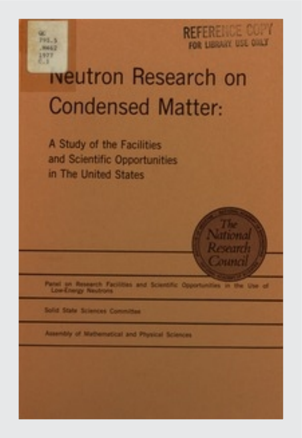

Neutron Research on Condensed Matter: A Study of the Facilities and Scientific Opportunities in the United States

DETAILS

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Panel on Research Facilities and Scientific Opportunities in the Use of Low-Energy Neutrons; Solid State Sciences Committee; Assembly of Mathematical and Physical Sciences; National Research Council

Neutron Research on Condensed Matter:
A Study of the Facilities and Scientific Opportunities in the United States

Panel on Research Facilities and Scientific Opportunities
in the Use of Low-Energy Neutrons
Solid State Sciences Committee
Assembly of Mathematical and Physical Sciences
National Research Council

National Academy of Sciences
Washington, D.C. 1977

NOTICE: The project that is the subject of this report was approved by the Governing Board of the National Research Council, whose members are drawn from the Councils of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine. The members of the Committee responsible for the report were chosen for their special competencies and with regard for appropriate balance.

This report has been reviewed by a group other than the authors according to procedures approved by a Report Review Committee consisting of members of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine.

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PREFACE

Over the last several decades, low-energy neutrons have been recognized as offering unique opportunities for research that could probe the nature of condensed matter, matter that varies greatly in atomic and molecular composition and structure arrangement. This probing often reveals with great precision physical interactions that account for observed properties of matter under study. Use of this powerful research tool in investigations of the multitude of structural problems with which the scientific community deals has surely been limited because of the size and complexity of nuclear reactors and their necessary location at large national centers or at a relatively few university centers. The sharing of such facilities with external users within the scientific community and the purposeful use of slow neutrons as a tool by a wide variety of scientists present a complex situation solvable only by a broad and perceptive consideration of numerous factors, some scientific and some managerial. Only then can the important impacts foreseen by the creative specialists in neutron research come to fruition. Moreover, in the last few years, a new type of neutron source, the pulsed-spallation source, has been conceived, which stands to offer dramatic improvement in neutron intensity and to open whole new areas of research opportunity.

Such considerations as these led, in early 1977, to the establishment of the Panel on Research Facilities and Scientific Opportunities in the Use of Low-Energy Neutrons by the Solid State Sciences Committee of the Assembly of Mathematical and Physical Sciences, National Research Council. The impetus for this study came from the desire of the two principal research supporting federal agencies, the Energy Research and Development Administration (ERDA), now part of the Department of Energy (DOE), and the National Science Foundation (NSF), to formulate guidelines for future activity in this area.

The general charge conveyed to the Panel at that time was designed to include (a) an assessment of the scientific and technological opportunities in the use of low-energy neutrons as a tool for research on the properties and structure of matter; (b) an evaluation of existing experimental facilities and the need for new, advanced, facilities for pursuing research in this area; and (c) a survey of the scientific community with present or potential interest in this area and the policies and procedures by which this community gains access to the necessarily centralized neutron facilities. Because it was recognized that the fruits of low-energy neutron research investigation flow to many disciplines, the Panel was to have representation, in either its core or subpanel groups, from the fields of condensed-matter physics, chemistry, geology, biology, and polymer science. Moreover, this broad-based representation was to be drawn with appropriate balance from research specialists and generalists from university, national laboratory, and industrial sectors such that a broad view of the future role and results of low-energy neutron science could be provided.

At an early meeting of the core Panel, it was decided to form subpanel groups for concentrated study in areas of (1) excitations and fluctuations in condensed matter, (2) biology and polymers, (3) interatomic structure, (4) radiation damage, (5) facilities and techniques, and (6) users and education. All of these groups have been very active in their deliberations in the short time interval since their formation, and individually they have supplied position reports that have become the major portion of the Panel report. From these subpanel reports and from its own overview activities, the core Panel has arrived at certain conclusions concerning the general health and vitality of this area of the U.S. science and what measures are called for in meeting future challenges. These conclusions are crystallized in the form of summary recommendations given in Chapter 2.

To foresee future needs for specific action successfully the members of the Panel have sought to assess to the extent possible the most important applications of present low-energy neutron science and technology and the more important impacts that might result. The scientists involved in various ways with the Panel have contributed a wealth of significant observations, conclusions, and recommendations that can only be adequately appreciated by the reader's careful consideration of the report as a whole.

My selection as Chairman of the Panel resulted from the desire of the Solid State Sciences Committee that the Chairmanship be in neutral hands. While I have remained neutral, I hope that the reader of this report will sense within it an enthusiasm for the future potential of low-energy neutron research as a tool for enlarging our knowledge of the properties and structure of matter in important fields--both scientifically basic and technologically practical.

I wish to acknowledge my personal indebtedness to Robb M. Thomson, Secretary of the Solid State Sciences Committee, and call attention to the key role he played in the directions taken by the Panel's activities. He was tireless in suggesting positive strategies for solving Panel problems as they arose, in contacting individuals who could furnish unique information of special value in various areas, and in contributing from his own wealth of knowledge.

Finally, I would like to express a healthy respect and high regard for the talents and opinions of my associates on the core Panel and the subpanels and to express my appreciation to members of the Panel Steering Committee, composed primarily of subpanel chairmen, who assisted in the operations of the study. Many scientists having no direct connection with this field of research have joined in this study, and we are particularly indebted to them for contributing to the breadth of the scientific considerations. I want to thank all the participants in this study for the dedicated and hard work that they have done in collaboratively preparing the report. The report benefited from the thoughtful comments of the reviewers. We hope that our endeavors will throw enough added light on the problems and potentials of several types of low-energy neutron research that the report will be of material assistance in the solution of some major national needs.

Gordon K. Teal, *Chairman*
Panel on Research Facilities
and Scientific Opportunities
in the Use of Low-Energy
Neutrons

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1

INTRODUCTION AND OVERVIEW

1.1. INTRODUCTION

This Panel has reviewed in depth the present status and future potential of the applications of low-energy neutron scattering to research in the condensed-matter sciences, including physics, chemistry, biology, and metallurgy. The study has shown that neutron-scattering technology has proven to be of enormous importance to research in the above areas and especially to those of solid-state physics and chemistry. Further, there is every reason to believe that this will continue to be the case for the indefinite future. Our main attention in this report will be focused on the scattering of low-energy neutrons by condensed matter; in addition, however, the same type of neutron source facilities can be used for the study of radiation damage. Hence this related topic has also been included in our study and discussed in our report.

Neutron research in condensed-matter science is an exemplary illustration of the catalytic interaction between different areas of science. In this case, a major discovery in nuclear physics--the production of neutrons from nuclear reactions--resulted in the development of nuclear reactors for studies in that field. These facilities in turn, have catalyzed interdisciplinary research in a large number of other fields of science. These have separately evolved into important research efforts with broad implications for their own respective subdisciplines. For example, studies of radiation damage of solid materials by neutron irradiation have accompanied fission reactor development over the past 35 years. Basic scientific studies of neutron irradiation effects have contributed to the understanding of technologically important forms of damage and to the general understanding of defect solid-state physics as well. This information in turn is essential for the development of successful energy sources based on nuclear reactions of both fission and fusion varieties.

In introducing neutron studies of condensed matter, it is useful to describe qualitatively the type of information obtained about condensed matter in neutron-scattering experiments. This will, of course, be discussed in much greater detail in later chapters of this report. From neutron-scattering experiments one is able to obtain detailed information on a microscopic scale of the positions of the atoms in a material and the manner in which these atoms move as a result of thermal excitation. Similarly, from the magnetic scattering, one obtains information on an atomic scale of the location of spins in a magnetic material, the relative alignment of the spins, and their thermal motion. Neutrons thus provide basic information about the structural and magnetic properties of condensed matter that is not accessible via any other experimental technique. Such information is essential for the understanding of all properties of materials.

The United States has occupied a leading position in neutron spectroscopy, and this preeminence in turn has contributed greatly to American leadership in condensed-matter science and technology as a whole. In many cases, completely new areas of research investigation have been opened up by the exploitation of the unique properties of neutrons. A salient example is the field of magnetism; most of our experimental information about the origin of magnetism on an atomic scale and the cooperative behavior of large numbers of magnetic ions derives from neutron experiments. Magnetic materials in turn have a vast range of technological applications ranging from transformers to computer memories. Neutrons have also provided a new and remarkably deep understanding of the dynamical interactions between atoms; this, for example, has led to a detailed microscopic understanding of the mechanisms for certain phase transitions in solids. A third, increasingly important, application of neutron scattering is in studies of the positions and dynamical behavior of hydrogen in materials. Hydrogen is, of course, a ubiquitous constituent of matter, including water, polymers, and all biologically active molecules.

Because of their unique properties, low-energy neutrons have been widely exploited by many scientists throughout the world wherever neutron sources of sufficient strength have been available. The principal thrust has been in solid-state physics and solid-state chemistry. Initial research efforts have concentrated on simpler types of materials in order to provide a basic theoretical understanding. In this respect, the technique has reached a high degree of maturity and sophistication. It is now possible to obtain a level of understanding in important but more complicated materials that would have been considered totally intractable only one decade ago. We are thus at an exciting junction point in neutron spectroscopy, where the field has evolved from studies of the basic properties of simple "model systems" to research into the microscopic static and dynamic properties of more complex and novel materials. Examples of the latter include metallic glasses, one- and two-dimensional magnets and conductors, fast ion conductors, hydrogen-containing metals, and adsorbed molecules and atoms on surfaces. It is also quite encouraging that many of these systems are of immediate technological importance. In the near term, we anticipate a major expansion with concomitant major accomplishments in the fields of molecular biology and polymers. These results will surely have a major impact in medicine and in the life sciences as a whole. Neutron-scattering studies in these fields are still at their earliest stages, but the initial results enable one to predict confidently that this will evolve into a major effort.

It is anticipated that neutron crystallography and neutron dynamics studies will become increasingly important in the structural analysis of materials at extreme temperature and pressure--an area of basic significance and enormous technological importance. Metallurgy is a field in which neutron scattering has so far seen limited applications, but the Panel strongly believes that there are future applications in a number of areas where this technique should lead to significant new developments.

As we have noted previously, observations of the alteration of many properties of solid materials by neutron irradiation have accompanied fission reactor development over the past 35 years. Basic scientific studies of neutron radiation effects have contributed to the understanding of technologically important forms of damage and to the general understanding of defect solid-state physics as well. Insofar as radiation defects are concerned with the properties of low concentrations of simple defects, such as isolated vacancies in interstitials, understanding is fairly complete, but many challenging problems remain concerning defect clustering and the behavior of materials in regions of very high defect density, especially at high temperatures. The economic and societal impact of improved scientific understanding of these problems cannot be overestimated. The Panel believes that continued and extended research in the area of neutron radiation damage can lead to acceptable solutions to many of the technological problems associated with nuclear power sources, as well as clarification of the physical principles underlying defects in the solid state.

In assessing the impact and future needs for neutron scattering, the Panel has been especially mindful of the complementary role and importance of alternative experimental techniques, including light scattering, infrared spectroscopy, electron scattering, and x-ray diffraction. Indeed, the Panel and all the subpanels included a number of scientists who are leaders in these other fields. It was our opinion that where possible photon or electron probes should be used in preference to neutron scattering, since those techniques generally involve facilities that are significantly less costly than neutron sources. In this report, therefore, we have emphasized those areas of condensed-matter research in which neutrons provide unique information not available by other techniques.

1.2. HIGH-INTENSITY NEUTRON SOURCES

A characteristic property of thermal neutrons, which is at the heart of their usefulness as probes of condensed matter, is their weak interaction with target material relative to other forms of radiation. This in turn means that high-intensity fluxes are required in order to carry out definitive experiments. Hence neutron-scattering research requires large-scale installations, where extensive radiation shielding can be installed, where appropriate measures can be taken to handle radioactive materials, and where the large amounts of heat generated can be readily dissipated. Currently, the principal sources of neutrons are steady-state nuclear reactors, which produce a *continuous* flux of thermal neutrons. In the United States, the two leading facilities are the High Flux Beam Reactor (40-60 megawatts) at Brookhaven National Laboratory and the High Flux Isotope Reactor (100 megawatts) at Oak Ridge National Laboratory.

Recently, very promising alternative techniques have been developed that will produce a very-high-flux *pulsed* beam of thermal and epithermal neutrons (below and above 0.15 eV, respectively). In essence, these techniques involve the injection of high-energy protons from a particle accelerator into target nuclei from which neutrons are dislodged. Since the neutrons in such devices are delivered in short pulses, the *peak* flux can be made much higher than that available in the highest flux reactors. Steady-state reactors, on the other hand, produce higher *average* flux. Because of the quite different characteristics of the two types of source, it turns out that the research problems best suited to the separate techniques are most often quite distinct. The Panel has given detailed consideration to this distinction as formulated in later chapters of the report, and *it foresees that exciting and important advances will occur with each type of facility.*

Continued operation and expansion of research facilities at the present highest flux reactors together with the development of a high-flux pulsed source are essential for the continued development of this field of science in the United States.* Detailed consideration has also been given to the development of a next-generation higher-flux steady-state reactor. Such a source would represent an outstanding facility for neutron research. We have concluded, however, that technological advances would be required to make such a source possible, and one of our recommendations deals with this.

The class of frontier accomplishments that we envisage demands highly sophisticated state-of-the-art instrumentation. We consider first instrumentation needs at the steady-state sources. Particular need exists for the development of ultra-high-energy resolution neutron spectrometers. These would provide improved precision in studies of such processes as diffusion in solids and of the phase transition behavior in many materials including those of technological importance such as ferroelectrics and superconductors. The advent of large-area detectors

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*It should be mentioned that these views parallel those expressed in the recent report, *Future of Nuclear Science*, prepared by the ad hoc Panel on the Future of Nuclear Science, Committee on Nuclear Science, Assembly of Mathematical and Physical Sciences, National Research Council (National Academy of Sciences, Washington, D.C., 1977).

would open up the systematic use of neutrons for protein crystallography and for studies of ordering in other complex systems. Similarly, well-instrumented small-angle scattering facilities would permit investigations of polymer conformation, inhomogeneities in colloids, and defect aggregate structures in metals and alloys. The development of high-efficiency neutron polarizing crystals would permit greatly extended investigations in magnetism, and indeed they would open up completely new areas of investigation.

Current designs for new-generation pulsed sources propose a peak thermal flux of 10^{16} neutrons cm^{-2} sec^{-1}, which is an order of magnitude larger than that of the highest flux steady-state reactors. This flux will provide important new opportunities for structural and dynamical studies of condensed matter by the use of time-of-flight techniques. The requisite use of time-of-flight techniques in turn means that the class of research problems best pursued at such a facility would, in many respects, be different from that most effectively pursued at the laboratories with steady-state facilities. Examples include the nonequilibrium properties of liquids, the general structural and dynamical behavior of amorphous materials, and studies of the dynamics of adsorbed species, especially those with immediate relevance to the heterogeneous catalysis problem. The pulsed sources also promise abundant neutrons in the epithermal range, that is, with energies greater than 0.15 eV. They will thence simply open up that area of neutron research, permitting new advances in studies of magnetism, hydrogen in metals, and the electronic structure of materials. The pulsed nature of the spallation source may also be exploited to study time-dependent phenomena such as defect migration in materials. All the above will require detailed instrumentation development in order to optimize the use of such a new facility. Many other potential forms of experimentation that would be possible with a pulsed source are detailed in later chapters of this report. *It should be emphasized that the most exciting and significant accomplishments with a new-generation neutron source may very well not be among those envisaged today.* This has certainly been the case in the history of our experience with the present high-flux reactor sources.

Improved facilities are also necessary for the continued development of radiation damage research. It is possible at present to produce technologically significant radiation effects only with prolonged exposure in high-flux steady-state reactors. With the present irradiation facilities at such sources, there are limitations in providing precise control of environmental parameters such as specimen temperature and stress as well as in being able to install proper instrumentation. These shortcomings along with the difficulty of retrieving irradiated samples for further investigation without altering their environment (temperature, vacuum, etc.) seriously hamper research in this area. Improvement in these facilities would make possible many types of experiments that cannot now be performed reliably, and these would in turn contribute directly to the successful achievement of national energy production goals. It is important that these shortcomings would be greatly alleviated for irradiations performed at a high-flux pulsed neutron source because of its special characteristics. Furthermore, a pulsed source would allow observations of the transient aspects of radiation damage that are not currently possible.

In the light of the scientific impacts highlighted in this chapter, and analyzed in detail in the main body of our report, the Panel finds that neutron science as covered in this report is an area of high scientific vitality and potential. It finds that the United States has held a forefront position in this field from its beginning and that a continuing national effort to maintain this leadership is justified.

2

SUMMARY OF FINDINGS AND RECOMMENDATIONS

Our major recommendations below fall into three categories, which deal, respectively, with our current facilities, the new generation source, and more effective use of all facilities. Within each category, the recommendations are listed in priority order, but we emphasize that *both our current high-flux steady-state reactors and the recommended new generation source each play an essential role in the field in the United States.* Within this context, it should be understood that steady-state and pulsed-source facilities are largely complementary as probes of condensed matter. We have not attempted, nor do we believe that it is possible, to rank order the separate classes of research amenable to the two techniques. Rather, as noted above, the Panel has concluded that both the steady-state and the newly proposed pulsed sources represent essential symbiotic tools for continued frontier research in physics, chemistry, biology, and metallurgy. *A national program combining both types of facilities would be singularly effective and would enable the United States to maintain its traditional leadership in low-energy neutron research and, more broadly, in condensed-matter science as a whole.* We have endeavored to confine our primary recommendations selectively to those that are considered to be truly necessary in meeting the future challenges and opportunities that lay open to this area of science. More detail on the specific form of our primary recommendations is given in later chapters of the report.

CURRENT FACILITIES

- Our existing highest-flux reactors at the Brookhaven and the Oak Ridge National Laboratories (HFBR and HFIR) are *national resources whose uninterrupted operation with full support for scientific programs and state-of-the-art instrumentation must be ensured to permit continued forefront research investigations.*

- There are many important neutron-scattering investigations that do not need the highest neutron flux; therefore, a balanced national program to meet the expanding applications of neutron research requires well-instrumented and well-staffed intermediate-flux facilities. In particular, *the Panel urges federal agencies to augment their program support for the best university-based neutron sources and recommends continued strong support for the versatile National Bureau of Standards (NBS) reactor.* The university facilities are extremely important not only for their scientific contributions but for the effective training of new scientists with expertise in neutron technology, while the NBS facility serves as an excellent center for neutron-scattering research by scientists from a number of government laboratories and universities.

- Because of its technological importance and its scientific significance, *the future development and vitality of neutron-produced radiation-damage research must be assured through the availability of properly instrumented neutron sources with adequate program support.*

NEW FACILITIES

- We foresee that major scientific achievements can be made in both present and unexplored areas with new-generation pulsed-spallation neutron sources with their very-high-peak neutron flux and readily tailored neutron spectra. The Panel thus *recommends that an immediate commitment be made for procuring* *such high-flux sources. Design studies should be made and funding support should be scheduled for the phased development stages leading to the creation of a national center with a high-flux (10^{16} thermal neutrons cm^{-2} sec^{-1} peak) pulsed-spallation neutron facility provided with the associated instrumentation required for effective use.*

- In order to prepare for the eventual replacement of the HFBR and HFIR reactors, it is *recommended that a study group for the design of future high-flux steady-state neutron sources be established.*

TOWARD AN EFFECTIVE SYSTEM

- In order to take optimum advantage of the scientific opportunities that are offered by our neutron-source facilities, *the Panel believes that the facilities at our neutron centers should be more widely used in the future by the national scientific and technical communities.* As discussed in the report, neutrons are a powerful research tool that can be (and in Europe are being) exploited over a broad range of scientific and technological areas. Detailed recommendations concerning aspects of broadened usage are given in Chapter 6.

- For reasons that are documented in Chapters 3 and 5, neutrons can only be used for the scientific study of matter in conjunction with instruments of a very high order of sophistication. *There is a strong current need to augment and modernize the instrumentation of the presently existing U.S. neutron facilities.* Since new developments in instrumentation are often equivalent to an increase in neutron flux at nominal cost, the development and deployment of new instruments is a very cost-effective action.

  Further, the new high-intensity pulsed source operates in a different mode than the traditional steady-state reactors and will require a different type of instrumentation (time of flight). *In order for the new-generation high-flux pulsed source to achieve its predicted potential, an instrument* *development phase will be required in parallel with the facility development.* Hence, the expensive investments in both types of national neutron source facilities must be supported by a corresponding program of instrumentation development at the centers.

3
LOW-ENERGY NEUTRON SCATTERING AND RADIATION-DAMAGE SCIENCE: GENERAL ISSUES AND CHALLENGES

3.1. BACKGROUND AND SCIENCE COMMUNITY ISSUES

3.1.1. Background

Developments in the science of low-energy neutrons have paralleled historically the development of neutron sources. Rudimentary information on low-energy neutron (LEN) scattering, and indeed on diffraction (cooperative, coherent scattering by many centers), was available in the late 1930's using neutrons generated with naturally occurring or accelerator-produced charged-particle radiation. It remained, however, for the wartime development of self-generating nuclear reactors to permit LEN scattering to flourish as a science in its own right. The much higher LEN intensity, of flux, that became available from such sources permitted controlled experimentation through use of collimation and monoenergetic neutron beams, and the early postwar years saw many fundamental developments that have formed the base of our present science.

It became clear that LEN radiation, with its unique characteristics, was to offer significant competition to the earlier-exploited sister radiations, x-ray and electron, as a tool in investigating atom assemblies constituting all forms of matter. Because neutrons are unchanged subnuclear particles, they can penetrate deeply into matter and can interact directly with the nuclear core of atoms over the full range of atomic size or mass. Unlike x rays and electrons, whose interaction with atoms originates in the charge structure of the atom, neutrons do not respond to atom charge. On the other hand, neutrons have a magnetic structure, and they do respond to atomic magnetization, which is sometimes produced by special configurations of the atomic charges or their spins. Of paramount significance is the fact that the momentum and kinetic energy of low-energy neutrons (those of energy of the order of 1 eV or smaller) are ideally matched to the equivalent quantities characteristic of excitations and fluctuations when atoms are collected together in the form of matter. In its uses, LEN radiation can be considered a "weakly interacting" probe in contrast to the "strongly interacting" electron and x-ray probes. These neutrons possess so little kinetic energy that they have minimal effect on what is being probed, whether it be a biological molecule involved in a life process or an organic substance undergoing catalytic reaction at a surface.

Because of these unique characteristics, the use of neutron radiation as a tool in investigating all forms of condensed matter has grown dramatically over the past two decades as new neutron sources have become available to experimentalists. Documentation of these many areas of application, which are still expanding, will be given in later chapters of this report. The form of experimentation behind these applications consists of projecting neutron radiation onto a specimen and searching for modification of its momentum, energy, polarization, and intensity. This affords information on the state of the specimen, namely, its most intimate structural detail (how atoms are arranged relative to each other) and how the atoms interact with each other, whether the specimen be of biological, chemical, or physical interest. This requires the existence of a radiation source of sufficient strength that these refined characteristics can be measured.

Before entering this discussion directly, however, it might be useful to supply some perspective information on the matter of "high flux" when applied to neutrons and why the matter of flux is so crucial in neutron experimentation. Very simply stated, in neutron-scattering experiments one is always faced with the problem of statistics in the limited intensity that is available to the detector at the end of the spectrometer instrument. This intensity with its statistical fluctuation is many orders of magnitude lower than the isotropic flux at the neutron source because of factors such as spatial collimation, incident energy selection, and final energy analysis. Long periods of data collection are often required in order to obtain meaningful intensity data.

The distinction between x rays and neutrons is a dramatic one when it is recognized that the quantal brightness of an x-ray tube source is one hundred thousand times larger than that of our present high-flux neutron sources. To this matter of brightness must be added the fact that neutrons are weakly interacting with matter (one of its advantages in many applications) so that coin-sized samples are generally required in order to get useful scattered intensity. In many cases, specimen crystals of such size are not available as in biological specimens, in surface films, or in esoteric materials. Moreover, there are frequently restrictions placed on sample size by the requirements of a special environment such as in high-pressure work. All of these factors have forced neutron experimentalists to become skilled in the art of optimizing a situation, often on the ragged edge of the possible, and many highly interesting scientific questions cannot yet be addressed at all.

3.1.2. Neutron-Source Facilities

It is significant that the United States held a virtual monopoly on the early developments in this field because of its unique position in having available, immediately after the war years, reactor neutron sources at the early Argonne and Oak Ridge National Laboratories. With the realization that nuclear reactors were important both in nuclear and solid-state science and in the technological experience of designing and operating such facilities with implications for future power generation, other reactor sources of advanced design have been procured in the intervening years at the national laboratories and at university centers. A tabulation of presently operating research reactors in the United States is given later in this report (see Table 5.1), and it includes a listing of 13 research reactors of operating power ranging from 100 MW to 1 MW with corresponding, roughly proportional, neutron-source flux. The commissioning dates of these reactor facilities extend back to 1951, and Figure 3.1 gives a histogram of the characteristic quantity, reactor-source power versus commissioning date. The activity that occurred in the mid-1960's was occasioned by the commissioning of the High-Flux-Beam Reactor (Brookhaven National Laboratory) and the High-Flux-Isotope Reactor (Oak Ridge National Laboratory). These two sources remain today as the dominant forces in this country's effort in this area of science.

Along with the various types of facilities operating at the national laboratories (Brookhaven, Oak Ridge, National

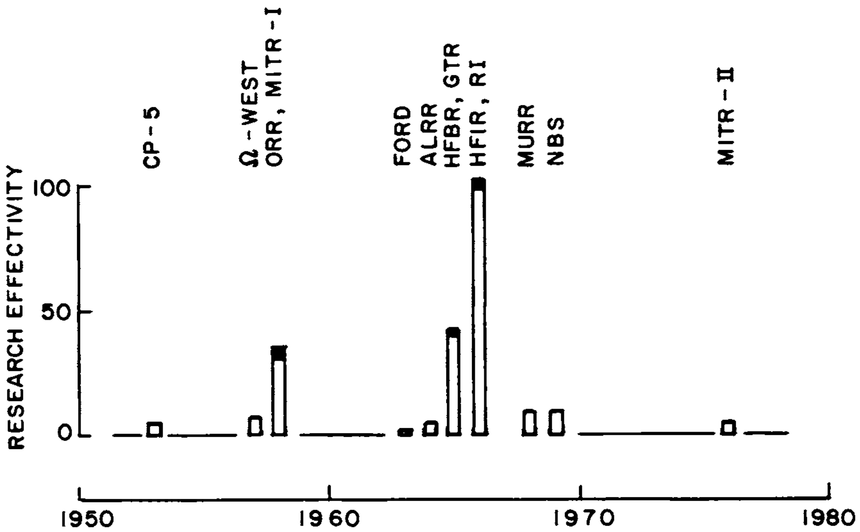

FIGURE 3.1 Commissioning of United States reactor neutron sources.

Bureau of Standards, Argonne, and Los Alamos), there are intermediate-strength facilities located at university centers (Missouri, Iowa State, Massachusetts Institute of Technology, Rhode Island, Michigan, and Georgia Institute of Technology). With the exception of the Iowa State (Ames Laboratory) reactor, the construction of these intermediate-strength facilities and their continuing operational costs have been financed mainly from university resources rather than by direct federal allocation. The justification of these expenditures to the universities has been that the facilities serve educational needs in the training of graduate students and other specialists and that they provide a home-based seat of research opportunity for independent, academic groups--a philosophy to which this Panel subscribes and which will be addressed again in this report.

Neutron scattering has always been an intensity-limited technique, and improvements in source intensity have invariably led to new accomplishments in neutron science. In attempting to overcome the practical limitation of neutron flux, which seems to characterize steady-state reactor sources, designers have concentrated on neutron sources of pulsed type in which bursts of high-energy charged particles are used through various nuclear reactions to generate high-energy neutron bursts. Upon neutron moderation to an easily controlled degree, LEN radiation bursts of adjustable energy range and of very high peak intensity are made available. *The most promising of these new-generation sources under consideration offer nearly an order-of-magnitude improvement in neutron source strength over that of any existing steady-state reactor source.* For most effective use of this high peak intensity, a new spectrometer technology is needed in which flight time is the distinguishing characteristic. Considerable experience in this has been gained with time-of-flight (TOF) facilities at reactors and also with pulsed sources in Europe and Japan and to a lesser degree in the United States. In this country, we have operated a few pulsed neutron-source prototypes, which, although improvised and with characteristics not particularly attractive to LEN experimentalists, have afforded valuable experience with this technique. It is fair to say that our effort in this direction has lagged behind that of other national communities.

The impetus behind having such a collection of neutron source facilities has come from the science community: scientists have appreciated the opportunities offered by LEN research and have responded enthusiastically in supporting the establishment of these facilities. The existence of such sources does not automatically guarantee that forefront research will result. It is clear that the availability of good sources should be closely tied to having modern, state-of-the-art instrumentation at these facilities and adequate research program support so that proper staffs, in number and expertise, will utilize the facilities to full effectiveness. *During the past decade, our facilities have been on such a scale and our staffs have been of such size and quality that this country stands preeminent in the world in the quality of LEN research.* However, a strong commitment on the part of the federal agencies to continue active support in this area of science is needed if the United States is to maintain this position.

3.1.3. Funding Support Issues

In addition to supplying radiation for LEN scattering and radiation-damage research purposes, the 13 currently operating research reactors satisfy many other needs such as those of the nuclear-physics, nuclear-engineering, medical-physics, isotope-production, chemical-analysis, and radiographic communities. The total annual operating expense for this collection of reactors amounts to \$14.7 million, and this is distributed among the various programs at the centers. Of this total figure, about 40 percent, or \$6.0 million per year, is associated with programs related to the concerns of this Panel. Funding of this annual neutron-source expense has come mostly (about 60 percent) from the Energy Research and Development Administration (ERDA) through sponsorship of its national laboratories. The remainder has come from the National Science Foundation (NSF), the Department of Commerce, the Department of Defense, and private institutions.

A survey of the research program expenses at the various reactor sites associated with LEN scattering and radiation damage yields a total of \$8.7 million per year. This includes items such as staff salaries and overhead, supplies, and routine backup expenditures maintaining the programs but does not include pro-rated neutron-source expenditures. *Thus the total cost of supporting the research programs in this area throughout the United States may be taken as \$15 million per year.* It has been estimated that equivalent expenditures outside the United States amount to about \$45 million per year, making a worldwide total expenditure of about \$60 million per year for research program support in this area.

None of the above expenditure figures includes the capital cost of neutron-source facilities, and indeed there has been very little facility expenditure in recent years in the United States following the activity in the mid to late 1960's, as shown in Figure 3.1. In the eight years since the commissioning of the NBS reactor, the only capital expenditures for new neutron sources in the United States have been those associated with the reconstruction of the MIT research reactor (\$2.0 million from university funds) and the installation of a pulsed neutron source on a satellite Los Alamos Meson Physics Facility (LAMPF) beam, also at a cost of about \$2 million.

This building activity appears insignificant when compared with the scale of that being pursued in other countries at this time. Table 3.1 gives a listing of ten major neutron-source facilities (either steady-state or pulsed variety) that are under construction today or approved for construction outside of the United States and that, in due course, will offer much expanded research capability and much higher source intensity than is now available there or here. The total capital expenditures on completion of these projects have been estimated, based on equivalent

TABLE 3.1 Worldwide Neutron Sources Approved or under Construction (1977)

IBR-II Pulsed Reactor, Dubna, U.S.S.R.
4-MW thermal average, 5×10^{16} neutrons/cm^2 sec peak flux, 1978 completion

Harwell Linac II Pulsed Neutron Source, Great Britain
2-10^{13} neutrons/cm^2 sec peak flux, 1978 operation

Saclay Research Reactor, France
14 MW, dedicated LEN facility, mid-1979 operation

Leningrad Research Reactor, U.S.S.R.
100 MW, 3×10^{15} neutrons/cm^2 sec flux, 1980 operation

Trombay Research--Isotope Production Reactor, India
100 MW, relatively low flux, 10^{14} neutrons/cm^2 sec

Swierk Research Reactor, Poland
30 MW

Tsukuba (KEK) Pulsed Neutron Spallation Source, Japan
10^{14} neutrons/cm^2 sec peak flux, 1980 completion

JAERI Research Reactor, Japan
30 MW, 1981 completion

KUR Research Reactor, Osaka, Japan
30 MW, 3×10^{14} neutrons/cm^2 sec flux, 1981 completion

Rutherford Pulsed Neutron Spallation Source, Great Britain
5×10^{15} neutrons/cm^2 sec peak flux, 1982 operation

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costs in the United States. Figure 3.2 shows the geographic distribution of this activity. Taken collectively, this flurry of current activity comes to the impressive total of $505 million. It must be emphasized that this current outside activity is not directed at merely matching the U.S. activity in the mid-1960's as represented in Figure 3.1. Rather to a large degree, it *represents a giant step forward in supplying facilities to the scientist of*

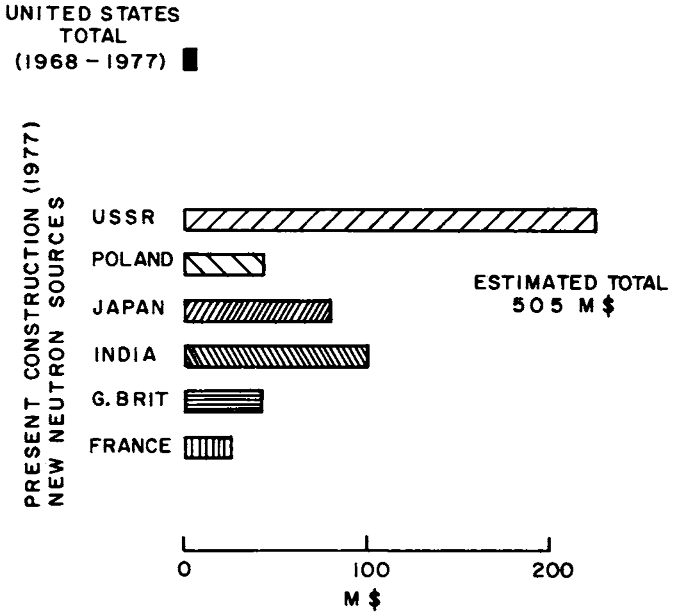


FIGURE 3.2 Estimated worldwide expenditures associated with new neutron sources (1977).

unprecedented source intensity and instrument sophistication. It signifies how a major, responsible segment of the worldwide scientific community views the future usefulness of new and more powerful neutron sources.

Of course, it is not preordained that our science effort should necessarily be competitive with, let alone be dominant over, other efforts. Responsive scientists in this country could very well conclude that the fruits flowing from a particular field of effort were diminishing in significance relative to national scientific and technological needs. During the course of its deliberations, it has *become clearly evident to this Panel (with representation from broadly separated fields) that such is not the case,* and that *the impact of this research discipline on many areas of science and technology can become far larger than it is now.* However, failure to provide neutron sources of advanced design will result in the surrendering of a valuable scientific asset and will relegate our effort to a minor position over the next decade.

Because of the unique scientific and technological opportunities that await us, *this Panel finds a commitment now by the federal research agencies toward maintaining a viable U.S. position in this area of science to be of paramount importance.*

The position of this country in this area of science is being further eroded at this time by planned or threatened withdrawal of neutron-source and associated experimental facilities. Thus, the ALRR reactor at Iowa State University with ERDA sponsorship is scheduled for shutdown at the end of 1977, the operation of the ORR reactor at Oak Ridge National Laboratory is to be used primarily by the Magnetic Fusion Energy program, and eventually within the next few years the aging CP-5 facility at Argonne National Laboratory will be withdrawn from service. Additionally, the intermediate- and lower-flux facilities at the university centers have all been operating in recent years under budgetary deficit conditions, with a continuing strain on university resources: *accordingly, their continued operation is seriously threatened.* Universities are perfectly willing to accept a reasonable share of costs attributable to their educational function, but research program funding is in the main beyond their resources. This Panel believes strongly that there does exist a real function of the educational centers with their introduction of new scientists into the field and their housing of home-based independent research groups.

The importance of having distributed independent research groups, functioning of necessity at less powerful and less costly university facilities than those at the major, key centers, should not be underemphasized. The major centers have tended to concentrate on exploiting particular areas of research--dynamics in the solid state at Brookhaven, magnetic structure and dynamics and crystallography at Oak Ridge, liquid dynamics at Argonne, hydrogen in metals and amorphous magnets at NBS. Outstanding contributions to our scientific and technological understanding of materials have evolved from this type of group attack, as documented in this report. On the other hand, the case can also be presented for the existence of smaller groups with the broad perspective that characterizes university research outlook, particularly when it is recognized that the scale of university research support in this field is an order of magnitude below that at the major centers. It is worth noting that developments such as dynamical diffraction, subtle neutron interactions, polarized beam technology, magnetization mapping, and neutron interferometry have all been generated in a university atmosphere.

Some of the implications on the neutron-science community of this planned or threatened reactor withdrawal are indicated in Figure 3.3, which illustrates the spectrometer unit and budgetary loss associated with these curtailments. This is a matter of serious concern since if all of these

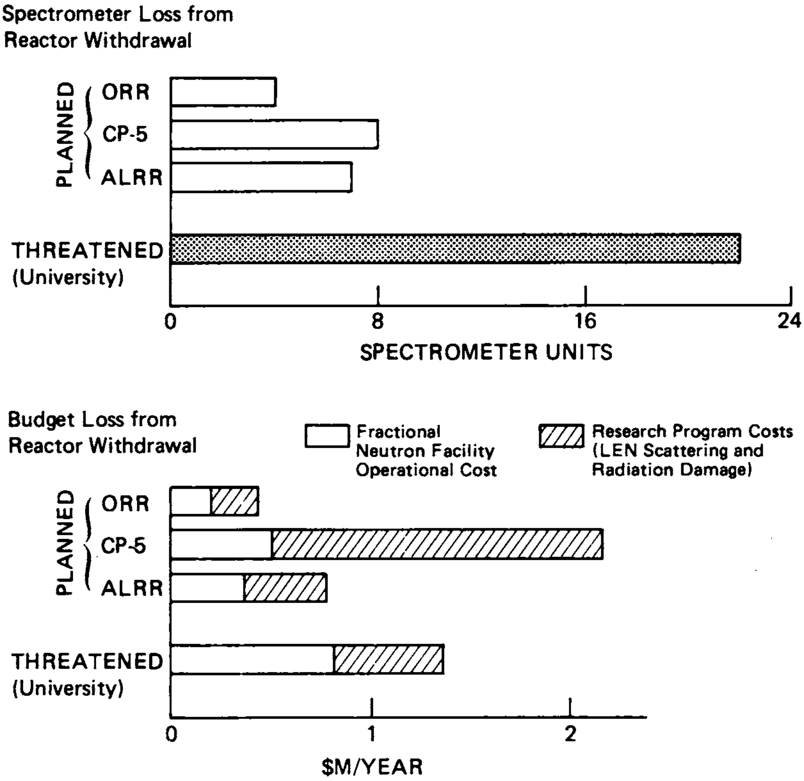


FIGURE 3.3 Effect on neutron-scattering (radiation-damage) community of planned or threatened reactor withdrawal in the United States.

curtailments were to materialize without other provisions being made to continue the research, it would mean a withdrawal of 32 percent of the research program support and a significant fraction of the spectrometer resources available to scientists in this field. The Panel believes that it is, therefore, essential to lessen the potential losses of scientific productivity that these threats present by the actions covered in our summary of recommendations.

3.1.4. Experimental Facilities and Users Topics

A survey of existing experimental facilities located at U.S. neutron sources indicates the presence of 73 spectrometer units and 5 radiation-damage units. The distribution of these spectrometer units installed at sources of different flux characteristics is given in a later chapter of this report (see Table 5.3). This distribution indicates that 20 spectrometer units are located at the two high-flux reactors (HFBR and HFIR) with the remainder and all the radiation-damage facilities to be found at lower-flux sources. It is significant that practically all of these facilities are of early design (having originated at the time of new reactor commissioning), and although operational improvements have been effected over the last decade, only a few can be classified as being state-of-the-art instruments. While conventional inelastic scattering and diffractometer instruments in the United States are generally competitive with the best in the world, it must also be recognized that our existing facilities do not include some of the highly sophisticated, special-function features that have evolved in recent years. *This problem of instrumentation development is an important one in the view of this Panel* and is addressed in our primary set of recommendations in Chapter 2. In particular, there is a great need for modern-design small-angle scattering and ultra-high-energy resolution spectrometers as discussed in Section 5.4.1. and at various places in Chapter 4.

In addition to these instrumentation improvements at our reactor sources, it is of equal importance that a vigorous program of instrumentation development be pursued that will permit the most effective exploitation of our present and planned pulsed neutron sources. As mentioned earlier, pulsed-neutron sources call for the use of time-of-flight (TOF) spectrometer units, and experience with this type of instrumentation in the United States to date has been limited. At present, there are five TOF spectrometers operating at our reactor sources and one TOF spectrometer operating with the WNR pulsed-source facility at the Los Alamos Scientific Laboratory. Useful experience is being gained with these units, but it must be emphasized that their sophistication is far below that which is needed to match the performance of a high-flux pulsed-neutron source.

Included in the distribution of 73 spectrometer units in the United States are two other spectrometer groups of 46 two-axis units (useful for structure analysis) and 21 three-axis units (inelastic scattering). It is informative to compare this census of experimental facilities in the United States with that in existence elsewhere. The histogram of Figure 3.4 draws such comparison with spectrometer delineation among the three groups mentioned above. Our survey leads to a worldwide total of 345 spectrometer units, of which 73 are located in the United States, 119 in Western Europe, 78 in Eastern Europe, and 75 in other widespread countries. Considering the availability of spectrometer units as an indicator of scientific activity, it is seen that about 60 percent of the effort is expended in elastic, diffraction-type studies with two-axis units. However, a sizable 40 percent (and growing) effort is devoted to inelastic-scattering studies in which the peculiarly unique characteristics of LEN radiation are exploited. This distribution seems to be common in both the United States and elsewhere.

It is apparent from the display in Figure 3.4 that a pronounced gap exists in the exploitation of the flight-time technique between the United States and elsewhere. As mentioned above, this has been occasioned by the lack of pulsed neutron sources of suitable capability in the United States to make this technique attractive to experimentalists. An important feature of a pulsed neutron source of modern design is the very-high-peak neutron flux that can be generated, as high as an order of magnitude larger than that of any existing steady-state reactor. To attain such peak intensity with a steady-state reactor would require very large expenditures of effort and monies even if it were technologically promising. Much of the attention of this Panel and the present report has been devoted to an assessment of the potential usefulness of such high neutron intensity, and one of our primary recommendations is devoted to this topic.

Scientists using these existing spectrometer facilities can be divided into two general classes, full-time-equivalent

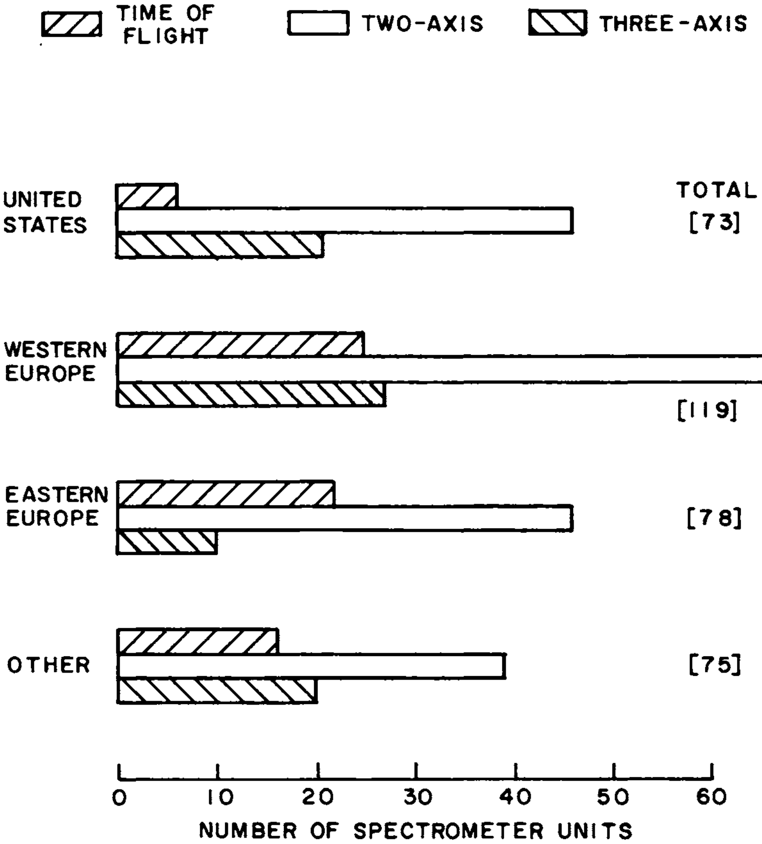


FIGURE 3.4 Worldwide neutron spectrometer distribution.

(FTE) users and part-time users. The former group includes full-time staff members at the national laboratory centers and those academic scientists at the university neutron centers whose research activity is dedicated to LEN-type research. The latter part-time users group includes those scientists who have interest in obtaining neutron-scattering information as a part of some more general research program. A survey of the users' community in evidence during the past year indicated 102 FTE users and 284 part-time users, of which the numbers 70 and 255, respectively, indicate approximately the user activity at the national laboratories, including the National Bureau of Standards. The research productivity of this group of scientists is demonstrated in the publication rate statistics, which show numbers of 204, 217, and 313 per year over the three-year period 1974-1976. Since the FTE staff size at the centers has been essentially unchanged during this period, this growth in productivity is presumably associated with the growth in the number of part-time users reported by the neutron centers. This growth trend in the number of outside users of the neutron facilities is encouraging. Since the usefulness of LEN techniques in providing information to both fundamental and technological developments has been amply demonstrated, the Panel believes that an enlarged user group can deliver increased benefits to an expanded segment of the national scientific community.

3.2. PERSPECTIVES AND CHALLENGES OF LOW-ENERGY NEUTRON SCIENCE

In previous pages we have reviewed the developments that have led to the present status of the low-energy neutron facilities and the associated scientific community in the United States, and we have placed this in the context of the general world scene. It has been shown how the United States invested heavily in neutron sources and instrumentation in the mid-1960's, giving to the scientists in this country the very best facilities available at the time. It has also been shown that over the past decade there has been no further strengthening or expansion of these facilities, and in fact the science community presently faces the threat of curtailment of some of them. In this section we shall consider the scientific aspects of the picture, drawing heavily on details laid out in the next chapter. We shall first discuss the impact that these facilities and the scientists associated with them have had on the general scene of science in the United States. We shall then proceed to a discussion of the future opportunities and challenges in this field and how they can best be faced.

3.2.1. Scientific Accomplishments and Present Status

Because of the research activities permitted by the facility developments in the decade previous to this one, as shown in Figure 3.1, and most notably from the commissioning of the high-flux beam reactors at Brookhaven and Oak Ridge, the United States has come to occupy a preeminent position in the field of neutron spectroscopy. These accomplishments in neutron-scattering studies of condensed matter have, in turn, contributed greatly to the strength of American condensed-matter science and technology as a whole. The success of the U.S. neutron spectroscopy programs has rested on (a) the high-flux sources themselves, (b) the ability of the neutron centers to attract unusually creative and productive scientists, and (c) the availability of high-quality ancillary support facilities. The three of these are equally important components.

To date, LEN research has made important, and in many cases dominant, contributions to an impressive array of basic problems in the condensed-matter sciences and, to a more limited extent, the life sciences. In support of this contention, we note in passing that three awards over the years of the Oliver E. Buckley Solid State Physics Prize of the American Physical Society have been given to LEN scientists in recognition of signal contributions to solid-state science. We will discuss these varied contributions, past, present, and future, in detail in later chapters of this report. It is of interest nevertheless to highlight here some of the contributions and the challenges that loom ahead.

In essence, in a neutron-scattering experiment one determines the position of atoms and/or the direction of magnetic spins (or orbital currents) associated with unpaired electrons in matter. This may be done on three different time scales: instantaneously, between 5×10^{-13} and 10^{-9} sec, and averaged over long periods of time as in elastic scattering. Further, the neutron is a passive probe so that one effectively determines the properties of the material with minimal perturbation of it. Thus, using neutron spectroscopy one obtains basic static and dynamic structural and magnetic information about condensed matter. Such information is, and will continue to be for the indefinite future, an essential component of our explorations of condensed matter.

Even at its earliest stages, neutron scattering introduced qualitative changes in several areas of research. One of the most dramatic of these developments was in the area of magnetism. Pioneering experiments at Oak Ridge National Laboratory demonstrated the ability of neutrons to determine magnetic ordered structures in antiferromagnetic materials. This work opened up a completely new approach to studies of magnetic ordering in a vast range of materials and its implications to fundamental theory. A major accomplishment in this area was the elucidation of the complex magnetic structures of the heavy rare-earth metals carried out at Oak Ridge. Neutrons also proved centrally important in a vast range of crystallographic problems. We note in particular that, in contrast to x rays, neutron-diffraction intensities are sensitive to hydrogen atom positions. Thus neutrons have provided detailed information on the structure of hydrogenous materials, a classic example being the structure of ice. This, in turn, has contributed greatly to our understanding of the hydrogen bond.

Measurements of the very-low-energy excitation (phonon-roton) spectrum in superfluid helium by neutrons represents a cornerstone of modern low-temperature physics; much of the current work in liquid helium relies crucially on these data. Small-angle neutron-scattering (SANS) studies of clustering and giant moment distributions in metals has contributed greatly to the field of metallurgy and to magnetism in metals. Polarized-beam studies of magnetic form factors have yielded information about electronic wavefunctions in a variety of insulators and in metals; these, in turn, have had a large impact on the theory of bonding in solids and on quantum chemistry as a whole.

With the development of the higher-flux sources, detailed inelastic-scattering studies became possible and, in fact, almost routine. Initial efforts were concentrated on mapping out the elementary excitation spectra (phonons and magnons) in prototypical systems. The measured dispersion relations could in many cases be inverted to obtain the interatomic forces, neighbor by neighbor. Such data thence have been central in the development of microscopic theories of the structural and magnetic interactions in insulators and semiconductors and especially in metals. For example, most available microscopic information on the magnetic structure, excitations, and microscopic interactions in rare-earth metals and alloys originates from neutron-scattering studies.

The past few years have seen a dramatic increase in the diversity, depth, and general sophistication of neutron-scattering experiments. Perhaps the most significant recent application of neutron spectroscopy has been in the interdisciplinary area of phase transitions and critical phenomena. This includes the development and full exploration of the soft-mode concept in structural and magnetic-phase transitions, the elucidation of the role of spatial dimensionality, and detailed tests of static and dynamic scaling theories of critical phenomena in a variety of systems. Forefront work in this area has been concentrated at the Brookhaven National Laboratory.

In solid-state physics, neutrons have provided detailed microscopic information on such diverse problems as hydrogen diffusion in metals, single-particle and collective excitations in itinerant magnets, anharmonic multiphonon effects especially in quantum solids such as ^{4}He, crystal field effects in metals, and phason excitations in incommensurate solids. In many cases, this basic information is only accessible via the neutron technique. Important inroads have also been made in studies of the dynamics of disordered materials including both alloys and glasses.

It is probably fair to say that, with the exception of quantum liquids, the principal triumphs of neutron spectroscopy have been in the areas of solid-state physics and chemistry. However, in the past few years, encouraging developments have occurred in other areas including liquid-state physics, polymers, organic chemistry, and biology. For example, recent small-angle neutron-scattering experiments at Grenoble have substantiated the Flory theory of excluded volume effects in flexible polymer conformations, including semidilute solutions. In liquid-state physics, work at Argonne National Laboratory has provided a detailed mapping of the dynamical structure of liquid argon, rubidium, and rubidium bromide. Rather close agreement is found with molecular dynamics calculations using model potentials.

In general, the recent applications at Brookhaven National Laboratory of neutron-scattering techniques for problems in molecular biology have given clear evidence that neutrons are particularly useful for the elucidation of architectural details of biological structures like nerve membranes, viruses, enzymes, and protein complexes. Neutron crystallography has been used to characterize structural hydrogens in oxygen-storing proteins, where these atoms control the processes that regulate oxygen uptake and release according to physiological conditions. Small-angle neutron-scattering techniques have had a great impact in the understanding of the large biological complexes that translate the genetic code into proteins, the hardware of biological machinery. Neutron research is unique in the analysis of these noncrystalline gel-like complexes utilizing the scattering difference between deuterium and hydrogen atoms, thus imposing contrast on the molecular constituents in cases for which other techniques cannot. Small-angle neutron analysis of lamellar structures like those found in membranes has introduced a scientific rigor into membrane studies eliminating the controversial "model techniques" of small-angle x-ray scattering.

3.2.2. Scientific Challenges

Looking ahead to future developments in LEN science, we are at an exciting stage in the history of neutron spectroscopy. As outlined above, neutron scattering has been and, in the judgment of the Panel, will continue to be for the indefinite future an essential experimental tool for many subdisciplines of solid-state physics, chemistry, biology, polymers, and materials science and technology. The health of American research in these areas would be seriously impaired, in the opinion of the Panel, if our current reactor-based neutron programs, especially those at the high-flux sources, were not maintained.

In later chapters of this report, we shall enumerate many examples of research topics that have been either limited in execution or impossible to study with our present-day sources and instrumentation. These limitations are of serious concern to the Panel in view of what might be accomplished toward meeting general scientific objectives. Because of the importance attached to this aspect of future low-energy neutron research in the United States and its bearing on the Panel recommendations, we shall summarize in the following some noteworthy illustrations of the new scientific areas that we foresee as being opened with new instrumentation at the existing steady-state sources and with a new generation neutron source. The discussion will assume in addressing questions of flux limitation that the next generation source will be a high-flux pulsed source (HFPS) as described above in Section 3.1.2. and later with more detail in Section 5.5.

Biology and Medicine

Biologists are currently trying to understand living organisms in terms of the structure and interactions of biomolecules. Progress in this area will help to provide the basis for understanding disease mechanisms and suggest rational biochemical approaches. Neutrons can make a unique contribution in the study of three aspects of biological structure: (1) the atomic organization of macromolecules such as proteins, (2) the conformation of flexible molecules as they interact with the bioenvironment, and (3) the larger-scale organization of complexes formed from many macromolecules.

As mentioned in an earlier section, the use of neutrons in the study of biological problems has begun to make substantial contributions and may become an activity of major significance in the future. Areas of special promise are the studies of the assemblies responsible for the expression of genetic information (ribosomes, RNA polymerase), the structure of proteins (myoglobin, lysozyme), model systems that mimic cholesterol-ester-containing deposits in arteries, the structure of connective tissue (collagen), and the molecular organization of viruses and cell membranes.

Steady-state neutron sources are ideally suited for small-angle studies of biological structuring, such as found in assemblies of proteins and nucleic acids in functional complexes. In addition, these sources provide the high average neutron flux for efficient protein crystallography using large two-dimensional position sensitive counters. The pulsed nature and the high-peak flux of a spallation source will provide exciting new prospects in the examination of biochemical transient effects and may permit the characterization of biomolecular dynamics.

Polymers

Plastics and rubbers are the most rapidly developing commercial materials in present-day use. They owe their unique properties to their large molecular size and to the interaction of these molecules one with another. Neutron studies on these materials have only barely begun, and we believe they will ultimately be important to our understanding of the structure and atomic properties of polymers. Indeed, recent work with LEN scattering has already confirmed some of Flory's earlier theoretical predictions that led to the 1976 Nobel Prize in chemistry. Another possible application of major importance is the sensing of "labeled" atoms in polymeric chain-folded crystals for studies of the detailed molecular environment at that site, e.g., the cross-linking reaction that produces strong polymers. The labeling would be accomplished by substituting deuterium for hydrogen.

Energy-Related Materials

Studies of metal hydrides and of hydrogen dissolved in metals are important because hydrides have been proposed as a means for storing hydrogen in energy systems and because of the serious degradation that hydrogen causes in the mechanical strength of high-strength steels and other metals. Neutron diffraction and scattering studies are the methods of choice in examining the relation between the structure and properties of these materials. Solid electrolytes are materials that are ingredients of new high-performance batteries for mobile vehicle and stationary energy storage systems. They are good electrical conductors with the conduction occurring by ionic diffusion. There are many new solid electrolytes that have been discovered recently, few of which have been examined structurally. Again, neutron diffraction is the method of choice, since these materials contain both heavy and light atoms. Also, the mechanisms of conduction can be examined by neutron scattering.

Structure and Dynamics of Amorphous Materials and Liquids

The general structure and excitations of matter in the liquid phase and noncrystalline solids remains one of the forefront areas of condensed-matter science. Neutrons will continue to play a crucial role in this field. Measurements of scattering at large scattering "angle" and higher resolution and energy will be easier with HFPS, both for determination of static or average structure and for excitations. The HFPS will make possible opening up a completely inaccessible range of momentum transfer which will probe the crucial range between the crystal-like and fluid-like motion of the atoms in a liquid. Opening up the area of fluid and amorphous materials structure is likely to be one of the major achievements of HFPS. The steady-state sources will also supply important experimental results on these materials. Studies of low-energy excitations and dispersion relations are best carried out with steady-state instruments. Also, long-wavelength, low-energy excitations, currently inaccessible, could be studied at reactors equipped with small-angle scattering and high-energy-resolution instruments.

Surfaces

Although there are many techniques for studying adsorbed species on surfaces, neutrons are unique because of their sensitivity to light atoms, hydrogen in particular, and because of their ability to study atomic motions. In this area, the highest possible flux, thermal and epithermal, is clearly needed, since the volume associated with surface coverage is necessarily exceedingly small. Measurements of the structure and collective excitations of the adsorbed layers can be made at the high-flux reactors, while high-flux pulsed sources will open up the detailed study of the diffusional and vibrational behavior of individual adsorbed molecules. Major contributions to surface science and catalysis can be expected from surface-layer studies.

Complex Materials

The development of large two-dimensional counters coupled to either steady-state or pulsed sources will allow structural analysis of the most complex chemical structures. The HFPS also will enhance the analysis of complex vibrational spectra, particularly in the higher-energy region. The high-peak flux of pulsed sources will be particularly useful in extending the range and resolution of the new powder profile analysis technique for polycrystalline samples. Thus, detailed structural information would become available for many materials that cannot be prepared in single-crystal form.

Defects, Impurities, and Chemical Reactions in Materials

The high-flux and resolution capabilities of HFPS together with new instrumentation at steady-state reactors will allow investigations of the properties of isolated defects and impurities. Although other techniques are available for these types of studies, neutrons serve as a unique probe because of their sensitivity to hydrogen, atomic magnetic structure, and atomic dynamics. High-flux steady-state reactors equipped with small-angle scattering facilities can be used to study defect aggregate structures, while triple-axis spectrometers at these sources can be used to study the interaction of impurities with specific lattice modes. The high-flux and resolution capabilities of HFPS will allow determination of the static distortion of the lattice surrounding isolated defects and the specific site of the defect. Neutrons from HFPS can also be applied to research on solid-state chemical reactions and to high-resolution studies of the details of the diffusive processes in solids.

Unique Experimentation with HFPS

The inherent physical and experimental arrangement of the HFPS provides obvious advantages for certain types of experiments. The fact that the neutrons are produced in short bursts makes possible the measurement of transient effects on the structure and dynamics of material of many kinds, e.g., chemical, thermal, and pressure relaxation effects and magnetic and electrical transients. Also solid-phase changes and transformations could be followed in time. Extremely high magnetic fields can be obtained by pulsing, and it is natural to combine this with neutron bursts from a pulsed source. The same can be said of pressure pulsing with geological applications. Thus, additional experiments on materials in extreme environments, an area of importance to technological applications as well as to science, are made possible by HFPS.

Magnetism

Neutron scattering plays a central role in the study of the magnetic properties of matter and their correlation with other physical properties. The study of magnetic impurities and systems with very small and subtle magnetization would be greatly extended by the provision of efficient neutron polarizers in instrumentation at both steady-state reactors and the HFPS. Improvement factors as large as an order of magnitude may very well become possible. Thus studies of organic free-radical molecules with unbalanced electron spins or of diamagnetic currents in both organic and inorganic systems may be susceptible to study with fundamental implications on chemical bonding. The HFPS would permit the study of high-energy, short-wavelength excitations in insulators and metals and studies of magnetic colloids, ferroliquids, and amorphous magnets. The study of these materials, along with the establishment of fine detail near magnetic phase transitions, would also be greatly facilitated by the use of area detectors and polarizers at steady-state sources.

Radiation-Damage Studies

In radiation damage, the principal current challenge is to generate a greater understanding of the complex interactions between defects that occur in the agglomerates or cascades created in neutron irradiation. This should also include the relation between these cascades and their ultimate effect on the macroscopic mechanical properties of the material, a matter of great technological importance in the design of advanced fission and fusion power sources. The only effective way to gain this understanding will be to conduct a variety of well-conceived experiments in which other tools and measurements are combined with the neutron irradiation, in some cases *in situ* and in others by transfer of the sample under controlled conditions to other laboratory locations. In these experiments, it is also a great advantage to be able to work under conditions where the gamma-ray flux is low so that the ambient temperature of the sample can be controlled. All of these conditions can be faced more easily with the pulsed-spallation source configuration because of its convenient accessibility and adaptability.

Overall, the scientific opportunities that are available to the LEN scientific community are impressive and challenging. They emphasize to this Panel that the future contributions of LEN science to widely varied research disciplines can be even more pronounced than they have been in the past. *Other significant developments, not foreseen at this time, may very well surface in the future as has certainly been the case since our last generation facilities were planned.* In order to translate these opportunities into real benefits, it is clear that strengthened and new facilities will be needed. In the view of this Panel, the scientific benefits that will accrue justify the investments that are required. This belief is embodied in our two major recommendations as discussed and summarized in Chapters 1 and 2. Only in this way can low-energy neutron science in the United States be fully exploited and a strong U.S. position be maintained during the next decade.

The evaluation of specific proposals for new facility construction or instrumentation development has not been within the scope of this study. Nevertheless, it is appropriate to consider briefly the general magnitude of the costs associated with the developments that are suggested in the Panel's recommendations. In Table 5.5

the cost estimate for constructing a high-flux pulsed source of the type envisaged here is given as $62 million. Its operating costs would be comparable with those of one of the present high-flux steady-state reactors, i.e., in the neighborhood of $5 million per year. The costs of upgrading the instrumentation at the present reactors and for instituting programs of instrument development have not been estimated in detail. An initial expenditure for instrument development on the order of $3 million and an added annual expenditure of approximately half that amount are estimates that would meet the goals set out in this area for the present reactor centers.

4

LOW-ENERGY NEUTRON SCATTERING AND RADIATION-DAMAGE SCIENCE: SCIENTIFIC ISSUES

4.1. NEUTRON RADIATION AND ITS FUNCTIONAL USE

The essential features of neutron scattering can be understood by noting a number of simple basic facts. (1) The neutron is a neutral particle and therefore can typically penetrate very deeply into solids. (2) The neutron couples directly to the nuclei of the target via purely nuclear forces; the nuclear scattering at thermal energies is isotropic in space with a typical cross section of 10 barns. The neutron-scattering length for a nucleus has no simple relationship to Z, the atomic number. (3) The neutron has a magnetic moment that couples to the magnetic moment of unpaired electrons in the target; this cross section is also typically 10 barns. Further, the neutron may be polarized so that polarization information may be obtained. (4) At thermal energies the wavelength of a neutron is comparable with the separation of atoms in condensed matter. Thus thermal neutrons display pronounced interference effects when scattered from condensed atomic systems; the interference patterns in turn contain detailed information about the spatial distribution of atoms and spins in the target. (5) In general, the neutron energy is in the thermal range (~500 K). Hence neutrons provide a very sensitive means of studying the thermal energy spectra of the structural and magnetic elementary excitations and the dynamics of the target system on an atomic scale.

The general theory of neutron scattering is rather complicated. For illustrative purposes we consider the simplest nontrivial scattering process, coherent nuclear Bragg scattering. In this case,

$$\frac{d\sigma}{d\Omega} = \left| \sum_{l} b_l e^{i\vec{Q} \times \vec{R}_l} \right|^2,$$

where b_ℓ is the scattering length of an atom at position $\vec{R}_\ell$ and $\vec{Q}$ is the momentum transfer. For a crystalline solid of infinite extent this becomes

$$\frac{d\sigma}{d\Omega} = N \frac{(2\pi)^3}{v_0} \sum_{\vec{\tau}} \delta(\vec{Q} - \vec{\tau}) \; |F_N(\vec{\tau})|^2,$$

where the structure factor

$$F_N(\vec{\tau}) = \sum_d b_d \exp(i\vec{\tau} \times \vec{d}).$$

Here $\vec{\tau}$ is a reciprocal lattice vector of the structure and $\vec{d}$ defines the positions of atoms within the unit cell. Thus, in a standard crystallographic experiment one measures a series of Bragg peaks at positions $\vec{\tau}$ with intensities that depend on the position of atoms within the unit cell. Hence in most cases one determines the unit cell and the space group from the observed τ's, and one deduces the positions of the atoms from the intensities. The atomic species, in turn, is identified by its scattering length b_d. An exactly analogous expression obtains for magnetic Bragg scattering. Using polarized-beam techniques one may also observe interference effects between nuclear and magnetic scattering. This technique is, in fact, of considerable practical importance.

For disordered systems, either liquids, glasses, or alloys, the elastic or quasielastic scattering is observed at all Q's and not just at special positions in reciprocal space. In that case, it is necessary to use correlation function language. Similarly, inelastic scattering occurs everywhere in Q space so that again a correlation function approach is most appropriate. This is discussed explicitly in Section 4.3.

There is, of course, a vast range of experimental techniques used in neutron scattering, diffraction, and spectroscopy. Essentially, the neutron *direction* is determined by collimators, whereas the neutron *energy*, and hence its vector momentum $\hbar\vec{Q}$ is determined by measuring either the neutron wavelength, $\hbar Q = 2\pi\lambda^{-1}$, or its energy, $\hbar Q = \sqrt{2mE}$. For inelastic scattering studies there are thence two basic approaches: (a) triple-axis spectrometry, which uses crystal reflections for energy analysis and Soller collimators for direction, or (b) time-of-flight (TOF) spectroscopy in which, in essence, one measures the time of arrival and hence the energy of the neutrons at a fixed counter position. These techniques may both be used in crystallographic applications in a straightforward fashion.

Triple-axis techniques are uniquely suited for use with a steady-state source. TOF techniques, on the other hand, may be used either with a reactor or with a pulsed source such as an accelerator-based spallation system. The time-averaged flux of a pulsed source is much smaller than that of a reactor. However, since it is only the flux within the time frame of the neutron burst that is generally relevant in TOF spectroscopy, the *usable* flux in a high-intensity pulsed-source system may be appreciably higher than that from existing reactors. For example, the proposed Argonne facility, IPNS II, promises an order-of-magnitude improvement in thermal flux over current U.S. high-flux reactors for TOF spectroscopy. Further, there will be orders-of-magnitude improvement in the epithermal range above 0.15 eV. As documented in the following subsections, this increased flux may be centrally important in many applications.

For illustrative purposes we discuss in more detail the experimental arrangement for triple-axis spectrometry. We show in Figure 4.1 a schematic diagram of a triple-axis spectrometer. Thermal neutrons from the reactor pass through a horizontal collimator C_1 and are incident on a crystal G_1, which is typically well-oriented pyrolytic graphite for low energies or single-crystal beryllium for higher energies. Neutrons that satisfy the Bragg condition within some bandwidth $\Delta\lambda$ pass through collimator C_2 and are incident on the sample. This first part of the spectrometer thence determines the neutron energy, via its wavelength, and its momentum. The sample may sit in a cryostat, an oven, a high-pressure press, or a superconducting magnet. Because many materials are almost transparent to neutrons, studies of matter in a controlled environment are generally quite straightforward. The second part of the spectrometer determines the outgoing energy and momentum. Consequently, in this simple fashion one determines the energy and momentum transferred to the sample in a particular scattering event.

As we noted above, since the neutron is a passive probe, the scattering cross section is simply related to the fundamental properties--specifically, the two-particle space-time correlation function--of the material being studied. In a typical scan, one chooses the spectrometer angles so that the momentum transfer $\vec{Q}$ is fixed, and one then scans the energy. By contrast, in a TOF experiment the energy and Q both vary across a scan. As we discuss in detail in Section 4.3, the triple-axis technique is well suited for problems in which the response is localized in

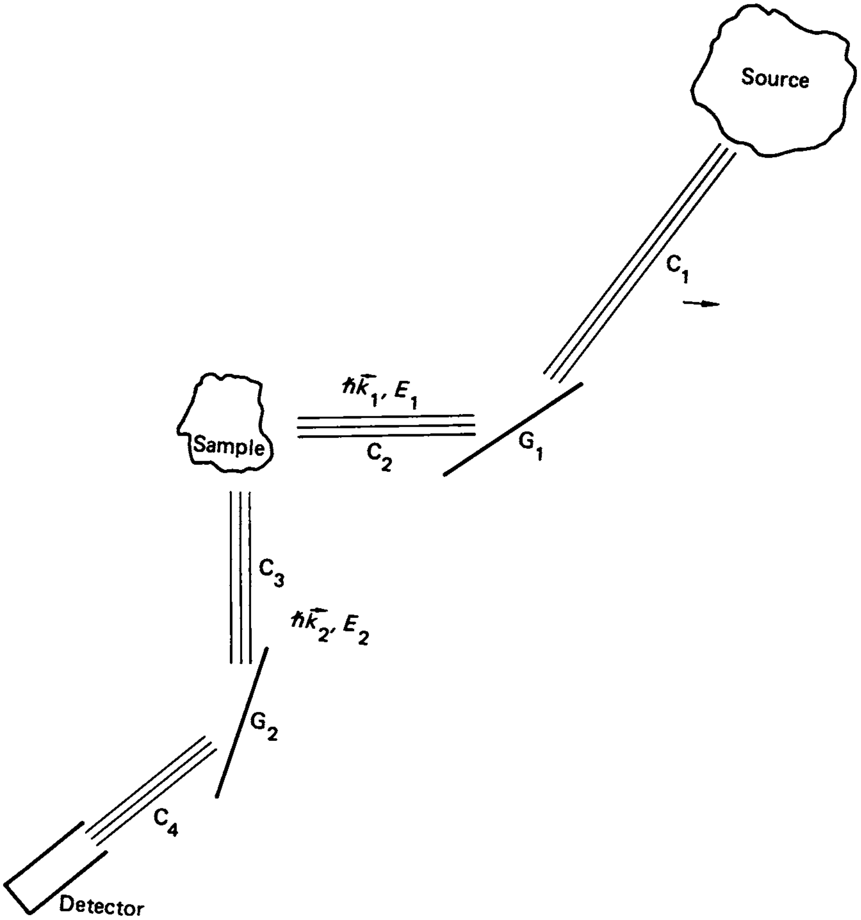


FIGURE 4.1 Schematic diagram of triple-axis spectrometer. Energy transfer = $E_1 - E_2$. Momentum transfer = $\hbar k_1 - \hbar k_2$.

energy-momentum space (e.g., critical scattering, phonon dispersion relations in crystals, etc.), whereas TOF spectroscopy is preferable for broad responses, diffuse in either energy or momentum space.

For standard diffraction applications, the spectrometer illustrated in Figure 4.1 can be simply set for zero energy transfer, since Bragg scattering is elastic. Alternatively, and more conventionally, the second crystal G_2 is removed so that the neutrons pass through collimator C_3 directly into a detector.

In the neutron TOF technqiue, the beam is split into a periodic sequence of short bursts and the distribution of wavelengths (hence energies and momenta) is measured by recording the detected neutrons as a function of the time taken to travel a known distance. To study inelastic scattering with this technique, the wavelength of either incident or scattered beam is fixed, while the wavelength distribution of the other beam is measured by TOF. In the example given in Figure 4.2(a) of a TOF inelastic scattering spectrometer at a steady-state reactor, the wavelength of the incident beam is fixed by the crystal monochromator system (as in the triple-axis spectrometer in Figure 4.1). The beam is split into bursts by a rotating mechanical chopper placed for convenience just before the sample, and the wavelength distribution of the scattered neutrons is deduced from the TOF spectra measured in the detectors.

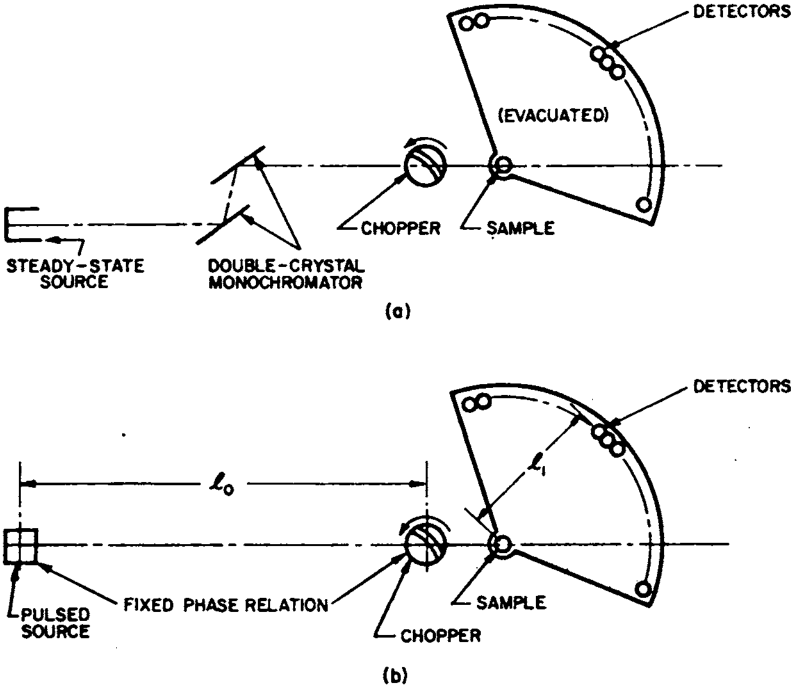


FIGURE 4.2 Schematic illustration of time-of-flight scattering instrument operating at (a) a steady-state reactor and (b) a pulsed neutron source.

Comparing this instrument with the triple-axis spectrometer there is (i) the obvious disadvantage of a low duty cycle (typically 1 in 500), (ii) the compensating advantage of measuring many (typically, 100 or 200) values of scattered neutron energy simultaneously, and (iii) the additional advantage of a detector array that can subtend a large solid angle at the sample (maybe a hundred to a thousand times that of a typical triple-axis spectrometer). Thus, as stated above, the TOF spectrometer will be more useful for studying those problems where extensive coverage of energy and momentum transfer is required and less useful if only limited energy and momentum transfer information is needed because then the duty-cycle factor becomes paramount. The implications of these features for the various types of problems studied with either instrument are discussed in Section 4.3.

At a source that is already pulsed, the TOF method compensates for the low duty cycle for reasons similar to those given above. In principle, one can use the apparatus shown in Figure 4.2(a) and simply dispense with the chopper. In practice, the apparatus shown in Figure 4.2(b) is more efficient, dispensing with the crystal monochromator and fixing the incident-beam wavelength through the phase relation between chopper and source.

4.2. LOW-ENERGY NEUTRONS FOR STRUCTURE STUDIES

The use of low-energy neutrons has proved to be invaluable and often indispensable for many studies of the structure of condensed matter. Among the factors that can make neutron diffraction the method of *first choice* for a structure determination are the following:

1. Precise location of the positions of light atoms, most notably hydrogen and deuterium.
2. Many opportunities for discrimination between neighboring elements in the periodic table.
3. Possibilities for altering the cross section of an element by isotope substitution.
4. The essential constancy of the nuclear form factor with scattering angle.
5. Absorption characteristics that usually are very different from those of x rays and electrons.

In addition, the presence of a magnetic moment on the neutron allows the direct determination of a magnetic structure--the arrangement of spins, their magnitude and direction. Neutron diffraction is the *only* means for obtaining such information for magnetic materials.

The various advantages in the use of low-energy neutrons as discussed in Section 4.1 have been exploited in structure studies of all the classes of crystalline materials--magnetic materials, inorganic compounds, organic compounds, and metals and alloys. Nonetheless, despite the impressive number of contributions to present-day knowledge of condensed matter we shall show in the following that the full potential of neutron diffraction has by no means yet been attained. Great advances toward attainment of that potential can be expected in the foreseeable future, however, with the prospect of a new high-intensity source, recent developments in instrumentation (e.g., two-dimensional detectors), and the availability of new techniques such as powder profile analysis and small-angle neutron scattering (SANS).

4.2.1. Accomplishments and Future Opportunities of Structure Studies with Neutrons

In this section we note some of the past and current accomplishments of neutron diffraction studies and particularly the opportunities for both scientific and technological advances for the different categories of materials.

Inorganic Structures

This is a classical area for neutron diffraction, the one in which the first major impact of neutron diffraction on structural science was felt. The earliest structure triumphs of the new tool came from the precise hydrogen location studies in important model compounds such as NaH, KHF_2, D_2O ice, NH_4Cl, and KH_2PO_4. Each of these studies provided answers, obtained in no other way, to long-standing questions of structure in relation to properties and function and provided the incentives for rapid development of the new research area.

Metal Hydrides Some of the earliest neutron studies dealt successfully with proton locations in metal hydrides. Currently, technological applications of metal hydrides for hydrogen storage and for energy storage and transport systems require extensive study and evaluation of specific systems for these applications. Intermetallic compounds such as FeTi and $LaNi_5$ are currently under intensive study. Doping with other metals to form ternary alloys is known to influence the formation and decomposition of the hydrides that can be formed. Neutron diffraction and scattering studies are the methods of choice to examine the relation between structure and stability. Some of the hydrides must be maintained under high hydrogen pressure and examined over wide temperature ranges. Pulsed neutron techniques offer considerable advantages for such measurements since the data may be collected at a single scattering angle.

Solid Electrolytes Materials that conduct electricity by extensive movement of ions from site to site in the crystal are known as superionic conductors or solid electrolytes. They are of potential interest in high-capacity storage batteries. An important group of these materials includes the beta aluminas, which are composed of layers of $Al_{11}O_{16}^+$ blocks separated by planes of conducting ions such as Na^+ or Ag^+. Neutron structural studies, as well as x-ray diffraction studies, have established the basic structural arrangement showing that there are more available sites than ions for the conducting cations such as Na^+ (see Figure 4.3). More significantly, however, the use of neutrons has allowed a clear-cut discrimination (see Figure 4.4) between Al and Mg sites in the commercially important material β''-alumina, which is stabilized by small additions of Mg^{2+} ions. The differentiation between Mg and Al is practically impossible with x rays.

The ions move by diffusive jumps from one site to another but remain localized between jumps. Quasi-elastic neutron scattering permits observation of the jump times and the distances traveled. Alpha silver iodide is another prominent example of a cation-disordered solid-electrolyte for which neutron studies have contributed to the basic understanding of the cation motion. In this case, the silver ion appears to be carrying out two different types of motion on two different time scales. One of these is a diffusive motion in a restricted region, while the other is a translational diffusion, which might be of jump-diffusion type with a larger time constant. In the case of fluorides such as BaF_2 and PdF_2 the high ionic conductivity is due to the mobility of fluoride ions. Neutron studies have shown that at temperatures well below

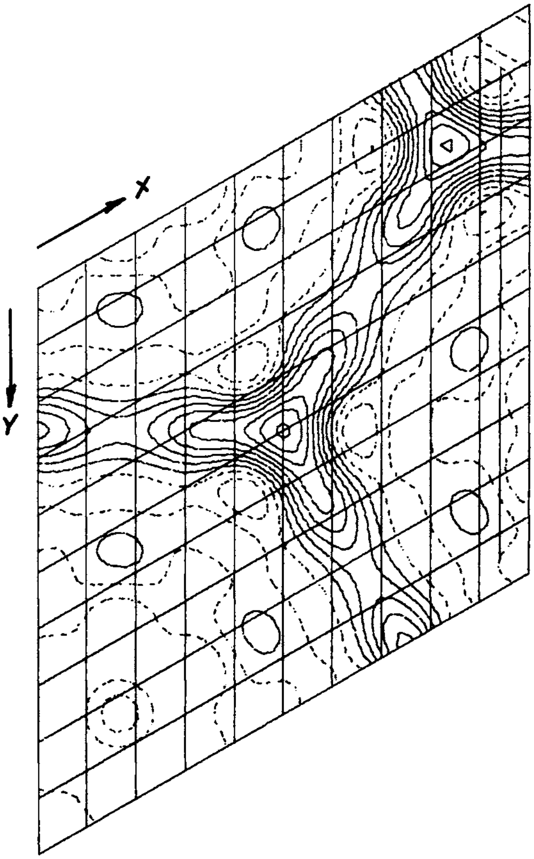


FIGURE 4.3 Sodium (nuclear) density in a conduction slab of β" alumina projected on the basal plane showing conducting cation positions in solid electrolyte structure. The difference Fourier was calculated to eliminate the oxygen nucleus located at $X = -1/3$, $Y = 1/3$. From W. L. Roth, F. Reidinger, and S. LaPlaca, in *Superionic Conductors*, G. D. Mahan and W. L. Roth, eds., Plenum Press, New York, 1976, p. 225.

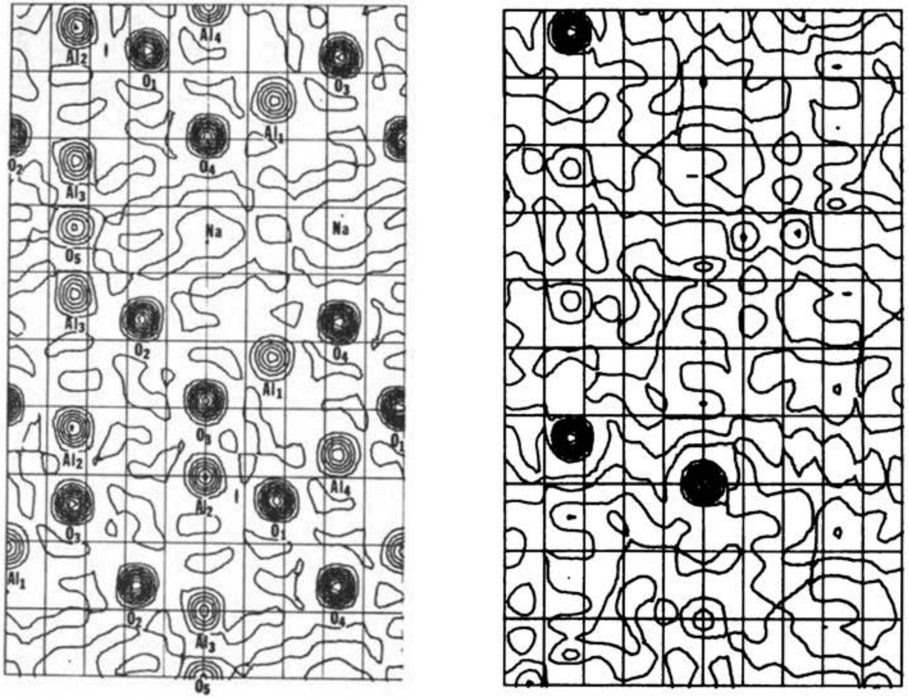


FIGURE 4.4 Nuclear density distribution (a) in the (110) plane of the solid electrolyte β"-alumina and (b) the difference Fourier map showing excess density at Al(2) sites due to the addition of stabilizing Mg atoms. From W. L. Roth, in *Crystal Structure and Chemical Bonding in Inorganic Chemistry*, C. J. M. Rooymans and A. Rabenau, eds., North-Holland Publishing Co., Amsterdam, 1975, p. 90.

the melting point, a large fraction of F^- ions have moved to interstitial positions.

There are many new solid electrolytes that have been discovered recently, few of which have been examined structurally. Neutron diffraction is again the method of choice, since these materials often contain heavy elements such as Ag, Hg, and Pb in the presence of oxygen or fluorine. The combination of neutron structure determination and quasi-elastic scattering analysis offers a most powerful combination of tools for the study of the mechanism of conduction.

1-D Conductors A new class of compounds based on square planar complexes of Pt and Ir has been shown to have very short metal-metal distances and high, anisotropic, essentially metallic conductivity. The most studied example is $K_2Pt(CN)_4Br_{0.3} \cdot 3H_2O$; this compound is partially oxidized, nonstoichiometric, and displays a basic electronic distortion that shows up in weak superlattice reflections. There is some hope that compounds with such properties may evidence high-temperature superconductivity, which would be of extreme technological importance in electric power transmission.

Neutron diffraction studies have provided the unequivocal answers concerning the basic structure of these 1-D materials. In addition, neutron study of the weak superlattice reflections has given a detailed picture of the nature of the Pt atom displacements that result from the charge density wave of a Peierls electronic distortion. These findings are an important key to the basic understanding of the electronic properties of these materials. This field is showing very rapid expansion and growth, and new and more complex materials are being produced. These new materials offer much promise, and investigations of structure in relation to physical properties will require even more powerful methods. Thus, both high neutron flux and high resolution will be required for detailed study of the complex superlattices that are increasingly in evidence.

Ferroelectrics and Antiferroelectrics Materials that have interesting electrical properties, such as ferroelectrics and antiferroelectrics, often undergo phase transitions to a disordered phase that is stable at a higher temperature. The ordered phase is often difficult to study because of the inevitability of twinning or because crystals often shatter when undergoing a phase transition. These problems can be circumvented by application of anisotropic pressure to a crystal to maintain an untwinned state or by using high-resolution powder methods with profile refinement when crystal shattering cannot be prevented. These methods are simpler to apply when high-intensity pulsed beams are available and fixed sample positions can be maintained during data collection.

The low-temperature structure of the antiferroelectric material $NH_4H_2PO_4$ has been accurately determined recently using such a profile refinement technique. This can easily be extended to more complicated materials, and measurements can be made on very small samples. The pulsed nature of a neutron source can be advantageously used to study transient effects such as was done recently in experiments with ferroelectric $NaNO_2$. By synchronizing an applied external electric field with the neutron pulse, it was possible to demonstrate that the NO_2 group rotates about the (001) axis when the polarization direction is reversed. Such measurements promise many new insights into mechanisms of phase transition.

Metal-Insulator Transitions Many rather complicated new mixed valence materials that undergo metal-semiconductor or metal-insulator transitions have been produced in recent years. These are often mixed alloy oxides or sulfides of rather complex structure, e.g., $Ba_{15}Fe_7S_{25}$ and $4Nb_2O_5 \cdot 9WO_3$. Some of these have interesting electrical properties, and careful structural studies are needed to investigate the phase transitions and to understand the electronic changes that are taking place. Neutron studies are again most suited to follow the subtle changes particularly in the light-atom distribution.

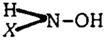
Mineralogy and Geology Actual minerals often are very complicated chemically and morphologically and appeal to neutron diffraction is only occasionally indicated. One area where neutron diffraction studies would be helpful concerns the ordering of Al and Si in various silicates, most importantly the feldspars. Deductions as to the ordering have been made mainly on the basis of interatomic distances derived from x-ray work. The direct discrimination between Al and Si is difficult with both x rays and neutrons but less so with the latter. The industrially important zeolites also are not well characterized generally as to the Si-Al ordering.

Minerals that are hydrates also would be better characterized with respect to the role of water by means of neutron diffraction. The other area where neutrons would be beneficial is in the study at high pressures, high temperatures, or both of minerals. Such studies would have considerable relevance to the geology of the earth.

Coordination Complexes In recent years, a wide range of coordination complexes has been prepared, especially of the organometallic type, and many have been studied by x-ray diffraction techniques. Neutron diffraction, on the other hand, can provide more precise information on lighter atom positions, on the hydrogen positions in unsaturated hydrocarbon groups such as $C_5H_5^-$ and C_2H_4, and on the positions of hydrogens directly attached to or bridging metal atoms. It can also furnish information on the orientation of water molecules coordinated to metals and readily distinguish between carbon and nitrogen in CN^- groups, where the difference in x-ray scattering would be very small. Such information should serve as a valuable input to theories of metal-ligand bonding and provide a sensitive probe for the evaluation of asymmetry effects in such coordination complexes.

Thus far, the number of neutron diffraction studies has been quite limited, largely because of difficulties in obtaining crystals of suitable size for such studies. The availability of high-flux steady-state or pulsed sources would greatly alleviate this problem and open the use of neutron diffraction to a wide range of potentially important materials.

Organic Structures

Chemical Binding Since roughly half of the atoms of organic compounds are hydrogen, neutron diffraction is often the best and may be the only practical tool for obtaining the most significant information pertinent to binding in organic compounds. For example, establishing that the hydroxamic acid group  ${}^{H}_{X}{>}N\text{-}OH$ is nonplanar in the compound hydroxyurea ($H_2N\text{-}CO\text{-}NHOH$) is impossible without accurate location of the hydrogen atoms.

Availability of accurate atomic and molecular geometric parameters is essential for current and future studies into the nature of chemical forces. Accurate structural data can be analyzed in terms of interatomic and intermolecular potential functions, which yield improved understanding of the origin and nature of chemical forces and may ultimately make possible prediction of the structure and properties of new and different materials. A pertinent example is the field of overcrowded organic molecules, in which the distribution of strain in response to the stresses from overcrowding yields directly the relative resistance of a molecule to various modes of distortion. Hydrogen atom locations are essential to a proper analysis of these stresses.

Organic free radical molecules may have electrons with unbalanced spins that can be explored by means of the magnetic interaction with neutrons. This type of study, as yet untried, promises important future results.

Recent experiments started at the Massachusetts Institute of Technology and pursued at Argonne National Laboratory have explored the feasibility of measuring interatomic diamagnetic currents in materials. The effects that are measured are very weak, and the source intensity is a real limitation. Such measurements can lead to a spatial mapping of the diamagnetic currents and should be of great interest in connection with bonding models.

Catalysis Well-conceived studies on structures of model compounds can be expected to yield valuable clues in this field of overriding practical importance. We can look forward to the synthesis of molecules imitating the active sites of biological enzymes and to molecules in which such sites are deliberately modified. The accurate determination of such molecular structures, including the hydrogen atoms, will be needed for an understanding of their behavior and as a guide to further synthesis. Already, molecules composed of clusters of several metal atoms bridged by organic ligands are being synthesized and studied by standard techniques, but neutron diffraction has not yet been applied.

Many catalysts, for example, uranium alkyls, and other organometallics contain heavy metals along with organic ligands; hydrogen positions, and possibly hydrogen-deuterium exchange distributions, would be highly informative.

Ferroelectricity Ferroelectricity is a property of much scientific interest and practical importance that has its basis in molecular structure. Hydrogen atoms are inevitably of basic importance, and neutron studies would be needed for their accurate location. An example is the ferroelectric crystal copper formate.

Mechanisms of Reactions Since neutrons can distinguish between different isotopes of an element, neutron diffraction can be used to trace the course of chemical reactions.

The H-D distribution at specific atomic sites can be quantitatively determined by neutron diffraction.

Isotopic Equilibrium The isotope effect on chemical equilibrium in systems with labile hydrogen atoms has been studied and can be further exploited. For example, it has been shown that short, strong hydrogen bonds tend to be enriched in the ^1H isotope, while the longer and weaker ones become enriched in ^2D.

Metal Structures and Metallurgy

Crystal Structures The importance of neutron diffraction for crystal structure determination of alloys was demonstrated in the earliest days of neutron diffraction experimentation. The clear-cut evidence for ordering in FeCo immediately suggested the advantages of neutron diffraction over x-ray diffraction for structure determination of alloys containing elements adjacent in the periodic table. This advantage has been considerably exploited for a variety of systems, including complicated structures such as the sigma phase, which contains varying proportions of transition elements, e.g., Co_7Cr_8, $Ni_{13}V_{17}$, and FeCr. Another case for which the use of neutrons may be the method of choice over x-ray diffraction involves structures containing light elements combined with heavy elements, e.g., UH_3.

There are many other situations for which the nuclear-scattering cross sections involved are such as to dictate the use of neutron diffraction over x-ray diffraction, although there are converse examples. Accordingly, neutron and x-ray diffraction are complementary tools for crystal structure determination of alloys.

Solid Solutions In addition to the well-ordered structures, alloys quite commonly occur as solid solutions over extensive ranges of composition. The characterization and understanding here is much less clear despite a great deal of effort in the past. It is an area where there is a potential for new significant contributions from neutron investigations, especially with the availability of higher fluxes. Some of the problems that could be tackled profitably with new techniques are short-range order and clustering, distortions, interatomic potentials, interstitials, precipitates, and kinetics.

With regard to short-range ordering or clustering phenomena, the power of neutron diffraction has previously been demonstrated in various cases, most elegantly for $Cu_{0.52}{}^{62}Ni_{0.48}$, a null matrix, in which the normal coherent scattering is balanced to near zero but in which the distinction between the two atomic constituents becomes very pronounced. Clearly it would be unthinkable, for Cu-Ni alloys, to attempt to obtain the information on clustering of atoms with x rays. Other alloy compositions can still present problems even with neutrons, but here the possibility of energy analysis of scattered neutrons allows clarification of the process of clustering, or short-range order, by recovering only the elastic scattering.

For comparison with alloy theory, one would wish to study very dilute solutions or even investigate defects such as self-interstitials for pure elements. One could then hope to obtain interatomic potentials. It has not been feasible to make such studies because of the very weak effects resulting from the low concentrations of solute or defects. A promising start has been made with the advanced facilities at Grenoble and Jülich. There is a clear need here for higher flux and improved techniques of analysis.

Other interstitials such as carbon, nitrogen, or oxygen as well as self-interstitials in low concentrations are very difficult to detect with conventional neutron or x-ray scattering but might be readily investigated with the new advanced techniques of pulsed sources. These studies could make important contributions toward understanding mechanisms of diffusion and phase transformation.

It would be desirable to learn more details of precipitate nucleation and growth. Some of the required information might be provided by small-angle scattering but also could be obtained with pulsed neutron sources and time-of-flight spectrometers. There is also the prospect of observing the kinetics of the various processes--ordering, clustering, transformations--by studying diffuse scattering as a function of time.

Glassy Metals Many studies have been made with x rays and only a few with neutrons. The advantages of neutrons for analysis of other glass structures should be utilized for these technologically interesting systems.

Preferred Orientation A study with neutrons of preferred orientation in uranium bars has indicated the obvious advantages in this case over x-ray methods, and recent work has been done in developing neutron diffraction as a tool for studies of grain orientation as a function of sample depth. Another application is preferred orientation with respect to the magnetization distribution in magnetic materials, a virtually untouched area.

Topography Neutron diffraction topography is a recent technique with considerable potential for study of crystal inhomogeneities. A very interesting application [J. Baruchel, M. Schlenker, and W. L. Roth, *J. Appl. Phys.* *48*, 5-8 (1977)] has been the first *direct* observation of antiferromagnetic domains in NiO. As illustrated in Figure 4.5, different magnetic reflections image one type of domain at a time yielding unambiguous identification of the magnetic arrangement in each domain. Further application of the technique for observation of dislocations and other structural defects needs to be explored.

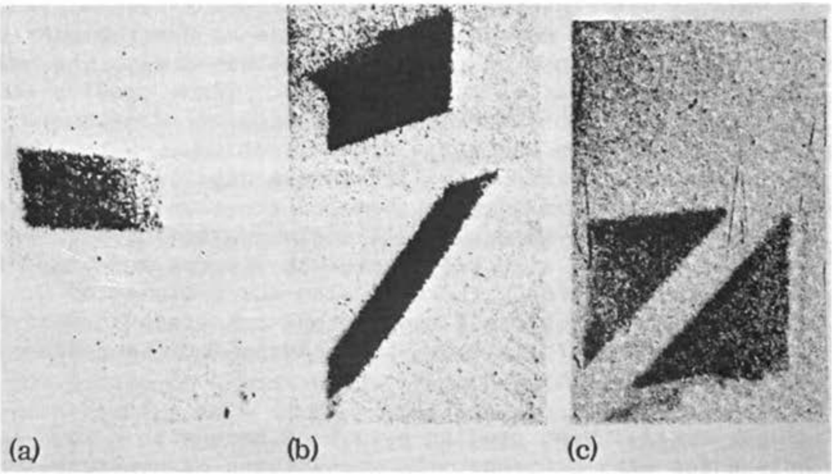


FIGURE 4.5 Magnetic neutron diffraction topographs of an antiferromagnetic NiO single crystal showing domain structure. Scale marker: 1 mm. From J. Baruchel, M. Schlenker, and W. L. Roth, *J. Appl. Phys.* *48*, 5 (1977).

Magnetic Structures

The magnetic moment associated with the neutron has made it a unique tool for the study of magnetism on a microscopic scale. Those topics that are studied by *elastic* or quasi-elastic scattering are reviewed here; areas of research that are studied by inelastic magnetic-scattering techniques are treated in Section 4.3. It is fair to say that the full potential of magnetic scattering has yet to be realized. The recently developed powerful methods of spin spectroscopy and polarization analysis are still restricted by low intensities.

Magnetic Configurations The earliest investigations of conventional magnetic structures were associated with understanding the behavior of bulk properties of crystalline solids; and since the period of the first experiments in 1948-1951, a vast literature of magnetic structures has evolved. At present, it seems certain that a magnetic-structure determination will be a part of any program in which the macroscopic properties of materials are investigated. A knowledge of the magnetic structure of a solid is always necessary before any dynamical studies are undertaken; it gives the first clues toward understanding the nature of the exchange and anisotropy energies of the moments of the atoms in the structure.

Such magnetic-structure determinations are subject, more or less, to the same constraints as are crystal-structure investigations, and very high fluxes are not essential. Also, profile analysis can be applied to powder patterns for magnetic structures as well as for crystal structures. Furthermore, the solution of even complex magnetic structures has been aided greatly by the group theoretical methods developed by workers at Grenoble.

Magnetic Phase Transitions and Critical Scattering The transitions that magnetic materials undergo when they become ordered ferromagnets or antiferromagnets have attracted great interest from both experimentalists and theorists. These magnetic systems provide general information on a wider body of phenomena associated with the behavior of systems in the neighborhood of a critical point. The modern era in this field began in about 1965 with the development of two important concepts: *scaling* and *universality*. The fundamental idea of a second-order transition is that it is describable by an order parameter that serves as a measure of the amount and kind of ordering that arises in the critical region. In the case of a ferromagnet, as an example, the order parameter is the magnetization, M. The order parameter must be nonzero below $T_{Critical}$ and must approach zero continuously as $T \rightarrow T_C$ from below. The approach to zero is generally given by a power law

$$M \sim \left(\frac{T_C - T}{T_C}\right)^{\beta},$$

where β is a so-called "critical exponent." The singular behavior of other equilibrium properties such as susceptibility and specific heat are also described by power laws with critical exponents. "Scaling theory" leads to relations among the various critical exponents. The concept of universality relates to the classification of second-order transitions. Neutron-scattering experiments, particularly on magnetic systems, have provided the most comprehensive evidence for the validity of these phenomenological theories.

In 1971, a new and "revolutionary" concept of adapting renormalization group techniques to the problem of phase transitions turned the phenomenological theories into true calculations of critical-point behavior. Once again, neutron-scattering experimentation on magnetic systems has been a primary experimental technique in checking these calculations.

The experiments in this area are difficult and require the most intense neutron sources available.

Magnetic Form Factors--Magnetic Moment Densities An important contribution of magnetic neutron scattering is the information it has provided on magnetic moment densities, and hence to electronic wavefunctions, in condensed matter. This comes about because the elastic magnetic neutron scattering is given by the Fourier inversion of the time-averaged magnetization density. The aim of a form factor experiment is to measure the magnetic contribution to as many Bragg reflections as possible, obtaining the magnetic scattering amplitude as a function of $K = 4\pi \sin \theta/\lambda$.

Historically, measurements of magnetic form factors were first made, and are still being made today under certain conditions, with an unpolarized neutron beam. (Every magnetic structure determination involves the measurement to greater or lesser precision of a magnetic form factor.)

In such a case, the nuclear and magnetic contributions to the intensity of a Bragg peak are additive. This may be satisfactory when the magnetic, *p*, and nuclear, *b*, amplitudes are about equal, the case for the inner reflections in some substances; but for the outer reflections where $p \ll b$ the magnetic scattering is completely dominated by the nuclear intensities. There are cases for which one needs to make significant measurements in the range $p/b \lesssim 0.01$.

The next level of complexity designed to make measurements of small amplitudes is the standard polarized beam method exploited first in research work at MIT. One measures the ratio $R = I^+/I^-$ of peak intensities of a Bragg reflection as a function of the direction (+ or -) of beam polarization. This flipping or polarization ratio can be determined to very high statistical accuracy, and its sensitivity to small values of *p* is greatly enhanced over the case above. The technique has the great advantage that each observation is self-calibrating assuming that the nuclear amplitude is known.

The ideal way to make flipping ratio measurements is to use a polarized beam with polarization analysis after the sample. This technique was first explored at MIT and then exploited in work at ORNL. As yet, this type of experiment has been performed only in a few special cases because the intensity attrition is too severe with present-day monochromators and polarizers to use this method routinely.

Precise measurements of magnetic form factors have been reported for a large number of ferromagnetic and ferrimagnetic materials--Fe, Co, Ni, ^{160}Gd, Tb, and Er, for example --as well as for compounds, ferrites, and intermetallic compounds. For the most part these were made, or could have been made, at reactors with modest fluxes, although source intensity always places a limit on the amount of available magnetic-scattering data. Having high intensity at high neutron energy means that larger momentum transfer (higher Bragg reflections) becomes accessible for study.

The same techniques have been applied to paramagnetic metals Sc, Y, Nb, Lu, Pd, Pt, Cr, and V, for example. In these experiments, a magnetic moment of several millibohr magnetons is induced in the specimen by means of a strong magnetic field. The distribution of the induced magnetization is then inferred from polarization ratio measurements, and information about the wavefunctions of electrons near the Fermi surface is thereby obtained. Generally, such experiments *require long counting times even at high-flux reactors.*

Moment Distributions in Disordered Systems A very important application of magnetic neutron scattering is to the measurements of moment distributions in disordered systems. Of these, the greatest effort has been in the study of ferromagnetic systems. The first experiments of this type were carried out at the early Oak Ridge graphite reactor now decommissioned. Very extensive measurements were later made at the Harwell Atomic Energy Establishment in England. This type of experimentation is presently being done at Oak Ridge and by scientists at the Institut Lane-Langevin (ILL) in Grenoble.

In the formation of an alloy, the electron shells of the atoms exert a mutual influence that is revealed by a change in the magnetic properties of the atoms. Neutron-scattering measurements on disordered systems can provide quantitative information about the influence of the surroundings on a given moment and about the interactions between the alloy components. For a purely random alloy, measurements of the Bragg reflections yield information about the average moment. To assess the values of the individual moments, it is necessary to make measurements of the magnetic disorder scattering.

In practice, the problem of determining the individual moments is complicated by several factors. These include the nuclear disorder scattering, multiple scattering, and inelastic effects. In addition, alloys are rarely perfectly random so that short-range ordering plays a role. Several experimental methods are in use to study disordered ferromagnetic systems, and these differ in the method used to extract the magnetic disorder scattering without the perturbing contributions mentioned above. The use of both polarized and unpolarized neutron beams produces the greatest amount of information.

Diffuse scattering intensities are generally quite low so that high-intensity sources are important for such studies. It is probable that experiments of this nature will be continued and the concentration of impurities pushed to very low values. More importantly, the diffuse scattering studies will be extended to antiferromagnetic and paramagnetic systems. For these, it seems probable that the technique of spin spectroscopy will be required. The technique utilizes a polarized beam with polarization analysis after the sample.

Long-Range Moment Density Oscillations--Small-Angle Scattering Small-angle neutron scattering can be used to examine any phenomenon that produces localized changes in small regions of a material that result in a different net neutron-scattering amplitude for those regions. It can be used to study, for example, long-range spin correlations in magnetic systems, fluxoid lattices in superconductors, clustering in alloys and amorphous magnets, domain wall effects, and magnetic polymers. The requirements for magnetic small-angle scattering experiments are, generally, the same as those for nuclear small-angle scattering. One would prefer to have a high-intensity source and, if possible, to use long-wavelength neutrons produced by a cold source. Many experiments can, however, be performed effectively without a cold source, using neutrons in the wavelength range 2.5-4.5 Å. Some of the difficulties of multiple scattering can be lessened by going to pinhole geometry and regaining intensity with an area detector.

Small-angle diffraction by fluxoid lattices has been carried out with a nearly perfect double-crystal system at the HFIR, which provides extremely high resolution. Studies of magnetic polymers could be investigated with both types of system. Critical scattering from ferromagnets (EuO), superparamagnetic clusters, and investigations of correlations in spin glasses provide a few of many applications.

Polarization Analysis--Neutron Spin Echo A number of new applications of polarized beams are under investigation. Among the more interesting devices is the neutron spin-echo device invented by Mezei at Grenoble. This is a device with which very-low-energy changes can be measured with precision and will be used to study quasi-elastic scattering phenomena. High fluxes and long wavelengths are required, and the device should be useful for the study of colloids and macromolecules. Neutron spin spectroscopy and polarization analysis have application to systems that do not have electronic magnetic moments but rather use the moment of the neutron as a probe to give additional information about the target system.

Conclusions It seems quite certain that in the next generation of magnetic scattering studies--elastic, quasi-elastic, and inelastic--polarized beam methods will play a dominant role. There is a clear need for very high neutron flux in this work. However, in order to obtain polarized beams from a pulsed source, it will first be necessary to adapt techniques now in use for nuclear studies in a different neutron energy range. This instrumentation development of efficient neutron filters must therefore be accomplished before a pulsed source can be used for these kinds of studies.

4.2.2. New Horizons

Several techniques have been developed recently that could greatly extend the scope of neutron-structure studies. In addition, the availability of a high-flux pulsed source provides further opportunity to do various unconventional and unusual types of study that have not been feasible heretofore.

Neutron Powder Diffraction and Profile Analysis

The technique of neutron powder diffraction has long been used in studies of materials having simple structure. With it, information can easily be obtained about the effect of temperature on crystal structure, magnetic properties, and phase transitions. However, in the past, severe limitation on its applicability was imposed by the requirement that complete or nearly complete resolution of the diffraction pattern had to be obtained before any interpretation could be attempted. This permitted useful work on problems having at most ten variable structural parameters. The scope and promise of neutron powder diffraction has recently been greatly expanded by two recent developments. The first is the realization that to obtain the most information from a diffraction pattern, the entire diffraction profile must be utilized in any structural analysis. The second is the use of a pulsed neutron beam of high intensity suitable for time-of-flight analysis.

Pattern Fitting Structure Refinement The technique of pattern fitting structure refinement (PFSR), also called profile analysis, was first described by Rietveld and has become the method of choice for the analysis of neutron powder diffraction patterns. In this method, each Bragg reflection is assumed to be Gaussian in shape, and the profile is calculated from the sum of Gaussian peaks corresponding to the set of reflections. Thus each intensity observation along the profile is the sum of contributions from the reflection peaks that overlap at that point, and the weighted difference between the observed and calculated profiles is minimized by a least-squares refinement. The calculated profile is obtained by summing the results of a structure factor calculation based on a set of atomic positions, thermal parameters, and occupancy factors with the peak shapes and positions dependent on lattice parameters, diffractometer zeropoint, and three half-width parameters. All of these parameters, both structural and instrumental, are evaluated in the above-mentioned least-squares refinement.

It is clear that this technique utilizes all the information inherent in a given neutron powder diffraction pattern, but how much improvement can be gained? In a recent survey of PFSR studies, over 150 structure refinements were listed, of which 10 involved structures with 20 or more refined atomic parameters. In one case, *ortho*-$Ti_2Nb_{10}O_{29}$, 41 atomic parameters were refined by full-matrix least-squares analysis as given by R. B. Von Dreele and A. K. Cheetham [*Proc. R. Soc. Lond. A338*, 311 (1974)]. The diffraction pattern (Figure 4.6) is a composite of 843 unique Bragg reflections and consisted of 1062 observations with a contribution above background level. This example illustrates one of the best uses of neutron diffraction because it took advantage of the extreme variation in neutron-scattering lengths for the various atoms to accurately determine the cation distribution over the six unique octahedral sites in the structure. In addition, the refined oxygen positions were more precise by a factor of ~10 than those obtained in an earlier single-crystal x-ray study.

As to further possible extensions of the PFSR technique, it is evident that for small problems with less than 20 parameters most of the presently existing low-resolution instruments can be used. Clearly these instruments are unsuitable for examination of complex materials. To be able to examine sensibly the structures of complex materials having 20 or more structural parameters, neutron powder diffractometers with better resolution characteristics are required. There are several such instruments currently available of which only a few are present in the United States. The pattern shown in Figure 4.6 was taken on one of these instruments (PANDA diffractometer at the PLUTO Reactor, Harwell, England). Experience suggests that problems with ~100 structural parameters can probably be refined using neutron-powder-diffraction data taken on one of these instruments. A further improvement in the

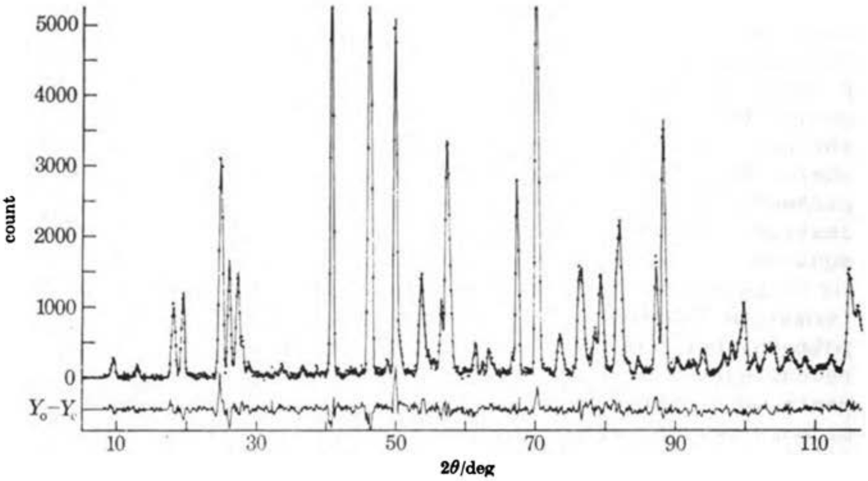


FIGURE 4.6 Neutron powder diffraction profile for *ortho*-$Ti_2Nb_{10}O_{29}$. Lines and points represent calculated and observed profiles, respectively. A difference curve is also shown. From R. B. Von Dreele and A. K. Cheetham, *Proc. R. Soc. London A338*, 311 (1974).

resolution of these instruments could extend the PFSR technique to more complicated problems but at the expense of intensity, although this can be compensated for by using multidetectors. To date, of the approximately 150 structures analyzed by PFSR only a small fraction of the analyses has been performed in the United States. Most analyses have been performed at Harwell in England, at the ILL at Grenoble, and at Petten in Holland. Dedicated instruments for profile analysis have been or are being developed at several U.S. laboratories.

Pulsed Neutron Sources The characteristics of pulsed sources will have a great impact on many different kinds of neutron-scattering experiments not the least of which is neutron powder diffraction. In the typical time-of-flight (TOF) powder diffractometer the geometry is fixed and a scan in time of the intensity gives the diffraction pattern of the sample as a function of neutron wavelength. A TOF powder diffractometer at a high-intensity pulsed-spallation source such as IPNS-II could have tenfold better resolution, with equivalent intensity, than a high-resolution diffractometer at a high-flux reactor.

Preliminary investigations using LINAC and spallation pulsed sources of known test specimens indicate that with the proper peak shape functions, the PFSR technique can be applied to these TOF patterns. It is clear that when pulsed neutron TOF diffractometry is combined with the PFSR technique of analysis, a considerable extension of the range of structures amenable to neutron powder diffraction is possible. In what could be a conservative estimate, it is felt that structural problems with 200-300 variable parameters could be examined successfully with these techniques. This would then be an extremely powerful structure determining tool which could handle nearly all small and medium size crystal structure analyses currently done by single crystal techniques. In addition, the experimental conditions can be varied over a wide range of temperatures and pressures.

Small-Angle Scattering

Low-energy neutrons can be used to examine large spacings and defect aggregates within solids in ways completely analogous to the well-practiced technique of small-angle x-ray scattering (SAXS). There are a number of differences between small-angle neutron scattering (SANS) and SAXS, however, that can make SANS a highly useful technique, complementing both SAXS and transmission electron microscopy (TEM). In fact, the Europeans have established a number of centers for SANS research, which have become saturated by interested investigators.

The two major research centers for SANS are the ILL in Grenoble, France, and the Institut für Festkörperforschung-Kernforschungsanlage (IFF-KFA) in Jülich, Germany. Grenoble is an international center for SANS studies, the ILL being administered by a troika from the United Kingdom, France, and Germany. There are currently two SANS guide tubes and spectrometers at ILL. What makes ILL especially interesting is a cold source, within the reactor, yielding a high flux of long-wavelength neutrons (up to 20 Å). This permits studies of crystals in which, previously, a problem was faced with double-Bragg scattering effects, for example, in plastically deformed crystals. Thus, the avoidance of double-Bragg scattering is an important aspect of SANS. Such troublesome scattering can be avoided by using a wavelength that is at least twice the lattice parameter of the crystal in question. It is important to recognize that the most efficient way, although not the only way, of performing SANS experiments in crystals is with a cold source.

Further uses of SANS have developed because of limitations of SAXS for examining clustering in solutions in which the atomic scattering factors are too close to achieve significant SAXS intensity. SANS can enter here as an effective alternative, since the scattering cross section does not vary in a predictable, smooth fashion with atomic number, as is the case for x rays. So SANS can be profitably used when SAXS cannot. Some cases in point include Pt-Au, Al-Mg, and Al-Si, to name but a few. There are large numbers of alloy and ceramic systems for which SAXS cannot be employed but for which SANS can. This is most important in metallurgy and materials science.

Another aspect of neutrons is their low absorption characteristic. Where x rays in transmission may have optimum thickness of tens of micrometers, neutrons can effectively penetrate a thickness measured in millimeters or centimeters.

Thus, alloys of Pb, for example, hitherto not studied by SAXS are natural candidates for SANS. Also, one can think about a range of applied experiments--nondestructive evaluation of, for example, turbine blades, which, under high-temperature centrifugal load-induced creep conditions, develop fine voids or cavities well before failure. This has actually been done by FIAT workers in Italy, where turbine blades after field service are routinely tested at a reactor neutron source. Applied SANS is a virtually untouched field with great potential.

Neutrons are influenced by the atomic magnetization produced by unbalanced electron spins or by orbital electronic currents. For a number of years, wide-angle neutron scattering has been used to probe the structure of many magnetic crystalline systems. Today, SANS is exploring magnetic clusters in alloys and amorphous magnetic alloys.

The list in materials science for the potential applications of SANS is impressive indeed: polymers, voids in irradiated metals, and, for applied SANS, coal and fine-particle analysis. Further activities will take place in chemistry, involving dilute solutions and catalysis. Biophysics has made extensive use of SANS to study deuterated molecules and the structure and dimensions of macromolecules.

There are a number of research reactor facilities within the United States where SANS experiments can be carried out.

However, we have no facilities approaching ILL at Grenoble. This dedicated SANS center attracts scientists from around the world and with them comes the excitement and enthusiasm that makes ILL such a successful establishment.

There should be a wider appreciation of the great potential and versatility of the SANS technique, and the United States needs expanded instrument facilities in this area, as stated in Section 5.4.1. It should be understood, of course, that in many of its applications SANS provides a complementary measurement capability to SAXS such that both of these techniques are required for the full exploitation of low-angle diffraction in science and technology.

Liquids and Glasses

By the term "structure" as applied to noncrystalline materials is usually meant the delineation of pairwise correlation functions (and sometimes higher-order correlation functions) for the component atoms or molecules. The importance of such information lies in the fact that these pair correlation functions constitute the most immediate common ground between the statistical-mechanical theorist and the experimentalist. Given reliable, sufficiently detailed correlation functions, the path to computing thermodynamic functions is (within the domain of two-body forces) straightforward.

Diffraction methods provide direct experimental access to pair-correlation functions but not in all desired detail. Combined use of neutron and x-ray methods and, in the case of neutrons, use of several isotopic compositions (in cases in which different isotopes have sufficiently different scattering amplitudes) can enhance the needed detail. The most successful research is likely to involve close collaboration between theorist and diffractionist, since the experimental results can serve as the first check on the validity of a theoretical model and a means of establishing values of disposable parameters.

Prospective applications of both practical and esoteric interest are numerous. The combination of a successful theoretical model, checked against diffraction data to prove its validity and to evaluate its parameters, can be a vehicle for predicting thermodynamic and transport properties of numerous systems, for interpolating such values when direct experimental measurements are sparse, and for extrapolating to conditions difficult to realize in the laboratory. One route to such results is, of course, computer simulation; for this method the effective interatomic potential is needed; and again the first and most direct check of a proposed potential, as well as computational procedure, is the comparison between diffraction measurements and computer correlation function.

In terms of the variety of materials of interest, one can list liquid metals and mixtures of metals, molten salts, liquid hydrocarbons, glasses, and aqueous solutions. For glasses, the need for higher spatial resolution makes neutrons of short wavelength and high intensity valuable. These systems are therefore best studied at pulsed sources or, less desirably, at steady-state devices equipped with a hot source.

Surface Phenomena

Neutron-scattering measurements have recently been used to study surface phenomena. These studies have indicated that there are cases for which the information obtained is not otherwise available using more conventional techniques. There are two aspects of the neutron measurements that make them appealing. The first relates to the study of lightly bound gas films on solid surfaces, which are usually *disrupted by the commonly used "strongly interacting" probes such as electrons or x rays* but which are *left intact by the "weakly interacting" neutron probe*. The second aspect relates simply to the study of hydrogen-containing materials on surfaces. The surface phases formed by hydrogen, deuterium, nitrogen, oxygen, and xenon on graphite have been characterized. The melting of these two-dimensional commensurate as well as incommensurate structures has been investigated, and whereas the commensurate phase melts abruptly, the incommensurate one seems to have a smooth and continuous change from the ordered (solid) to the disordered (liquid) phase. More complicated systems such as methane adsorbed on zeolite or hydrogen on Raney nickel have also been studied with emphasis on surface diffusion. Further work in this field is of interest for both practical applications to catalysis as well as for broadening our understanding of surface phenomena.

Definitive studies of surface structure phenomena must be performed with extremely small specimen volumes, and high intensity of the "probing" radiation is of paramount importance. This importance is even further emphasized in studies attempting to establish the dynamical properties of surface films or layers. Clearly, the high-intensity characteristic of a pulsed neutron source is a real advantage.

Structures under Extreme and Unusual Conditions

There is an increasing need for studying the structure of materials under extreme conditions, particularly of high temperature and high pressure. The information obtained from such studies would be useful for choice of materials in various energy technologies, for learning more of geological processes in the interior of the earth, and for general knowledge of phase equilibria. Neutrons offer various advantages in allowing simple design of temperature or pressure cells because of the smaller absorption of neutrons by most materials used for containment. An especially attractive feature is the possibility of using only one or two exit windows with TOF methods.

A high-intensity neutron source, used in conjunction with TOF techniques, affords possibilities for studies of crystals subject to peak compressions and of the *excited states* of molecules. A pulsed compression experiment could be carried out synchronized with the neutron pulses to investigate materials subjected to high pressures. Also, a strong beam from a pulsed laser or a strong magnetic field synchronized with the neutron pulses could be used to study excited states provided a sufficiently high percentage of the molecules were excited to allow coherent diffraction to be measured. There are a number of interesting possibilities that should still retain the overall crystal structure, such as those involving intramolecular hydrogen transfer or selective excitation of normal modes, thus leading to a direct determination of the displacement coordinates through analysis of anisotropic Debye-Waller factors.

The very high fluxes of pulsed neutron sources could be exploited on structure studies involving time-dependent phenomena such as phase changes, kinetics of precipitation, ordering or clustering in alloy systems, crystallization in glass-ceramic systems, and chemical changes, to cite a few examples.

Basic Crystallographic Studies

Electron Density The determination of experimental electron densities is still in its infancy when one considers what could be accomplished in this area. Here again accurate x-ray and neutron-diffraction measurements provide complementary information. Neutron diffraction allows one to determine the position and true vibrational motion of an atom, whereas x rays sense electron positions. The number of candidates for study thus far has been severely limited because of the large crystals necessary for neutron-diffraction investigations. In extreme cases, experimentalists have been forced to fabricate a crystal of required volume by stacking smaller crystallites with all the troubles of orienting and supporting them. Higher-intensity neutron sources would permit smaller and much more readily obtainable crystals to be used with concurrent reduction in effects due to extinction, absorption, and multiple reflection. Also the high-intensity source, used with convenient cooling equipment, would allow the study of the variation of charge densities in materials such as TTF-TCNQ, which exhibit temperature-dependent conduction. Such measurements coupled with density-of-state measurements by spectroscopic techniques may be of great importance in promoting our understanding of these solid-state properties.

Extinction Theories for primary and secondary extinction have seen considerable improvement in recent years, and extinction effects on measured intensities are predicted with reasonable accuracy unless the effect of extinction is severe. The term extinction is used to describe the effect on Bragg intensities of multiple scattering within the crystal, and the theories predict a specific wavelength dependence for the magnitude of this effect. However, it should be noted that such treatments are based on mosaic crystal models rather than a more realistic description based on crystal dislocations. In addition, the diffraction in mosaic blocks is described by intensity rather than amplitude coupling. An improved model recently proposed involves a continuous transition between the dynamical and kinematic treatments and thus allows for partial coherence in the diffraction for small misorientation in adjacent parts of the sample. One of the best tests of such theories is by careful examination of the predicted versus observed wavelength dependence. Such studies are much more readily and comprehensively carried out by use of white neutron and TOF facilities. An intense pulsed neutron source would be ideal for such experiments.

Anomalous Dispersion Certain nuclei (e.g., ^{149}Sm and ^{113}Cd) exhibit resonances in the thermal region, with the accompaniment of anomalous dispersion. These anomalous dispersion effects are markedly wavelength-dependent and are an order of magnitude larger than those observed in x-ray diffraction. These neutron anomalous dispersion effects have greatest potential for study of biological molecules but can also find use in other specialized inorganic systems.

4.2.3. Conclusions

Usefulness of a High-Intensity Pulsed Source (HFPS)

The scientific prospects in the area of structure determination provide strong justification for an HFPS with a flux of 10^{16} thermal neutrons/cm^2 sec. For example, in classical single-crystal structure determination, an HFPS with TOF analysis and area position sensitive detectors promise data collection rates an order of magnitude over that possible with the highest-flux steady-state reactors. (As a practical matter, however, considerable instrumentation development will be required before this goal can be achieved.) In addition, new areas such as surface structure can be opened up for thermal neutron research because of the high flux; a more powerful attack on the structure of amorphous materials and liquids becomes possible because of higher energy (epithermal) neutrons; and new types of time-dependent transient studies are possible because the repetitive pulsed nature of the source.

Steady-State Reactor Aspects

At the same time, it is important to maintain first-rate capabilities with existing high-flux reactors because many forefront scientific areas can best be studied with steady-state reactors. For example, small-angle neutron scattering is a new area not yet exploited in the United States with many exciting prospects. Although proposals have been made for coupling small-angle scattering instruments to a pulsed source, the concepts must be tested, and it is not yet clear which type of source would be best for such studies. Also, a similar situation prevails for the study of magnetic structures by pulsed neutrons, because much work remains to be done to develop an efficient polarizer for separating neutron spins.

Instrumentation Development

From the above, it is clear that the quality and sophistication of the instrumentation that surrounds a neutron source ultimately determines the usefulness and power of neutrons as a probe of matter. At various places in this section we have discussed the need for additional instrument facilities such as small-angle scattering spectrometers and high-resolution powder spectrometers and as well for operational improvement in polarizers, monochromators, area detectors, and other instruments. In addition, the successful use of a high-flux pulsed source will be based on the development of completely new forms of instrumentation. Further discussion of these instrumentation needs will be given in Chapter 5.

4.2.4. Summary of Recognized Areas of Structural Research Benefiting or Made Possible by Improved Neutron Sources and Facilities

1. General crystallography where there is sample size limitation.
2. Crystallography of complex systems where a great amount of data collection is necessary.
3. Subtle superlattice formation as with charge-density wave instigation.
4. Fully exploited profile analysis in powder diffractometry.
5. Noncrystalline states of matter, especially glasses, liquids, paramagnets, and amorphous ferromagnets with data of improved resolution or greater extent.
6. Interstitials, imperfections, and defects in solids and their diffusion, including self-interstitials.
7. Disordered crystalline solids and their magnetic analog.
8. Kinetics of solid-state reactions, glass transitions, and other time-dependent phenomena.
9. Structures under unusual and extreme conditions, particularly high pressure and temperature.
10. Structures under pulsed external stress such as extreme pressure or magnetic (electric) field.

11. Transient effects with magnetic and ferroelectric domains.
12. Magnetic phase transitions and critical scattering effects.
13. Surface monolayer structures.
14. Surface reactions as in catalysis.
15. Neutron polarization analysis after magnetic scattering.
16. Diamagnetic currents and atom bonding.
17. Organic free-radical molecules with unbalanced electron spins.

4.3. EXCITATIONS AND FLUCTUATIONS IN CONDENSED MATTER

4.3.1. Introductory Comments

As discussed in the first parts of this report inelastic neutron scattering has had an enormous impact on our understanding of the dynamics and phase transition behavior of solids and liquids. In this section, we shall, first, review the basic principles involved, second, discuss the accomplishments of neutron scattering, especially in the United States to date, and third, explore the probable future areas of emphasis and identify important new problems in condensed-matter science where inelastic neutron scattering could make significant contributions. Finally, we shall identify the current and future needs for facilities and instrumentation in order to guarantee the vitality and continued excellence of inelastic neutron scattering and, to a certain extent, concomitantly solid- and liquid-state science in the United States.

As discussed in Section 4.1, neutrons couple to the nucleus via the nuclear interaction and to the unpaired electron spins via the magnetic dipole interaction. In both cases, the coupling is relatively weak so that the inelastic scattering cross section may be simply calculated in the first Born approximation. Formally, for a monatomic system one obtains for the inelastic nuclear and magnetic coherent-scattering cross sections, respectively, *Nuclear:*

$$\left.\frac{d^2\sigma}{d\Omega d\omega}\right|_{\text{coherent}} = \frac{k'}{k}\,\frac{\sigma_c}{4\pi}\,\frac{1}{2\pi\hbar}$$

$$\int_{-\infty}^{\infty} dt\; e^{-i\omega t} \sum_{j,j'} \left\langle e^{-i\vec{Q} \times \vec{r}_j(o)}\, e^{i\vec{Q} \times \vec{r}_{j'}(t)} \right\rangle, \qquad (4.1)$$

Magnetic unpolarized neutrons:

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k'}{k} \left(\frac{\gamma e^2}{m_e c^2}\right)^2 \left(\frac{g}{2}\, F(\vec{Q})\right)^2$$

$$\sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \hat{Q}^\alpha \hat{Q}^\beta) \times S^{\alpha\beta}(\vec{Q},\omega), \qquad (4.2)$$

where

$$S^{\alpha\beta}(\vec{Q},\omega) = \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} e^{i(\vec{Q} \times \vec{r} - \omega t)} \left\langle S^\alpha_{\vec{o}}(o)\; S^\beta_{\vec{r}}(t) \right\rangle$$

$$= \frac{\hbar}{\pi}\, \frac{1}{g^2 \mu_B{}^2}\, \frac{1}{1 - e^{-\hbar\omega/kT}}\; \text{Im}\, \chi(Q,\omega). \qquad (4.3)$$

The symbols in these equations have their usual meaning.

For the case of the magnetic scattering we have written the cross section both in correlation function language and in linear response form. The formal equivalence of Eqs. (4.2) and (4.3) is known as the fluctuation-dissipation theorem. In words, Eq. (4.2) means that in a neutron-scattering experiment one measures in Fourier transform the probability that a spin at position $\vec{r}$ at some time t is pointing up given that a spin at position $\vec{o}$ at the origin time is also pointing up. Subject to resolution considerations, the experiment yields this information for all $\vec{r}$ and for all t. Such an experiment thus provides complete information about the microscopic dynamics of the spins in a magnetic material. A similar description for the nuclear positions applies to Eq. (4.1). In most cases, neutron scattering is the only experimental technique that can yield this most important microscopic information.

As discussed in Section 4.1, an essential feature, which is at the core of neutron scattering's impact on condensed-matter science, is that neutrons from a nuclear reactor are well matched both in energy and in momentum to thermal excitations in solids and liquids. There are, of course, important exceptions that we shall discuss later in this section, but for now we limit ourselves to thermal phenomena. In general, with neutrons from a nuclear reactor one can readily study vibrational and magnetic excitations with energies varying from 1 to 1000 K and with momenta varying from 0.01 $Å^{-1}$ to 10 $Å^{-1}$, that is thermal excitations over the complete Brillouin zone in a solid are accessible to neutrons. The above ranges may also be extended with special techniques. It is important to note that neutrons, unlike electrons and photons, do not couple to the electron charge, and hence one does not measure charge density fluctuations. Thus, for dynamical phenomena, x-ray and electron scattering are complementary to neutron scattering. It is, however, possible to measure vibrational and magnetic excitations at very long wavelengths using light scattering or infrared techniques. In certain cases then, photon spectroscopy is to be preferred to neutron scattering because of its intrinsic high resolution, although, more often than not, the two techniques reinforce each other. Finally, because of the relative weakness of neutron sources, illuminated volumes, that is, samples of order 1 cm^3 are typically required. Again, however, except for certain isotopes, most materials have penetration lengths of much greater than 1 cm for thermal neutrons so that such studies are possible provided, of course, that appropriate single-crystal samples may be grown. In addition, because neutrons do not couple directly to the charge, one may study insulators, semiconductors, and metals with equal facility.

Excitations in condensed matter may generally be subdivided into two broad categories: (a) those that are localized in energy-momentum (hereafter referred to as ω-Q) space; (b) broad excitations extending over a wide ω range for a given Q and vice versa. Examples of class (a) are phonon and magnon excitations in solids, while vibrational excitations in liquids are typical examples of class (b). The experimental techniques used for studies of sharp and diffuse excitations are generally quite different, the former involving triple-axis measurements and the latter TOF techniques. We should note that, especially in the United States, studies of type (a) are ubiquitous, while those of type (b) are still relatively rare. It is convenient then to subdivide our discussion of dynamics into those best suited to triple-axis and TOF spectrometry, respectively. Finally, because of the Maxwellian pile spectrum, studies of dynamics above 0.15 eV are extremely difficult using a steady-state source. However, new proposed accelerator-based pulsed sources promise abundant fluxes in the epithermal region above 0.15 eV. It is of interest therefore to consider the possible areas of research that these new sources open up.

4.3.2. Triple-Axis Spectrometry

The experimental technique of triple-axis spectrometry is discussed in Section 4.1. In a typical experiment one fixes the momentum transfer $\vec{Q}$ at a particular value and one then varies the spectrometer angular positions so as to scan the energy $\hbar\omega$. A phonon or magnon excitation with energy $\hbar\omega(\vec{Q})$ then manifests itself as a peak in the $\hbar\omega$ spectrum. This method would at first seem to be exceedingly inefficient since one selects a narrow range of the pile spectrum and one only looks at a single-energy transfer at a particular time. This inefficiency, however, is greatly compensated for by the fact that one obtains specific information that may be immediately interpretable. A second and equally important feature of triple-axis spectrometry is that the resolution characteristics of the instrument are understood in great detail. This means first that the effects of the instrumental resolution may often be deconvoluted to obtain the intrinsic profile. Second, one may use well-understood focusing techniques to obtain enhanced resolution.

We may now consider various applications of inelastic neutron scattering using triple-axis techniques. The overwhelming activity in this field has been the mapping out of phonon and magnon dispersion relations in solids. These dispersion curves may generally be unfolded to obtain the interatomic force constants atom by atom (or spin by spin). They may also be used to calculate various thermodynamic properties of the solid. This is by now a mature, well-developed field. Studies of the phonon and magnon dispersion relations in a vast range of insulators, semiconductors, and metals have been reported. We show in Figure 4.7 as a typical example the phonon dispersion relations in $PdD_{0.63}$. It seems that most of the elemental and simple compound solids have now been studied. We may anticipate, however, that important experiments will be performed for the indefinite future. As materials scientists generate new interesting systems, a knowledge of

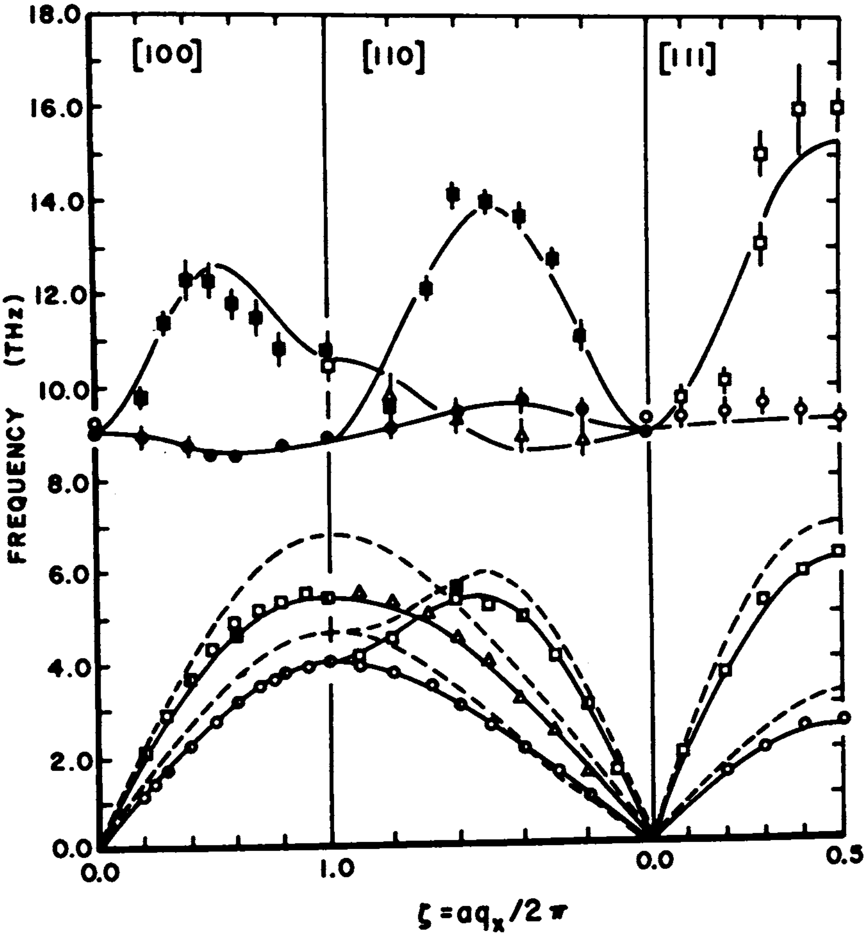


FIGURE 4.7 Phonon dispersion curves in $PdD_{0.63}$. The points represent the measured energies for single phonon excitations propagating in the directions indicated with wave vector [ζ00], [ζζ0], or [ζζζ]. The lower-energy modes represent mainly palladium motion, while the higher-energy modes involve primarily deuterium motion. The solid curves are calculated based on a Born-von Kármán model with 12 parameters. The solid (open) symbols are phonon groups obtained at 150 K (295 K). The dashed curves are the dispersion curves observed for pure Pd at 300 K. Figure taken from J. M. Rowe *et al.*, *Phys. Rev. Lett.* *33*, 1297 (1974).

the basic excitation spectra will always be essential. In current research the major limitation inevitably seems to be the size of single crystals obtainable. Correspondingly, one requires the highest steady-state flux possible (e.g., on the HFBR at Brookhaven or on the HFIR at Oak Ridge). Nevertheless, many such experiments may be performed on intermediate or smaller reactors. We should also note that in these experiments useful information about the excitation eigenfunctions may be obtained from the intensities. We anticipate more emphasis on dynamical intensity information in the future.

Besides conventional magnon and phonon studies, there are also a variety of other experiments on excitations that couple directly to neutrons and are therefore amenable to neutron spectroscopy. These include a wide range of magnetic dipole active excitations. The simplest of these are crystal field and atomic (i.e., fine structure) excitations in solids. An example of the former is the dynamics of double-hexagonal-close-packed Pr, while the $^7F_0$-$^7F_1$ exciton in SmS provides a good example of a highly dispersive atomic transition in a solid. To date very few such experiments have been performed, but we anticipate many more such studies in the future. In cases where the dispersion is appreciable and the energies are less than 100 meV, triple-axis techniques are preferable. For crystal-field studies, in particular, the magnetic excitons have often very little dispersion so that TOF techniques are highly preferable. In addition, there are many important physical systems for which the magnetic excitons are at energies between 0.1 and 1 eV. For these experiments, a new source with abundant epithermal neutrons is required. Finally, we should emphasize that in metals neutrons provide the only realistic probe of magnetic exciton dynamics.

In all the above cases, important changes often occur in the dynamics as a result of some external parameter such as temperature, pressure, or magnetic or electric field. For example, many structural phase transitions are driven by the softening of a phonon branch as a function of temperature. With advances in materials science a remarkable variety of exotic transitions have been discovered; recent examples of special interest include the one-dimensional conductors KCP and TTF-TCNQ and the two-dimensional charge-density wave systems such as $NbSe_2$. In all cases, neutron scattering either has played or promises to play a central role in sorting out the microscopic interactions. We may anticipate an indefinite number of such experiments in the future. The pressure variable has been exploited less fully than temperature. However, new triple-axis techniques have made possible the study of dynamics as a function of both pressure and temperature from 1 to 40,000 atm and from 4.2 K to room temperature. Here the principal limitation is sample size, ~0.25 cc, so that abundant fluxes are essential. We should also note that in the structural phase-transition problem a high-intensity, ultra-high-resolution spectrometer would be invaluable in enabling one to sort out the very-low-frequency response.

Another area of research that has recently come to full fruition is the dynamics of disordered systems. Disordered materials may be broadly subdivided into two classes, amorphous systems, that is, those without lattice periodicity, and site-random crystalline systems. For the former, TOF techniques are generally preferable so we postpone the discussion of them until the next subsection. Disordered materials are, of course, ubiquitous in nature. They span a vast range of possibilities including magnetic alloys such as $Rb_2Mn_{1-x}Mg_xF_4$, alloys such as $K_{0.9}(NH_4)_{0.1}Cl$ of interest for lattice dynamical reasons, and defect systems such as split interstitials in single-crystal Cu. Such systems are of considerable scientific interest, and, in addition, they are often technologically important. A reasonable number of experiments on the lattice dynamics and magnetic excitations in concentrated alloys has been reported. For the latter, theory and experiment are generally in good accord (see Figure 4.8), while for the former there are still substantial disagreements in many systems. High-resolution measurements on model systems seem to be essential. Studies of the dynamics of very dilute defects are still in their infancy. This area promises to develop into a major thrust in materials science. Again very high resolution, which in turn necessitates the highest possible steady-state flux, is required. In general, neutron scattering has played an important role in studies of the lattice dynamics and magnetic excitations in alloys and defect systems. One can anticipate a continued expansion of this class of research for the indefinite future.

As mentioned previously neutrons have made a variety of important contributions to metals physics. The most straightforward of these involve measurements of phonon and magnon dispersion curves, which, in turn, have acted as stimuli for the development of pseudopotential theory and in general for theories of the conduction electron wave-vector-dependent susceptibility. Recent work has concentrated on high-resolution measurements of energies

NEUTRON SCATTERING FROM $Rb_2Mn_{0.54}Mg_{0.46}F_4$ AT 4.0 K AS COMPARED WITH COMPUTER SIMULATIONS

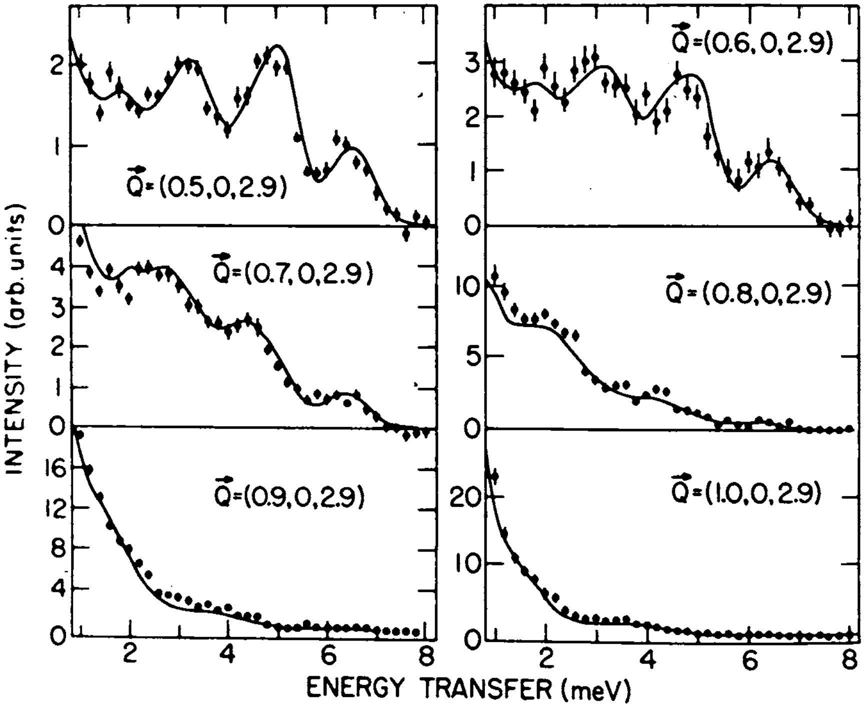


FIGURE 4.8 Dynamical response $S^t(\vec{Q},\omega)$ as a function of wave vector in the two-dimensional dilute antiferromagnet $Rb_2Mn_{0.54}Mg_{0.46}F_4$. As the wave vector component Q_x changes from 0.5 to 0.1, one is going from the magnetic Brillouin zone boundary to the zone center, that is, from very short to very long wavelengths. The individual peaks that are resolved at short wavelengths correspond to cluster modes in which one excites a Mn^{2+} spin in the presence of 4, 3, 2, and 1 nearest neighbors. The solid lines are computer calculations based on classical spin wave theory by R. Alben and M. Thorpe. Figure taken from R. A. Cowley *et al.*, Phys. Rev. B *15*, 4292 (1977).

and linewidths to elucidate Fermi-surface and electron-phonon effects. Particularly dramatic effects have been observed in the superconductors Nb_3Sn and Nb around T_C for phonon energies near the superconducting gap. In theory, high-resolution measurements of phonon lifetimes and energies (especially near Kohn anomalies) could yield important information on Fermi surfaces in metals and alloys, on the electron-phonon interaction in superconductors, and on the temperature dependence and anisotropy of the gap energy in superconductors. Typical results in such systems are shown in Figure 4.9. However, for

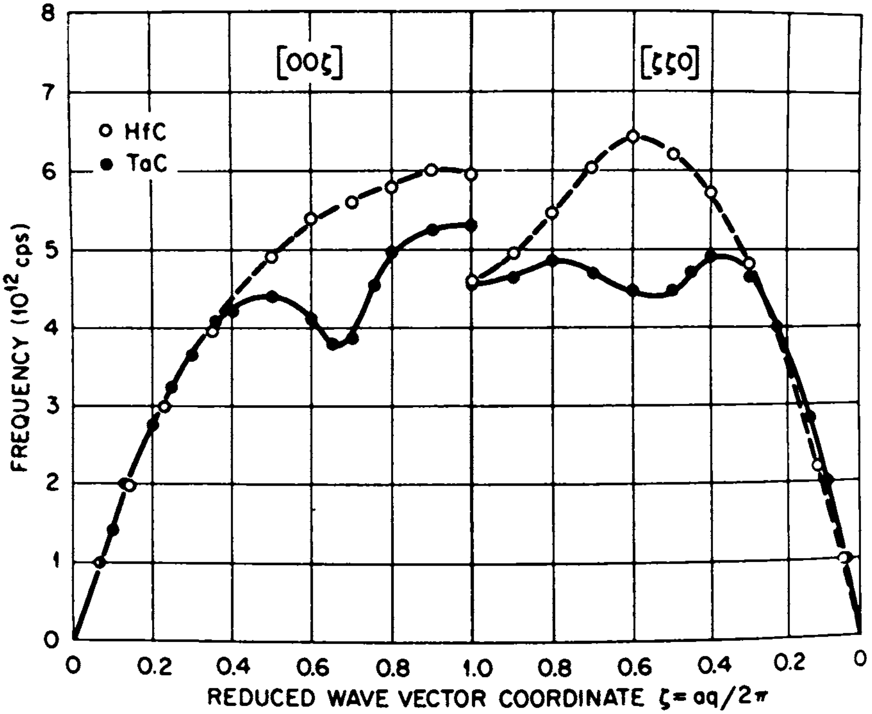


FIGURE 4.9 Longitudinal acoustic modes in TaC and HfC. TaC is a high-temperature superconductor, whereas HfC is nonsuperconducting down to 1 K. A marked difference in the phonon dispersion curves is observed at intermediate wave vectors indicating unusual electron-phonon effects in the high T_C material. Figure taken from H. G. Smith and W. Glaser, *Phys. Rev. Lett. 25*, 1611 (1970).

this to develop into a general technique, an order-of-magnitude increase in source intensity is required.

Another area in which neutrons continue to play a central role is in the dynamics of quantum fluids. Indeed one of the earliest and still one of the most dramatic contributions of neutron scattering has been the mapping out of the phonon-roton spectrum in superfluid ^{4}He. The measured dispersion curve is shown in Figure 4.10. More recent work has concentrated on ^{4}He films, on ^{3}He-^{4}He mixtures, and on ^{3}He itself. These experiments may be done effectively with both triple-axis and TOF techniques. We may anticipate interesting work on quantum fluids (perhaps including spin-aligned atomic hydrogen!) for the indefinite future. The ^{3}He studies would be greatly simplified by a factor-of-10 improvement in source strength.

Finally, we consider the impact of neutron scattering on studies of phase transitions and critical phenomena. There are, of course, many phase "transformations" that are included in the subjects already discussed. One might note especially martensitic transitions of interest to metallurgists. As we noted previously, this is an on-going area of research in which neutrons provide central metallurgical information. We are interested here, however, primarily in critical phenomena. In terms of the scattering cross-section Eq. (4.3), a second-order transition manifests itself as a divergence in Im $\chi(\vec{Q},\omega)$ at particular points in Q space as $\omega \to 0$. The essential feature of such a phase transition is the correlation length, ξ, which diverges at the phase-transition temperature T_C. Scattering spectroscopy in general is uniquely important in such studies since it provides a direct experimental measure of ξ. Neutron scattering is particularly useful in studies of magnetic, order-disorder, and structural phase transitions.

The last 10 years has seen significant progress in our understanding of the theory of critical phenomena at ordinary second-order transitions; indeed, it may now be said that this is a "solved problem." Neutron-scattering studies of the static and dynamic fluctuations in "model" systems such as $RbMnF_3$ have played a central role in this development. Attention now is shifting toward more complicated multicritical phase transitions. For example, at a tricritical point in a magnet both the ferromagnetic and antiferromagnetic order parameters behave critically. Because of the wide range of wave vectors available, neutrons can provide information *in situ* on both the ferromagnetic and antiferromagnetic fluctuations and order

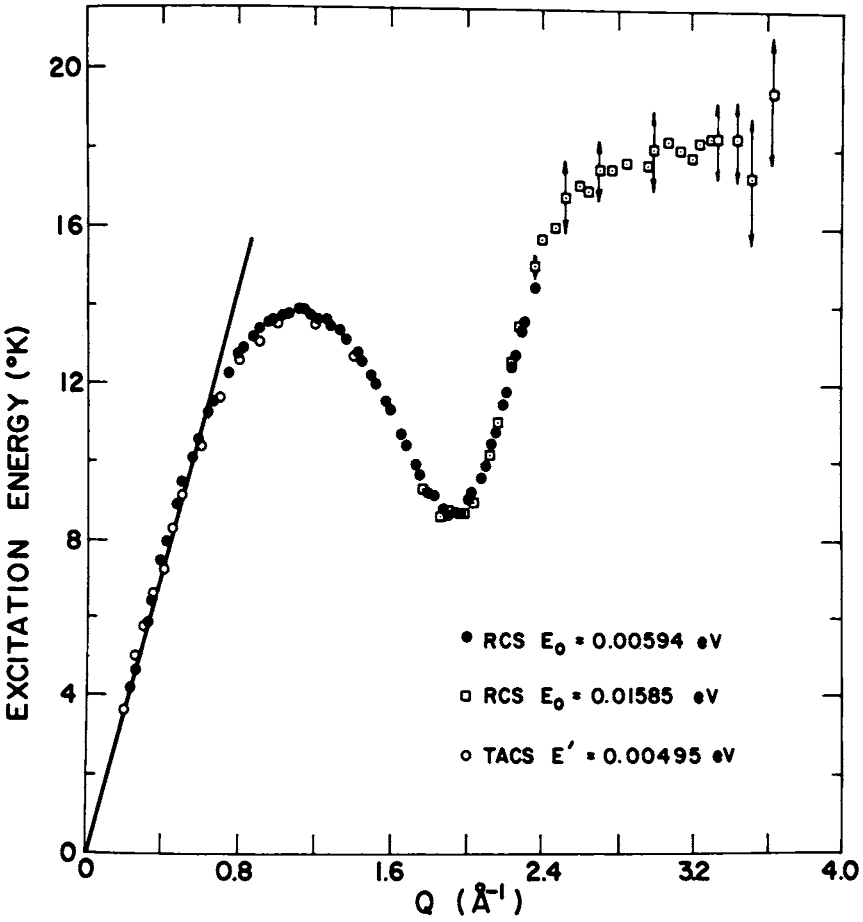


FIGURE 4.10 The experimental results for the energies of the one-phonon excitations at 1.1 K in liquid ^{4}He at 1 atm. These results were obtained using both triple-axis (TACS) and time-of-flight (RCS) neutron-scattering techniques. Figure from R. A. Cowley and A. D. B. Woods, *Can. J. Phys.* *49*, 177 (1971).

parameters. Another example is the critical behavior near the percolation concentration in dilute magnetic alloys. Detailed studies of the statics and dynamics of such systems clearly are, and will continue to be, of prime importance for the continued development of our understanding of phase transitions. In general, the essential requirement is very high-q resolution at small angles around Bragg peaks together with very good energy resolution. These requirements call for abundant sources of low-energy neutrons together with appropriate instrumentation.

In this subsection we have given a variety of examples of the applications of triple-axis neutron spectrometry. For purposes of conciseness we have omitted some subjects such as paramagnetic spin dynamics especially in lower-dimensional materials, surface phonons, mixed excitations such as plasmon-phonon and quadrupole exciton-phonon modes, valence fluctuations, and hydrogen tunneling modes, each of which is amenable to triple-axis techniques. Some of these will be discussed in the next subsection. Indeed there are many problems that are best studied via both triple-axis and TOF techniques, the former to give precise information localized in ω-Q space and the latter to give a broad overview.

It is clear, in general, that triple-axis techniques can give detailed information about both mass density and spin-density fluctuations in solids and that these in turn play a role in virtually all aspects of solid-state and materials science. This field has now reached a reasonable level of maturity, and it is possible to make realistic conjectures about future directions of research. Most of the simplest experiments have been done, and the evolution is now toward studies of more exotic materials and more complicated physical phenomena. In order to sustain traditional dominance of the United States in this area and for the health of American solid-state and materials science as a whole we strongly recommend the following:

(a) All the best existing facilities should be maintained and, if possible, the community of users should be broadened;

(b) Good support facilities should be provided including especially ancillary equipment for high and low temperatures, high pressures, and high magnetic and electric fields;

(c) Many of the next-generation experiments require abundant low-energy neutrons, thus vigorous support should be given to the development and installation of cold sources;

(d) Development of ultra-high-resolution spectrometers either of the backscattering or Mezei type is a necessity;

(e) Development of better polarizers for all energies and also of improved monochromaters for higher-energy neutrons (E > 40 meV) would facilitate a large number of new experiments.

4.3.3. Time-of-Flight (TOF) Spectrometry

In triple-axis spectrometry the energy analysis is done via the wave aspects of the neutron, that is, by Bragg scattering. This by necessity is highly inefficient since in a typical scan one looks at a succession of isolated points in ω-Q space. As we discussed in the previous subsection, in a large number of problems this inefficiency is completely compensated for by the immediate physical interpretability of the results and the fact that one often is only interested in quite focused information about the system being studied.

There are, however, equally many physical systems for which the converse is true. For example, for isotropic systems only the modulus of the momentum transfer $|Q|$ is relevant. Furthermore, the response function in ω-space may be quite diffuse, as, for example, is generally the case for liquids. For this class of experiments, TOF neutron spectroscopy is usually more efficient, and indeed in certain cases it may yield one to two orders-of-magnitude improvement in useful intensity.

A brief discussion of TOF spectroscopy has been given in Section 4.1. In this subsection we confine ourselves to a discussion of the various dynamical physical problems that are amenable to TOF techniques. It should be noted that in the United States triple-axis spectrometry has been developed to a high art form, whereas TOF experiments have been rather less extensive. Thus, by necessity, many of the areas of research to be discussed here are less well explored and concomitantly more speculative. We consider first experiments involving energy transfers less than about 150 meV.

Energy <150 meV

Quite generally, TOF techniques are especially useful for dispersionless excitations. In that case, one may add the spectra from many banks of counters. A simple example is that of crystal-field spectroscopy. For all the non-S-state rare earths as well as certain transition metal ions including Fe^{3+} and Co^{2+}, the crystal field lifts the degeneracy of the lowest Hund's rule multiplet, giving a set of energy levels with typical separations of 10-100 meV. The lowest-lying excitations in such a system may have appreciable dispersion, but in nearly all cases the higher-lying levels are flat. The spectroscopy of crystal-field states in insulators is, of course, an ancient and well-respected subdiscipline of optical spectroscopy. However, even in insulators neutrons may provide valuable complementary information since they pick out uniquely the magnetic-dipole active transitions. Further, because the coupling of the neutrons to the metal atom is well understood, the transition intensities may be used to obtain eigenfunction information. For crystal-field effects in metals, neutrons provide the only direct spectroscopic probe. The field of neutron crystal-field spectroscopy is still in its early stages; indeed the current theoretical models for crystal fields in metals are quite primitive. Thus we might expect much more research in this area in the future. There are a variety of other magnetic excitations of this sort including atomic fine-structure splittings, pair spectra, and singlet-triplet excitons where neutron TOF techniques might be expected to provide important spectroscopic information.

In certain rare earth compounds and metals the magnetic electrons fluctuate between distinct localized and itinerant states. This fluctuation manifests itself in a severely broadened spin-spin correlation function. This, in turn, means that the response functions Im $\chi(\vec{Q},\omega)$ is extended in ω-Q space. Initial experiments, for example in Ce_xTh_{1-x}, demonstrate that neutrons can provide a direct map of Im $\chi(\vec{Q},\omega)$ in these systems. Results for $Ce_{0.74}Th_{0.26}$ are shown in Figure 4.11. The characteristic energies are typically in the range of 10-100 meV. It is anticipated that similar effects will be observed in actinide metals. This area of research is in its infancy, but certainly the initial experiments indicate that neutrons, and especially neutron TOF techniques, will provide essential microscopic information.

A third area of research that is rapidly developing and in which neutrons will undoubtedly play a major role is that of the dynamics of amorphous materials. From the high-Q spectra one obtains a weighted phonon density of states that can be compared with microscopic theoretical models. Similar information is, of course, obtained using optical techniques in insulators and in semiconductors. However, neutrons are the only feasible probe for glassy metals. There is also considerable current scientific and technological interest in amorphous metallic ferromagnets.

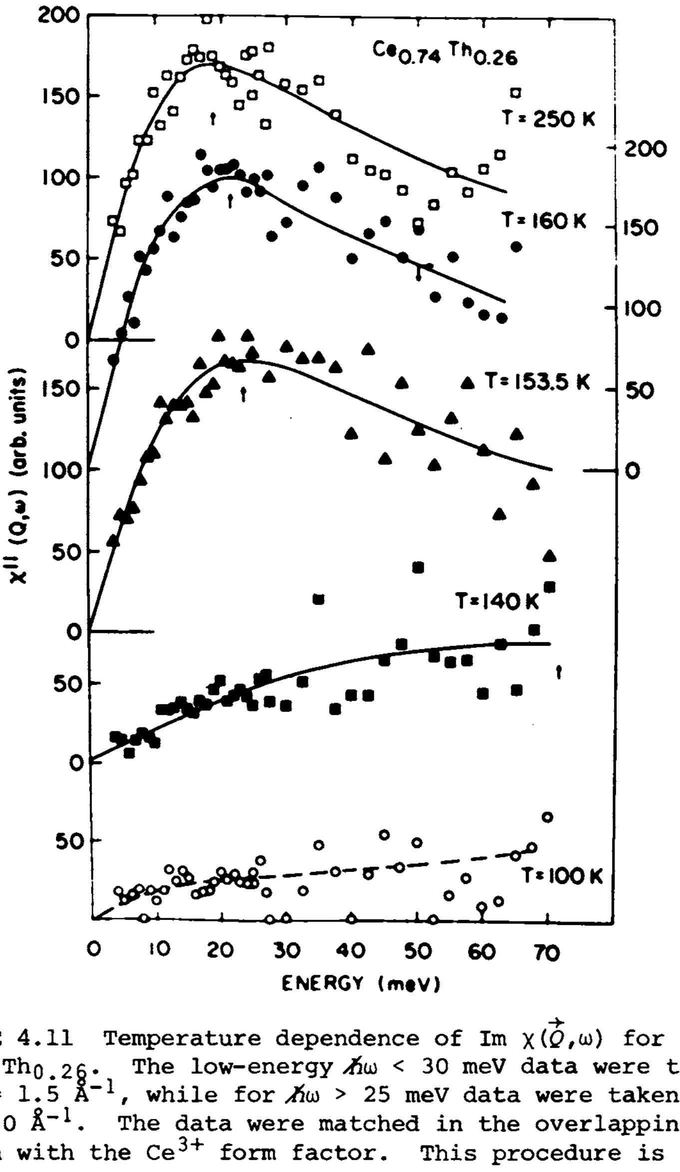


FIGURE 4.11 Temperature dependence of Im $\chi(\vec{Q},\omega)$ for $Ce_{0.74}Th_{0.26}$. The low-energy $\hbar\omega$ < 30 meV data were taken at Q = 1.5 $Å^{-1}$, while for $\hbar\omega$ > 25 meV data were taken at Q = 3.0 $Å^{-1}$. The data were matched in the overlapping region with the Ce^{3+} form factor. This procedure is justified by the fact that the shape of Im χ is Q-independent in this Q region. The solid lines are fits of the data to a Lorentzian response. Note that in this type of system the spin dynamics are spread over a wide range of energy and are at best weakly Q-dependent. Figure taken from S. M. Shapiro *et al.*, *Phys. Rev. B 16*, 2225 (1977).

Initial triple-axis experiments at Brookhaven, NBS, and Oak Ridge have yielded interesting results in such systems. In particular, in certain cases, one observes remarkably well-defined spin waves at long wavelengths. Currently, such experiments are limited to either small Q or very large Q because of kinematical considerations. An intense epithermal source should open up the entire Q spectrum. In general, one can expect that a combination of triple-axis and TOF spectrometry will be best suited for such studies.

In analogy with the above large-Q experiments, one can reasonably expect that with an intense pulsed source one may easily carry out broad surveys of the frequency spectra of solids and alloys including fine particles and more complicated organic systems and including the pressure, temperature, and concentration dependences of their frequency spectra. In the main, these at present require prohibitive amounts of time to obtain even reasonable statistical accuracy and good resolution. In spite of the fact that neutrons are not a surface-specific probe, recent neutron-scattering studies have provided the first determination, by any technique, of the collective dynamics of monolayer films. This area is just in its infancy, but it is clear that there are many fruitful possibilities, especially for studying hydrocarbon dynamics on various surfaces including catalysts. Here, TOF techniques and an intense neutron source will be of importance in extending the range of samples amenable to the technique.

A principal application of TOF spectroscopy to date has been the study of the dynamics of fluids. The classic example is, of course, the phonons and rotons in liquid ^{4}He. One might mention in addition the work at Argonne on rubidium, argon, and ^{3}He. We show in Figure 4.12, $S(Q,\omega)$ for liquid argon.

The very small-Q region ($Q \lesssim 10^{-5}$ Å^{-1}) in liquids is most accurately probed by laser light scattering. The dynamical response of simple fluids in this hydrodynamic region is now well understood. For larger Q's, neutrons represent the only possible probe. In general, neutron-scattering research in liquids is in its early stages of development. Only the simplest fluids have been characterized; further, the data do not cover the important crossover regime from hydrodynamic to high-Q behavior. This requires neutrons of very high energy combined with a spectrometer with very good resolution. There has also been very little work on molecular liquids. Finally, there is need for experiments in the vicinity of the

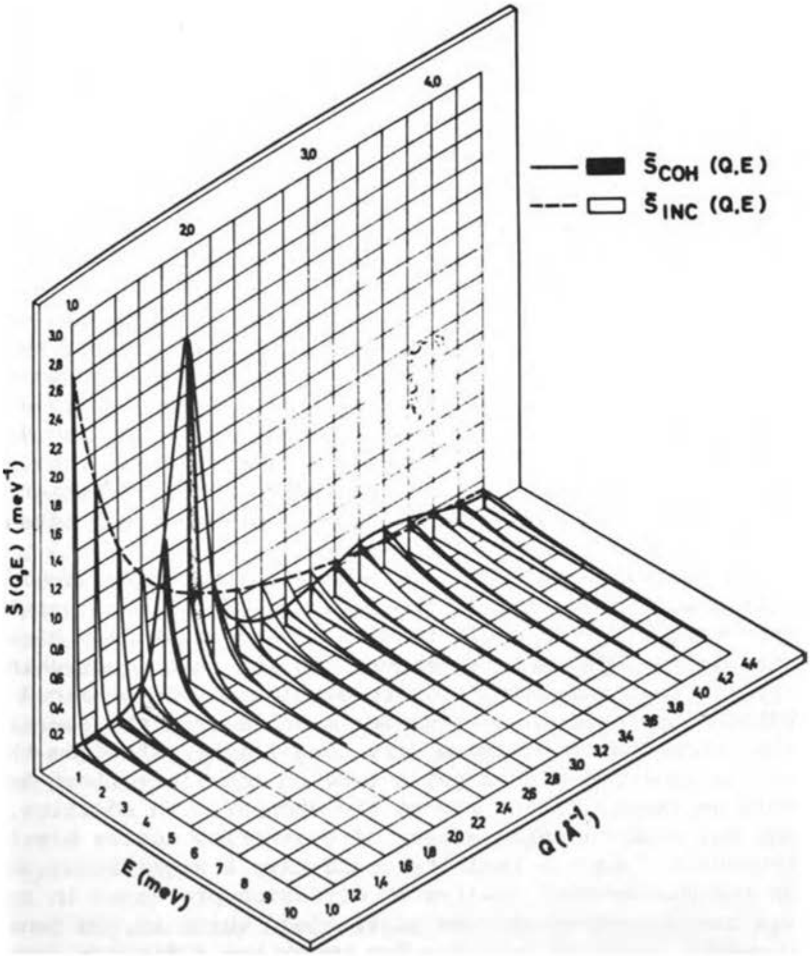


FIGURE 4.12 Smooth-scattering functions shown versus *Q* and *E* for liquid argon. The dashed line and the solid line show the envelope of the incoherent- and coherent-scattering functions, respectively. Figure taken from K. Sköld *et al., Phys. Rev. A6*, 1107 (1972).

gas-liquid critical point, for studies of the dynamics of H_2O and D_2O, for studies of the structure factor in liquid metals and alloys, and for measurements of $S(Q,\omega)$ in molten salts. Clearly, high-quality experiments on liquids are difficult. Further, the experimental results are rarely amenable to direct theoretical interpretation. Indeed, most successful interpretations to date have involved comparisons with molecular-dynamics calculations as opposed to any first-principles theory. In spite of the extreme difficulty of this problem, one can reasonably anticipate that detailed TOF neutron studies using a high-intensity source should lead to major advances in liquid physics. Further, this is clearly an important area of research that warrants a concerted effort.

Another area of research in which neutrons have begun to make important contributions is that of hydrogen in metals. The study of the structure and dynamics of hydrogen in metals is related to several important technological areas, including the development of new systems for energy storage and production and the effects of hydrogen embrittlement in metallic materials and structures. While a large number of incoherent-scattering studies of the dynamics of metal hydrides have already been made, the development of more intense neutron sources and better instrumentation holds great promise for important advances in this field.

An exciting aspect of an intense pulsed source equipped with a cold moderator is the prospect of inelastic spectrometers with microvolt resolution and relatively high intensity. With TOF techniques (unlike techniques using crystal spectrometers) high resolution may be obtained without appreciable loss of solid angle from the source by the procedure of having a very long flight path from the source equipped with a guide tube to totally reflect the cold neutrons all the way to the chopper. In addition, one may study a large number of scattering angles simultaneously. Such a facility could have a significant impact on the microscopic studies of diffusion processes in solids via quasi-elastic neutron scattering, which is, in general, currently limited to cases for which the diffusion constant is $\gtrsim 10^{-4}$ cm^2/sec and hence for which certain crucial temperature ranges are inaccessible. At present, the backscattering microvolt spectrometer at ILL is one of the few facilities where such studies can be carried out, although present data-gathering rates are very low. An example of such studies is the study of hydrogen diffusion in Nb metal and Nb doped with nitrogen impurities (carried out at ILL), which revealed the hydrogen-trapping effects of impurities in limiting diffusion. In addition, neutrons are the only known probe for studying diffusive dynamics at finite wave vector, thus providing a crucial test for our microscopic understanding of diffusion in solids and in particular in solid electrolyte materials. Much higher-resolution quasi-elastic scattering studies of hydrogen diffusion as a function of concentration and hydride phase should provide a more detailed understanding of diffusion mechanisms in metals and alloys, including a test of the lattice-gas and lattice-liquid models for solute hydride phases. Neutrons may be used also to study diffusion of molecules in submonolayer films on surfaces, a field that has just begun to be explored. It should be noted that because of grain-size limitations of most such surfaces, macroscopic techniques are not applicable and neutrons provide the only method of studying the true diffusive processes in these systems.

In anticipation of our next section we note that the availability of hot sources at steady-state reactors and new high-intensity pulsed-neutron sources will make possible the high-resolution study of hydrogen vibrations in metals, including the observation of several levels above the ground state. The derived level widths and splittings should provide significant information on the metal-hydrogen potentials in a variety of metals and alloys. Moreover, the availability of high fluxes in the 150-500 meV region will allow the detailed vibrational analysis of very low concentrations of hydrogen in metals, yielding unique information, for example, on hydrogen clustering effects as a function of concentration in these systems. Similar advances in detailed *coherent* inelastic scattering studies of the lattice dynamics of hydride single crystals at high neutron energies are also anticipated.

Energy >150 meV

One of the exciting features of the proposed accelerator-based pulsed sources is that they will produce abundant amounts of epithermal neutrons because of the $1/E$ rather than Maxwellian high-energy tail. We have already discussed a variety of problems such as dynamics in liquids and dynamics in amorphous magnets for which neutron kinematical limitations inhibit studies at small Q. Clearly, many of these experiments would be much easier using a pulsed source. There are, in addition, a wide variety of important problems in condensed-matter science for which the characteristic energies for mass-density or spin-density fluctuations are above 150 meV. This is especially the case for magnetic excitations.

The spin dynamics of low-lying excitations, particularly in localized moment magnetic systems, is by now fairly well studied by neutron spectroscopy. Similar studies for the so-called "mixed-valence" or "configuration fluctuation" systems (which represent an interesting borderline between the localized moment and itinerant electron systems) are just beginning, as mentioned earlier. In particular, it is expected that much will be learned about the relative importance of *f*-electron-conduction electron mixing and strong intra-atomic correlations from such studies. We should point out that the physics involved here is at the heart of a number of central problems in solid-state physics, such as the Kondo effect. One problem with such systems experimentally has been the large energy range over which the spin dynamics extends. Projections are, however, that such experiments could be easily realizable with a pulsed source of epithermal neutrons. The actinide metals represent an even more extreme case, requiring large energy-transfer studies, and currently there have been little or no spin excitations observed in such systems. Thus, the pulsed source could play a key role in opening up one of the last "frontier" fields of basic magnetism. Even the itinerant magnetism of the transition metals is a problem of fundamental importance that is still unsolved, for which the pulsed source offers the best hope of a breakthrough in our understanding of the electron spin dynamics in the range of 0.1 to 1 eV, such as the Stoner excitations or higher-lying collective modes currently predicted by several theories.

There is a wide variety of other magnetic excitations with energies of the order of 1 eV that should also be amenable to neutron spectroscopy. Among these we might mention transitions between the "atomic" 4*f* *J*-multiplets in rare-earth metals. It is difficult to assess the importance of the physics associated with such studies in the absence of any existing experimental results. Inevitably, however, nature will have many surprises in store for us.

We have already mentioned applications to studies of the vibrational modes of hydrogen in metals. Quite generally, neutrons may be applied to the problem of molecular vibration in each of gases, liquids, and solids. For the former two states, Doppler shift effects broaden the spectra, but nevertheless useful information may be obtained. In solids, recoil broadening effects are minimized so that many systems are open for high-resolution studies. Typical areas of importance are polymers, hydrogen-bonded solids, and organic molecular solids. It should be emphasized, however, that in this area of research neutrons are an ancillary tool compared with optical spectroscopic methods.

A completely different kind of spectroscopy that an epithermal source opens up is that of neutron Compton scattering. Such experiments should yield directly atomic, as opposed to electronic, momentum distributions in solids. These experiments require neutron momentum transfers of 100 $\mathring{A}^{-1}$ or greater. From order-of-magnitude considerations it seems clear that these experiments will be straightforward for helium. More importantly, it should be possible to use this technique to study proton momentum distributions in hydrogen-bonded solids and in metal hydrides. A resonance detector spectrometer utilizing ^{238}U resonance at 6.07 eV with 25-meV resolution may be used for such experiments. These experiments could, in general, involve neutron energies up to 100 eV.

It may also be possible to exploit the pulsed nature of TOF spectrometers to enable one to perform "real-time" experiments. In particular, one might study relaxation effects in response to laser excitation or pulsed magnetic or electric fields. This, of course, will depend on the characteristic time scales involved. Certainly physical phenomena with time scales of 0.1 to 1 msec (typical, for example, of phonon-assisted energy transfer times in solids) should be amenable to study.

Finally, we should note that since neutron spectroscopy at energies >150 meV represents new terrain, it is highly likely that many exciting experimental ideas will evolve after the initial experiments have been performed.

Conclusions

It is evident from the preceding discussion that there is a wide range of important experiments on excitations and fluctuations in solids and liquids that are ideally suited to TOF spectroscopy. Further, such experiments will involve a much wider range of scientists than those currently utilizing triple-axis techniques. We note, in particular, that there are numerous opportunities in chemical spectroscopy as illustrated by the ILL (Grenoble) experience (see Chapter 6). Many of the experiments could be done on TOF systems at the high- and medium-flux reactors. We urge, therefore, that there be continual development of TOF instrumentation at the centers and that the facilities become more user-oriented.

The most dramatic improvements in TOF spectroscopy are possible by going to an accelerator-based pulsed-spallation source. Peak fluxes of 10^{16} neutrons/cm^2 sec with a desirable duty cycle appear to be feasible. This marked increase in usable flux can be exploited in a variety of fashions. First, many experiments that are currently sample-size-limited will become feasible. Second, studies under extreme conditions such as ultra-high pressures will be greatly facilitated. Third, the much higher intensities will enable one to achieve much higher resolutions in many cases. The spallation sources also give abundant fluxes in the epithermal region; thus a new generation of exciting experiments will be possible.

4.3.4. Major Thrusts in Future Inelastic Neutron Research, Especially Those Facilitated or Made Possible by a High-Intensity Pulsed Source

1. Studies of the excitations and phase transition behavior of exotic materials such as lower-dimensional magnets and conductors and solid-state electrolytes.
2. Studies of matter under extreme conditions including ultra-high pressures, temperatures, and magnetic fields.
3. Very-low-energy dynamical processes such as diffusion in varied materials, central peak phenomena at structural phase transitions, and critical dynamics of magnetic phase transitions. These experiments would be facilitated by an ultra-high-resolution spectrometer.
4. Dynamics of amorphous materials including ferromagnetic metallic glasses, amorphous semiconductors, and conventional glasses.
5. Dynamics of liquids including quantum fluids, liquid metals, molten salts, molecular liquids, and liquid crystals.
6. Dynamics of atoms and molecules absorbed onto surfaces, especially hydrocarbons on catalytic metal surfaces.
7. Magnetic excitations at high energies including valence fluctuations in rare earth and actinide metals, and single particle excitations in transition metals.

8. Neutron Compton scattering, that is, studies of the ground-state momentum distributions of light atoms in condensed matter.
9. Structure and dynamics of hydrogen in metals.
10. Dynamics of very dilute defects.
11. Various aspects of metals physics including the mapping out of Fermi surfaces in alloys via Kohn anomalies and studies of the electron-phonon interaction in normal and superconducting metals.

4.4. BIOLOGY AND POLYMERS

Neutron scattering is an important technique for the analysis of biological and polymer structures since it permits the elucidation of structural details not attainable by other presently available techniques. Basically, neutron scattering is often preferable to x-ray scattering because of the higher contrast the sample constituents present to neutrons, facilitating, therefore, the identification of structural landmarks in chemical terms. The relative contrast of sample constituents can further be significantly increased by specific deuterations utilizing the large coherent-scattering factor difference between hydrogen isotopes.

4.4.1. Biology

Biological structures analyzed by scattering techniques are best divided into three experimentally different groups depending on the samples, degree of order, and other factors: (1) nonordered systems such as proteins or protein complexes in solution; (2) partially ordered systems like muscle, collagen, and membranes; and (3) crystalline samples such as protein crystals.

Solution Scattering

Small-angle scattering from biological complexes in solution permits the determination of their overall shape and size, as well as the organization of the various constituents. Such studies are particularly desirable since many macromolecular complexes in solution are in a form most closely resembling their functional state in living organisms.

The most powerful applications of small-angle neutron-scattering techniques rely on the use of specific deuteration to produce regions of high contrast with known chemical composition. Contrast variations are obtained by specific deuteration of constituents and variations of solvent scattering density by simply mixing H_2O with D_2O. Such contrast variation of the solvent amplifies the information that can be obtained from radius-of-gyration measurements. Internal density fluctuations arising from chemical differences can be explored; for example, a two-component system can be characterized with respect to the radius of gyration of each component and the separation of their centers of mass.

"Extended scattering curve" analyses attempt to exploit the scattering beyond the Guinier region (i.e., region where radii of gyration are measured) to extract more detailed shape information. The theoretical treatments for this form of analysis have not yet been fully developed.

"Label triangulation" is an important and exciting extension of the classical solution scattering techniques that allow only the determination of the solute shape and size. If two subunits of a multiunit complex can be deuterated, the interference term in the scattering curve can be analyzed to give the distance separating their centers of mass. One can then proceed to measure different pair distances until the three-dimensional organization of the complex is obtained by triangulation.

This type of analysis will give structural information on a molecular scale and presents an important link between the atomic resolution of the crystallographer and the observation of the microscopist without using preparative techniques that distort the structure by heavy metal staining or vacuum preparation techniques. In this regard, neutron-scattering techniques are able to provide structural information not attainable by other techniques now available.

Present Status Label triangulation is being applied to the structure of the ribosome and DNA-dependent RNA polymerase complexes of *E. coli*. These are the focal machinery of genetic expression, and their structures have not been approachable using conventional methods. Success in the future is assured by the present status of these studies, and many other structures are appropriate for this type of analysis.

Radius-of-gyration measurement and contrast variation have been used to characterize the size, shape, and internal structural features of proteins, ribosomes, lipoproteins, and nucleosomes. While the information obtained is less detailed than in label triangulation, perspectives have been gained that will be most useful.

Ordered Systems

Many biological systems, such as membranes, collagen, viruses, and muscle, exhibit a paracrystalline organization ideally suited for diffraction studies. The investigation of these structures by neutron-diffraction techniques is advantageous as compared with corresponding x-ray techniques because of the higher inherent contrast of such structures for neutrons and the ability to enhance this contrast by studying such structures in environments of variable contrast, e.g., aqueous media of variable H_2O/D_2O ratios. The full power of neutron techniques is achieved utilizing specifically isotopically labeled (e.g., H versus D) molecular components and submolecular portions thereof. In this manner, the structural organization of these systems can be determined to the submolecular and even atomic level, which cannot be achieved using x-ray techniques.

Present Status A number of studies on lamellar membrane systems, such as nerve myelin, retinal rods, and the purple membrane, and on lamellar artificial lipid and lipid/protein membranes have demonstrated the utility of neutron scattering to elucidate important structural details of such membranes at the molecular and submolecular levels. Specific deuteration of molecular and submolecular fragments, as well as variation of the solvent contrast, were utilized in these studies. These methods have also been used to study the nature of the interactions of small molecules (such as local anesthetics) and water with certain membranes. In addition, specific isotopic labeling (H versus D) has been utilized in the development of methods for directly phasing the diffraction from such systems.

Protein Crystallography

Neutron protein crystallography is able to determine H atom locations; hydrogen atoms make up nearly half of all atoms in a protein and play a major role in determining the protein's shape and function but cannot be determined by x-ray crystallographic techniques. Apart from locating H atoms, neutron analysis is able to determine the stability of different hydrogen interactions using H/D exchange. Furthermore, the interactions of water with the protein surface can be documented.

While phases for such investigations can be obtained from anomalous scattering of certain isotopes, it is best to carry out a neutron investigation after a fairly good "x-ray" structure is available that can be used to initiate a neutron refinement. Neutron-diffraction investigations of enzymes will be important to elucidate substrate binding and catalytic mechanisms involving proton transfer.

Present Status Two forms of myoglobin have been analyzed, and the locations and long-term exchange of hydrogens have been characterized. A number of bound water molecules have been identified on the surface of the protein. Structural refinement techniques are still being developed to determine density parameters of solvent water.

Neutron Usage during 1976

In the United States, 22 scientists have been directly involved in neutron studies of biological systems with a total of a 15-man-year effort. Apart from the 2-man-year effort at the National Bureau of Standards (sponsored by NBS and NIH), all U.S. efforts on neutron biology have been carried out on two stations at Brookhaven with a 7-man-year scientific effort provided by Brookhaven personnel and a 7-man-year effort provided by the university community. About 20 percent of the effort was expended on instrumentation development, about 20 percent on protein crystallography, 40 percent on solution scattering, and 20 percent on ordered systems.

In Europe, about 80 scientists have been doing biological neutron work with a total effort of approximately 40 man-years. About 80 percent of that effort was provided by university users, using two stations at ILL and one at Harwell. The technical support of the European effort amounts to about 1.5 technicians per scientist, while the

U.S. ratio is closer to 0.5 technician per scientist. While the use of neutron-scattering techniques for structural studies in molecular biology was pioneered at Brookhaven, funding levels have not allowed the full exploitation of this field, and the major thrust has now moved to the European centers, particularly ILL, although the United States still maintains a small qualitative lead.

Future Usage

The use of elastic neutron scattering in molecular biology will proceed along the directions described above, expanding gradually as workers in the field become familiar with these new techniques and as facilities and trained personnel become available to carry out such investigations. The uses of inelastic and quasi-elastic scattering have, however, hardly been investigated, even though they promise a wealth of information on molecular dynamics. It can only be guessed that pioneering efforts in this area will be made, opening up fertile new avenues for investigating living material.

At present, it can be estimated that about 20 percent of the small-angle-scattering community uses neutrons where applicable. It can be expected, therefore, that the present 8-man-year effort will grow to about 40 man-years over the next 5 years, assuming no major increase of funding in structural molecular biology as a whole. In protein crystallography, the growth rate will probably be higher, increasing from the present 2.5-man-year effort to about 20 man-years.

This minimum envisaged growth rate will require at least three versatile small-angle spectrometers and two protein crystallographic stations at high-flux-beam reactors staffed with adequate scientific and technical personnel.

4.4.2. Polymers

Small-angle neutron scattering is a unique tool to investigate conformations of polymers in solution, polymer melts, crystalline systems, and glasses. As for the biological applications, the power of the method is most fully developed in partially deuterated samples, using the large contrast in the coherent-scattering amplitudes between deuterons and protons.

Elastic scattering yields information on osmotic pressure, radius of gyration, and internal molecular distribution functions of macromolecules in both solid and liquid phases. Specific deuteration allows one to investigate the effects of intermolecular interactions on conformations. This is done by deuterating a low concentration of the polymer present. Specific labeling of parts of a chain, e.g., ends, may provide end-to-end distributions, locations of entanglement points in semidilute solutions, and degree of folding in crystalline systems.

A wide variety of polymeric systems may be studied by these techniques. A partial list includes the following:

1. Solutions of flexible polymers, e.g., polystyrene
2. Partially rigid systems, e.g., aromatic polymers, polypeptides
3. Polyelectrolytes, e.g., sulfonated polystyrene, nucleic acids, proteins
4. Copolymers
5. Rubbers
6. Gels
7. Polymer adsorption
8. Polymer mixtures
9. Coupled polymer-mesophase systems
10. Branched polymers

In addition, low-angle neutron scattering provides a unique method to study the response of rubbers, gels, solutions, and solids to external perturbations such as mechanical stresses, flow fields, and electric and magnetic fields. Such studies should lead to a microscopic understanding of plasticity and rubber elasticity, exhibiting the various roles of transient topological entanglements and chemical cross-linking.

Present Status Although the technique is in its infancy, the use of small-angle measurements (SANS) has already resolved several key polymer questions of historical importance. The only measurements of chain dimensions in the solid state have been carried out by SANS. The Flory theory of excluded volume effects of flexible polymer conformations has been essentially substantiated and extended to semidilute solutions. Initial studies have begun yielding preliminary information on copolymers, polyelectrolytes, and gels. Also, pioneering investigations have commenced on deformed gels and rubbers. It must be emphasized that most effort in this field has thus far been made in France and Germany, including work by British and American scientists. However, the NBS and more recently the University of Missouri have initiated construction of small-angle spectrometers for use in polymer science. Scientists at NBS have already examined the conformations of some copolymers.

Future Usage

The application of neutron small-angle scattering to polymer problems holds great promise both from a scientific and technological points of view. An enormous number of crucial experiments remain to be performed on those problems listed above. In particular, the roles played by entanglements and cross-linking in ladder polymers, stars, combs, and other nonlinear systems are not established. Furthermore, chain reorientation induced by static and cyclic stresses in solids should yield vital technological information on solid polymers. Probably the microscopic aspects of rubber elasticity will be elucidated in the near future by SANS. The relationship between polymers and mesomorphic systems such as lyotrophic liquid phases and thermotrophic polypeptide liquid crystals will form a link between the polymer scientists and the biologists.

The possibility of future very intense neutron sources opens the possibility of studying the dynamical properties of polymeric systems, i.e., the diffuse modes in solution and the phonons in the solids. At the moment, the only tool available for solution dynamics is light scattering. However, this technique is limited to a few extremely large molecules of molecular weight greater than 10^6. A very intense neutron source would eliminate this constraint. For example, in rubbers and gels it would be very exciting to be able to probe the dynamical response over a wavelength scale spanning the characteristic distance between cross-linking points. This is now out of the question.

In light of all the possible important experiments, it can be predicted that the U.S. effort on neutron application to polymer science and technology will grow at a fast rate, probably reaching a 100-fold increase over the present effort within 10 years.

4.4.3. Conclusions

Organization of Neutron Facilities

It is certain that initially, and for some considerable time thereafter, most users will be experts in their own field (polymers or biology) but inexperienced in neutron scattering. If results are to be obtained efficiently, it is, therefore, essential that both scientific and technical help be available at the facility. Resident scientists should be able to maintain their own continuing research programs both in neutron scattering and instrumentation development. Suitable arrangement for an equitable division of beam time between resident-scientists and visitors will need to be developed.

In order to accommodate the predicted growth and ensure that growth occurs in a strong scientific mode, a vigorous program should be developed to educate molecular biologists and polymer scientists into the present and possible future capabilities and techniques of neutron scattering. This educational function must be strongly supported by a critical mass of in-house experts through theoretical and practical workshops.

Instrumentation

For most true solution scattering experiments encountered in radii-of-gyration type determination in polymer chemistry or molecular biology, instrumentation is required with resolution of a few tenths of a degree, using wavelengths >4 Å, i.e., $Q \leqslant 0.005$ ($Q = \theta \times 2 \times \pi/\lambda$) and extending to a Q of 0.1. Adequate flux at long wavelength is probably best achieved with a cold moderated beam with either a multilayer monochromator or a velocity selector. For experiments from ordered systems and most "pair" type measurements, the resolution requirements are less and a minimum of 0.01 Q is quite acceptable. To fulfill these resolution requirements, pathlengths of a few meters are sufficient considering that most biological samples are small (typically 2-6 mm in diameter). For adequate peak-to-background ratios, two-dimensional high-efficiency counters are needed particularly for solution scattering. To permit a relatively small flight path, counters with resolution of ~1-3 mm are required. Present position-sensitive ^{3}He counters, either as 2-D or linear counters, are adequate and permit counting rates ~10^5 neutrons/sec with resolutions of 2.5 mm and efficiencies close to 100 percent. To service a variety of experimental conditions requiring an extended *Q* region, the counter should permit equatorial positioning to an angle of at least 40°.

For protein crystallography, low-angle resolution requirements are less. Typically wavelengths of 1 to 3 Å are used with data extending into the backreflecting region. In order to use smaller protein crystals or crystals with larger unit cell sizes (higher molecular weight), large bandwidth, i.e., limited Laue type, diffraction techniques have to be developed. For efficient data collection, large 2-D position-sensitive counters with real-time display and adequate data-handling systems are needed to handle the few hundred thousand reflections required for protein analysis.

Need for Higher Flux

The development of neutron-scattering techniques for the analysis of biological structures and polymers is seriously handicapped by the relatively low flux of present-day sources.

A tenfold increase in effective neutron flux will result in major qualitative and quantitative improvements as tabulated below.

For solution scattering:

- Measurements can be made at lower concentrations, so that concentration effects on the measurement of radius of gyration can be more completely removed.
- Smaller proportions of the total mass of a macromolecular complex can be mapped as units in labeled triangulation experiments.
- Experiments in monomer-dimer association kinetics and molecular dynamics can be considered.
- In general, the interrelationship of sample size, data collection time, and data quality will be greatly improved, permitting a range of experiments that is not now feasible.

For ordered systems:

- Improved statistics for the utilization of direct methods in the phasing of the diffraction data.
- Improved signal for the study of in-plane structure in lamellar systems (i.e., equatorial diffraction).
- Ability to use anomalous scattering for phasing and locating different isotopes, especially metals.

- Ability to use lower isotope concentrations (especially for biosynthetic incorporation), thinner multilayer specimens resulting in decreased mosaic spread yielding better data.

For protein crystallography:

- Weaker diffracting samples can be analyzed permitting either smaller crystals or larger unit cell sizes (higher molecular weights).
- Better data statistics permitting accurate anomalous phasing leading to more accurate structures.
- Ability to study binding of pharmacological agents with smaller molecular weight than now possible.

For polymers:

- Permits dynamic experiments on bulk polymer response with time as a function of applied stress and temperature change.
- Studies of cyclic stress response of technologically important polymers such as polycarbonate.
- Allows preparation of samples with much lower concentration doping, greatly improving sample reliability in partially crystallized systems.
- Quasi-elastic scattering to study diffusivelike motions of chains.
- Sample size can be reduced, simplifying preparation techniques, or data collection time can be reduced, permitting the analysis of a wider range of samples.

New Neutron Sources

The desirability of a substantial increase in effective flux is clearly established by the improvements described above.

The progressive development of pulsed sources does promise a considerable increase in peak source flux. In order to translate this high source flux into effective flux, extensive developments in data acquisition, reduction, and analysis will, however, be required in addition to the development of suitable spectrometers. In addition, one cannot ignore the necessity for the replacement of current high-flux steady-state sources that may have to be retired in the foreseeable future.

Short-Term Facilities and Manpower Requirements

1. A high-resolution small-angle station for polymer chemists needs to be established. Such a facility should be staffed by polymer scientists with adequate technical personnel.
2. Scientific and technical staff at the two existing biology stations at Brookhaven National Laboratory should be increased.
3. A second small-angle station for the biological sciences is required.

4.5. RADIATION DAMAGE

4.5.1. Introduction

The cumulative damage caused by the energetic products of nuclear reactions has been of great practical importance since the earliest days of nuclear technology. The attempts to control these effects, and in particular those due to medium-energy neutrons, have led to the growth of an important area of applied science and at the same time have stimulated efforts leading to greater fundamental understanding of defects in crystalline solids.

"Medium-energy neutrons" are taken in this section to include neutrons with energy as high as several MeV. Typical fast fission reactor neutrons with energies near 1 MeV are therefore included, while the 14-MeV neutrons characteristic of the fusion reaction are excluded. Neutrons of the latter energy are of great interest in the radiation-damage field because of fusion reactor applications. However, we shall not consider this area in this section because useful fluxes of these particles will not be produced by the neutron sources under present consideration.

We first identify the radiation-damage topics that are of current interest with respect to neutrons. Next, a brief review and discussion of presently existing major facilities is carried out. Scientific opportunities and needs are then discussed, and recommendations regarding the establishment of new facilities are made. A number of previous documents were found to be of value and are listed in References 1-5.

4.5.2. Radiation-Damage Phenomena of Current Interest

In this section we attempt to break the large field of radiation damage into the topics that are of most current interest with respect to neutrons. These topics can be classified conveniently under the following three major headings:

A. Basic phenomena associated with neutron irradiation
B. Complex phenomena associated with neutron irradiation
C. Study of radiation-damage type defects by means of neutron scattering

Category A incudes all the elementary phenomena that are basic to radiation damage by neutrons and includes the research areas listed in Table 4.1.

Category B includes the more complex phenomena that may occur under many applied conditions. These phenomena generally involve a number of concurrent basic mechanisms accompanied by complex interactions and are listed in Table 4.2.

Category C includes studies of radiation-damage phenomena by neutron scattering that are listed in Table 4.3.

TABLE 4.1 Basic Phenomena Associated with Neutron Irradiation

1. Elastic, inelastic scattering and reaction cross sections; slowing down of particles; energy distributions of secondary particles
2. Production of defects (vacancies, interstitials, clusters, and electronic defects)
3. Displacement cascade phenomena (depleted zones and replacement collision sequences)
4. Physical properties of the defects (formation energies, migration energies, electrical properties, etc.)
5. Defect interactions (binding energies)
6. Thermal and pressure spikes
7. Radiation annealing
8. Transmutation products and their behavior
9. Interactions of defects with impurity atoms and transmutation products
10. Surface phenomena (sputtering)

TABLE 4.2 Complex Phenomena Associated with Neutron Irradiation

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1. Complex defect structures resulting from high fluences at low and high temperatures (dislocation loops, voids, bubbles, etc.)
2. Redistribution of solute atoms
3. Phase stability
4. Effects of irradiation on mechanical properties (strength, ductility, creep, fatigue, fracture)
5. Complex surface phenomena (blistering)

TABLE 4.3 Study of Radiation-Damage Type Defects by Means of Neutron Scattering

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1. Study of the atomic configuration of defects by diffuse scattering
2. Study of vibrational modes associated with defects (point defects and clusters)
3. Study of spatial distributions of alloying elements
4. Study of the behavior of solute (or alloying) elements that are near the host element in the periodic table
5. Small-angle scattering from clusters, voids, etc.

This category is quite distinct from the previous two, but the Panel believes that several unique opportunities exist in this area and that this category should therefore be included explicitly in any general consideration of low-energy neutrons and radiation damage.

At present, there is an urgent need for further research and progress in the above areas. The development of nuclear-energy technology depends to a great extent on a sophisticated understanding of neutron-type radiation damage and associated phenomena in reactor materials. In addition, valuable additional information about crystal defect properties can be obtained from radiation-damage and neutron-scattering experiments. Such knowledge is of great importance in understanding defect-sensitive phenomena such as diffusion.

4.5.3. Review of Present Radiation-Damage Facilities

Ideally, facilities for radiation-damage studies should include the following:

1. An intense neutron source of well-characterized neutrons
2. Control over the irradiation conditions and flexibility in the scheduling of experiments
3. Adequate and accessible specimen volume
4. Control over specimen temperature and atmosphere
5. Facilities for bringing in signal leads and instrumenting the experiment
6. Low-gamma heating
7. Ability to transfer specimens to another location under fixed conditions of temperature and vacuum.

A selected list of neutron sources that are of particular interest for radiation-damage or scattering studies and that are either currently available, under construction, or potentially available in the United States is given in Table 4.4.

Of the fast reactors, EBR-II is the only fast-flux reactor currently available in the United States. This reactor produces a large fast neutron flux ~(10-20) × 10^{14} neutrons/cm^2 sec ($E > 0.1$ MeV). The reactor was not designed for materials testing, but it has been used extensively for that purpose in LMFBR research. Tests are conducted in hexagonal subassemblies, permitting experiments in a volume about 5.6 cm in diameter over a core length of 34 cm. Only certain positions have capability for instrumentation (e.g., bringing leads in and out), and because of limited space and access, instrumented experimentation is difficult and expensive. The ambient temperature in the test space is $\gtrsim$400°C, and therefore experiments at low temperature would be exceedingly difficult and expensive. This reactor is therefore fairly satisfactory for carrying out experiments related to phenomena in Table 4.2 that can be carried out at relatively elevated temperatures. However, it is not well suited to experiments involving complex instrumentation or low temperatures.

The FFTF reactor is being built as a test reactor and will be considerably more useful than EBR-II. This reactor will possess a higher neutron flux [~(20-40) × 10^{14} neutrons/cm^2 sec] and a larger experimental volume and will have a higher capability for instrumentation along with a favorable fast-neutron flux to gamma heating ratio. It

TABLE 4.4 Selected List of Current Neutron Sources in the United States

Neutron Source	Comments
Fast Reactors	
EBR-II (Idaho Falls)	High-T radiation-damage facility
FFTF (Richland 1980)	Large high-T radiation-damage facility
Mixed Spectrum Reactors	
HFBR (Brookhaven)	Mainly scattering source
HFIR (Oak Ridge)	Mainly scattering source
BSR (Oak Ridge)	Instrumented low-T radiation-damage facility
ORR (Oak Ridge)	Used for CTR radiation-damage research
CP-5	Instrumented low-T radiation-damage facility
Pulsed Sources	
LAMPF & WNR (Los Alamos)	
IPNS-I (Argonne--still in planning stage)	

should therefore be satisfactory for carrying out experiments related to Table 4.2 at elevated temperatures. (For example, there is adequate volume available for considerable mechanical testing.) Again, however, highly instrumented experiments at low temperatures would be extremely difficult. Also, a long operational cycle of ~1/3 year will limit flexibility in research programs.

Turning next to the mixed spectrum reactors listed in Table 4.4, the HFBR produces fast neutron and thermal neutron fluxes as high as ~5×10^{14} neutrons/cm^2 sec and has been used mainly as a source for scattering experiments. The experimental volume for radiation-damage studies is relatively small, but this situation could be improved by modifications. Also, the ratio of the fast to thermal neutron flux is well suited for radiation-damage studies.

The HFIR produces high fast neutron and thermal neutron fluxes (~15×10^{14} and 24×10^{14} neutrons/cm^2 sec, respectively) but has a very limited specimen volume available that is accompanied by large gamma heating. This reactor is heavily used as a neutron source for scattering experiments and as a producer of transuranic elements.

The BSR and CP-5 reactors are currently used for experiments concerned with basic phenomena of the type listed in Table 4.1. These reactors possess experimental spaces that can be relatively highly instrumented and that can also be cooled to low temperatures. (However, the CP-5 reactor is mainly used at present for scattering work.) Currently, the ORR is being used for CTR radiation-damage experiments. Spectrum tailoring can be accomplished in this reactor, and, for example, the amount of He produced by transmutation in an alloy containing nickel can be controlled by suitable manipulation of the spectrum.[5] Also, experiments concerned with the phenomena listed in Table 4.2 can be well instrumented in this reactor.

In general, therefore, the BSR and CP-5 reactors are the only reactors set up for Table 4.1 type studies at low temperatures. Since the oncoming FFTF will have a large capacity for Table 4.2 type experiments at higher temperatures, it is anticipated that HFBR and HFIR will continue to be heavily used for scattering work.

There are a number of additional mixed spectrum reactors including, for example, the following: N Reactor (United Nuclear), MITR-II (MIT), GTRR (Georgia Tech), MURR (U. Missouri), ATR (Idaho Falls), GETR (General Electric), and ETR (Idaho Falls). These reactors have no really outstanding and unique characteristics for performing radiation-damage work and therefore are not making noteworthy contributions to radiation-damage studies at present. In general, they produce modest fluxes of both fast and thermal neutrons and have relatively restricted capacity and accessibility, which therefore limit the specimen volume available and the capability for instrumentation. However, it has been pointed out in Reference 5 that some of these reactors could be highly useful for radiation-damage studies if a special effort were made to utilize them. At present, there is little incentive in this direction, since other more suitable facilities are available.

Of the pulsed neutron sources, Los Alamos has two facilities. The first is located at the LAMPF 800-MeV proton-beam stop and is used for a variety of purposes including radiation-damage studies, radiochemistry, and particle physics. The neutron flux is derived from the spallation process in a Cu beam stop and is in the range of 10^{13} neutrons/cm^2 sec. It should increase by a factor of 10 as the LAMPF beam reaches its design current of 1 mA in 1979. The irradiation facility provides three large volume (4 in. × 8 in. × 24 in. long) specimen positions with low-gamma heating. Instrumentation is expensive because of the shielding configuration, and low-temperature investigations would be costly for the same reason.

The second pulsed neutron irradiation facility is located at the associated Weapons Neutron Research facility (WNR) at Los Alamos. The multipurpose (scattering and radiation effects) WNR facility utilizes up to 10 percent of the 800-MeV pulsed proton beam to produce neutrons by spallation in a high-Z target. At 2 percent of the 1-mA LAMPF beam, the nominal operating condition, a neutron flux of 10^{13} neutrons/cm^2 sec will be available. The irradiation facility has hot-cell and remote-handling capability. The experimental volume is large, easy to instrument, and there are few restrictions with respect to facilities for high- and low-temperature experiments. Lower fluence experiments such as internal friction could use any of a large number of access ports in the target crypt without interference with other experiments.

IPNS is in the advanced planning stage at Argonne. The IPNS concept includes an independent radiation-damage source with bending magnets to switch the beam between separate radiation-damage and scattering target areas. This is an important concept, which is consistent with the different requirements of scattering and radiation-damage experiments. For example, the optimum target in the radiation-damage facility is tungsten (for maximum neutron productivity without gamma rays), whereas in the scattering target it is uranium (for maximum neutron productivity independent of gamma rays). The radiation-damage target requires a tungsten reflector to obtain a high flux of energetic neutrons with small energy degradation, whereas the scattering target requires graphite moderators to obtain a high flux of lower-energy neutrons. This concept allows a gamma-free, thermal-neutron-free, neutron spectrum with adequate experimental space without adversely affecting the scattering target or requiring a major modification in the target assemblies. A detailed description of this proposed facility is given in Reference 3. In the initial construction stage, IPNS will deliver an average flux of 10^{13} neutrons/cm^2 sec at the specimen in a relatively large volume in the presence of low-gamma heating. The experimental volume will be easy to instrument and includes facilities for low-temperature experiments. IPNS-II in the final stages will deliver about an order-of-magnitude higher flux (i.e., 2×10^{14} neutrons/cm^2 sec) to the specimen under otherwise rather similar conditions.

It should be re-emphasized at this point that a high-flux capability is of great importance in radiation-damage studies since it allows the study of (i) damage rate effects over a wide range; (ii) damage saturation phenomena; and (iii) neutron scattering from the defects produced by radiation damage. In case iii a high flux is doubly important since it allows the production of a relatively high concentration of defects by radiation damage in the first place, and then it provides intense scattering radiation for characterization of these defects.

4.5.4. Scientific Opportunities

The bombardment of atoms in a material by energetic nucleons results in the displacement of some of those atoms from their equilibrium sites by both elastic collisions (charged particles and neutrons) and inelastic collisions arising from nuclear reactions (high-energy neutrons). The defects in the material created by these collision processes are vacant lattice sites, interstitial atoms, and their agglomerates, and transmutation products. These defects can significantly alter technologically important properties of materials. As a result, such studies are of great economic significance in the commercial application of fission and fusion reactors to produce energy. Other study groups have devoted considerable time and effort in characterizing the materials problems resulting from neutron bombardment and have focused attention on the manner in which basic research can alleviate them.[1,2] These studies are summarized in the following.

Most of the basic radiation-effects research has been performed on the effects of bombardment of materials by low-energy electrons (1 MeV or so), high-energy heavy ions (5 MeV or so, Ni^+, for instance), and neutrons. The amount of energy transferred by the collision process varies widely from one type of irradiation to another. As a consequence, the use of one or another type of irradiation determines to a large degree the nature of the radiation damage by controlling the local densities of the defects and ultimately the clustering characteristics of the defects.

A great effort has been made in electron irradiation research because the energy of the primary electrons can be selected to produce single isolated vacancy-interstitial pairs, which permit detailed study of the isolated defects as well as their simple interactions. This research has been very successful, and there is now considerable fundamental understanding of the properties of these elementary defects, including their configurations.

As a result, the major challenge in radiation-effects studies now lies in the understanding of the nature of fast neutron bombardment and the cascades of defects that neutrons induce. Thus, studies of the size, defect densities, and defect configurations as a function of both energy and temperature within displacement cascades are now needed. A variety of techniques will be required to obtain this information, including the bombardment of neutron-irradiated samples with charged particles including electrons; diffraction studies by x rays, electrons, and neutrons; studies of dimensional changes; mechanical property changes; and electrical resistivity changes. All of these require transfer techniques that allow samples to be removed from the reactor at bombardment temperatures. In addition to the direct interest in the details of the cascades themselves, it is apparent that defect agglomerates play a key role in the observed changes in the mechanical properties of irradiated materials. Thus, simultaneous studies also should be undertaken to determine the interactions between dislocations and displacement cascades.

Heavy ion bombardment is used to simulate energetic neutrons because the results of several years of neutron irradiation can be introduced in a few hours with this technique. However, the thickness of the damage region is so small that electron-microscopy techniques must be utilized to study it, and the bulk effects of the damage cannot necessarily be inferred. Also, the precise correspondence between the two types of irradiation is not clear. Thus, in spite of the considerable success in such simulation efforts, an intense research effort with energetic neutrons is still necessary.

We have mentioned the requirement to transfer an irradiated sample from the reactor to a different site for additional measurements. Techniques for such transfers are already available but remain to be more widely exploited. In addition, *in situ* experiments involving highly sophisticated instrumentation and specimen manipulation seem possible in certain cases.

For use in such combined experiments, a whole new generation of extremely powerful high-resolution microscopic and microanalytical (microchemical) techniques is becoming available on a usable basis that could undoubtedly make large contributions to our current knowledge in well-designed radiation-damage experiments. These include field ion microscopy, atom-probe field ion microscopy, high-resolution, high-voltage electron microscopy, and scanning transmission electron microscopy using field emission guns.

Several of these techniques are capable of resolving individual point defects and individual impurity atoms and small clusters of these entities. The atom-probe field ion microscope is capable of chemically identifying single atoms removed from the specimen by pulsed field evaporation. Modern scanning transmission electron microscopes equipped with high brightness illumination systems are becoming capable of chemically analyzing exceedingly small regions consisting of a relatively small number of atoms in thin-film specimens.

Several new and valuable scattering techniques have been developed to the point where they are ready for exploitation. These include Rutherford backscattering, channeling spectroscopy, and scattering of intense hard x-ray beams produced by a synchrotron.

Other interesting current techniques include positron annihilation, EXAFS, and the use of the Mössbauer effect. Many current surface-physics techniques can be applied in a powerful manner to radiation-damage surface phenomena such as Auger spectroscopy, LEED, and HEED.

In the case of the more complex phenomena listed in Table 4.2, there is a current need for quantitative observations under well-controlled conditions. Once the phenomena are clearly identified and characterized, modeling studies accompanied by critical experiments would be valuable. In many cases, the experimental techniques mentioned previously would be useful. In the special case of the mechanical properties, recent developments based on the idea of a mechanical equation of state seem promising as an aid in dealing with irradiation effects.

Finally, some special opportunities exist[6] in the area of the study of radiation damage by means of neutron scattering (Table 4.3). These include the following:

(i) Study of the near-neighbor atomic environment of crystal defects using elastic diffuse scattering between the Bragg peaks. In this work the inelastically scattered neutrons can be filtered out.

(ii) Study of vibrational modes associated with crystal defects. Here, the information supplied by the neutron scattering is unique.

(iii) Study of spatial distribution of alloying and impurity elements in irradiated specimens. Here, the work is done better in many cases with neutrons than, say, with x rays, because complications with x-ray form factors are avoided. Also, elements that are nearby in the Periodic Table can often be distinguished.

(iv) Study of clusters, voids, etc. by small-angle scattering. In this case multiple scattering effects can be largely avoided.

4.5.5. Conclusions and Recommendations Regarding Facilities

Our recommendations regarding radiation-damage facilities may be divided into two main categories, neutron sources and instrumentation facilities.

Neutron Source Facilities

The Panel believes that there is a need to construct additional neutron sources for basic radiation-damage studies of the type listed in Table 4.1. The oncoming FFTF, as planned, should provide sufficient new experimental volume to satisfy the radiation-damage community with respect to experiments related to the more complex phenomena of the type listed in Table 4.2. At present, in the United States most of the experiments that fall in the basic-studies category are being performed in either the CP-5 reactor at Argonne or in the BSR reactor at Oak Ridge. These reactors are suited to highly instrumented experiments including low- (liquid He) and high-temperature loops. In addition, the ORR reactor at Oak Ridge is used by the DOE for a limited number of basic radiation-damage experiments and a more substantial program of experiments concerned with complex phenomena. Experiments dealing with basic phenomena to be conducted in the near future are in jeopardy, as these reactors apparently have short longevity at present (only a few additional years of life).

The pulsed neutron sources seem to be the most attractive sources as a replacement for either the CP-5 or BSR reactors. The pulsed sources produce low nuclear heating and deliver a neutron flux considerably higher than the older sources.

To meet the needs of the radiation damage community it is therefore recommended that the following steps be taken:

1. A pulsed neutron source with resources dedicated to radiation-damage research should be constructed providing

low nuclear heating and a time-averaged flux of 10^{13} neutrons/cm^2 sec with a capability of eventually reaching 10^{14} neutrons/cm^2 sec. These average flux levels correspond to those of the IPNS stages given later in Table 5.5

2. The Los Alamos beam-stop facility should be upgraded to provide easy experimental access and enhanced flux. The use of the WNR facility for radiation-damage studies should be encouraged.

3. At least one steady-state source for radiation-damage applications should be maintained in operation. This will be necessary since neutron radiation-damage phenomena may occur with the use of the pulsed source in temperature regimes where the relaxation times of the irradiation-induced defects become equal to or shorter than the period of the pulsing. In such cases, comparisons between results obtained under pulsing and steady-state conditions will be essential.

Additional Facilities

As in the other areas of neutron research, the Panel concludes that sufficient support should be made available for additional facilities that will allow the full utilization of the very expensive neutron sources discussed above. In the radiation-damage field, we believe that the full potential of both the existing and the proposed source facilities should be exploited. Specifically, more attention should be paid to the establishment of facilities in which specimens can be irradiated and then removed (often at low temperature and/or under vacuum) to other laboratories for further experimentation. Also, there is a need for more extensive and sophisticated *in situ* instrumentation.

In general, more ambitious radiation-damage experiments should be supported even though they may be relatively costly. In this connection, we note that much of the recent progress in basic electron radiation-damage research has been accomplished at the Kernforschungsanlage in Jülich, West Germany, as a result of unusually ambitious and well-instrumented experiments.

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6. See Reference 3, pp. 88-115, for more details.

5
NEUTRON FACILITIES AND TECHNIQUES IN THE UNITED STATES

5.1. SURVEY OF U.S. REACTOR FACILITIES

There are 13 presently operating steady-state nuclear reactors in the United States that serve as neutron sources for research studies in low-energy neutron scattering and radiation damage. These range in operating power from 100 MW to 1 MW and are listed in Table 5.1 along with a description of some of their characteristics. In addition to those tabulated, there exists a collection of small reacting facilities located mostly at university centers that serve for training and educational purposes, but whose flux characteristics are inadequate for research purposes. As is evident from the tabulation, the available thermal neutron flux from these sources varies over a wide range, over two orders of magnitude, since this quantity is roughly proportional to the operating power for this common class of enriched-uranium, condensed-core reacting systems. Seven of these reactors are located at DOE national laboratories (Brookhaven, Oak Ridge, Argonne, Ames, and Los Alamos), one at the National Bureau of Standards, and the remaining five are distributed at various university centers.

The commissioning dates of these reactors range from 1951 (for BSR) to 1969 (for NBS), with the two most powerful ones (HFBR and HFIR) being started in 1965 and 1966, respectively. This is significant in that the generally expected lifetime of a well-cared-for reactor is about 30 years. With the exception of some uninstrumented beam ports at ORR and lower-grade beam ports at HFIR, the available beam ports at the major reactors are very fully utilized. In many cases, the beam ports serve multiple functions in supplying neutron radiation to more than one spectrometer facility. This is not the case at some of the lesser facilities (MITR, University of Rhode Island, Georgia

TABLE 5.1 Neutron-Source Reactor Facilities Operating in the United States

Facility and Location	Reactor Power (MW)	Thermal Neutron Flux (10^{14} neutrons/cm^2 sec)	Commissioning Year	Yearly Operation Cost ($M)	Research Program Support[a] ($M/yr)	Number of Available Ports	Number of Spectrometer Units	Mode of Operation[b]
High Flux Isotope Reactor (HFIR) Oak Ridge National Laboratory	100	13	1966	4.1	1.10	{ 4 horizontal, 4 inclined	8	C
High Flux Beam Reactor (HFBR) Brookhaven National Laboratory	40	8	1965	2.8	2.50	9	12	C
Oak Ridge Research Reactor (ORR) Oak Ridge National Laboratory	30	2	1958	1.45	0.22	8	3	S
National Bureau of Standards Reactor (NBSR)	10	1	1969	1.20	1.60	12	11	C
University of Missouri Reactor (MURR)	10	1	1968	0.80	0.50	6	8	C
Massachusetts Institute of Technology Reactor (MITR-II)	5	0.8	1958 (rebuilt 1976)	0.50	0.20	13	6	C
Argonne National Laboratory Reactor (CP-5)	5	0.3	1953	1.32	1.65	7 plus rad.-damage facility	8	C
Ames Laboratory Research Reactor (ALRR), Iowa State University	5	0.3	1964	0.86	0.37	11	7	C
Omega West Reactor Los Alamos Scientific Laboratory	8	0.3	1957	0.36	0.20	7	3	S
Bulk Shielding Reactor (BSR) Oak Ridge National Laboratory	2	0.2	1951	0.40	0.28	4 rad.-damage facilities	-	C
University of Rhode Island Reactor	2.5	0.2	1966	0.30	0.04	5	3	S
Georgia Institute of Technology	1	0.2	1965	0.30	0.02	6	2	S
Ford Nuclear Reactor University of Michigan	2	0.1	1963	0.30	0.05	5	3	C

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[a] For LEN scattering and radiation damage, not including reactor operation expense.

[b] C, continuous three-shift operation; S, one-shift or intermittent operation.

Institute of Technology, University of Michigan), where a sizable number of beam ports remain uninstrumented because of staff and budgetary limitations.

Operational expenses for these reactors vary widely, depending on their size and function, with the collective total amounting to $14.7 million per year. These expenses are shared among the various functional programs at the reactors with about 40 percent, or $6.0 million per year, being assignable to programs related to the study topics of this Panel, namely, LEN scattering and radiation damage. Funding of these reactor operational expenses is borne principally by research divisions of DOE (about 60 percent), by the Departments of Commerce and Defense, and by university sources.

As mentioned in Section 3.1.3, it is planned that some of these reactor facilities will be shut down or withdrawn from service to the LEN community. Thus the ALRR facility at Iowa State University with DOE support is scheduled for shutdown at the end of 1977, the ORR facility is to be operated mainly for the Magnetic Fusion Energy program, and eventually within the next few years the aging CP-5 facility will be withdrawn from service. Additionally, the intermediate- and lower-flux facilities at the university centers have all been operating under budgetary deficit conditions (with a continuing strain on university resources) in recent years, and it can be said that their continued operation is seriously threatened.

5.2. PULSED NEUTRON SOURCES

In addition to neutron generation by the fission reaction in nuclear reactors, neutrons may also be produced by other nuclear processes activated by high-energy charged particles, such as electrons or protons. Energetic charged particles from accelerators invariably come in repetitive bursts, and the resulting neutron source is of pulsed nature. The primary fast-neutron spectra covers a wide energy range and after energy degradation by a moderator placed adjacent to the target a copious source of low-energy neutrons can be obtained. There are three characteristic features that distinguish this type of source from reactor sources: (1) the neutrons are generated in repetitive bursts whose time width and repetition rate can be controlled; (2) the neutron energy spectrum can be tailored by relatively easy changes in moderator characteristics and this means, in practice, that the epithermal (energies >1 eV) neutron intensity can be larger by several orders of magnitude than that available from reactors; and (3) the peak intensity in the neutron pulse can be very much larger than the time-average flux, which is proportional to the target power loading. When operated with high-performance accelerators, this type of neutron source can offer a useful-flux improvement of an order of magnitude over that of any existing reactor source and can open many new areas of experimentation. A different brand of experimental facilities compared with that at steady-state reactors is called for in using the pulsed source, and much of the present experience with such systems has been obtained in Europe and in Japan, where a number of pulsed neutron systems have been operated during the past decade.

Pulsed neutron beams with very-high-peak flux can also be produced by "flashing" nuclear reactors, and a very-large-scale development of such a system is currently under way in the Soviet Union with their IBR-II pulsed reactor (see Table 3.1). The expected peak flux from this pulsed source is nearly two orders of magnitude larger than that of any existing steady-state source. This large gain factor is vitiated somewhat by the associated lower repetition rate (5 pulses per sec) and longer pulse duration than those that characterize the spallation sources.

Table 5.2 shows a summary of existing pulsed neutron sources in the United States. The RPI facility was constructed in 1962 and has been used for many years in obtaining fundamental nuclear-physics data. It is currently in part-time use and is not now instrumented for LEN studies of condensed matter. This laboratory could be used as a valuable testing ground for new pulsed-source instrumentation as well as a center for training new people in the use of these techniques.

ZING P', which is just becoming operational at the time of writing, is the second prototype stage in the Argonne pulsed neutron source program; it is intended to provide experience with pulsed-source instrumentation. The previous prototype, ZING P, operated for three months in 1974-1975. ZING P' uses the Booster II proton synchrotron to produce pulses of 500-MeV protons, which impinge on a tungsten target. Neutrons are produced in this high-Z target by the spallation reaction with neutrons being evaporated from the heavy target nuclei. The peak thermal neutron flux will initially be about 10^{14} neutrons/cm^2 sec, equivalent to that of a medium-flux reactor, and is expected

TABLE 5.2 Pulsed-Neutron-Source Facilities in the United States

Name and Location	Accelerator and Target	Repetition Rate (pulses per sec)	Source Pulse Width (μsec)	Time-Average Neutron Production Rate $(\mathrm{sec})^{-1}$	Peak Thermal Flux $(\mathrm{cm}^2\ \mathrm{sec})^{-1}$	Peak Epithermal Flux Density at 1 eV $(\mathrm{cm}^2\ \mathrm{sec\ eV})^{-1}$	Number of Ports	Operation Date
ZING P' (ANL)	Booster II 500-MeV, 1×10^{12} protons/pulse tungsten target	30	0.1	2.4×10^{14}	8×10^{13}	2.0×10^{14}	4	1977
WNR (LASL)	LAMPF at 300-μA 800-MeV, 3×10^{11} protons/pulse tantalum target	120	10	6×10^{14}	5×10^{13}	3×10^{13} (for 2-μsec pulse)	12	1977
RPI Gaerttner Linac	50-MeV e^- 33-kW uranium target	360	4.5	2×10^{14}		1.2×10^{13}	5	1962
ORELA	140-MeV e^- 65-kW tantalum target	1000	0.1	1×10^{14}		3×10^{12}	5	1967

to increase as Booster II performance is tuned up. Operating costs are comparable with those of a medium-flux reactor if operated year round. At present, the primary support for Booster II operation is from the High Energy Physics program.

The WNR source listed in the table is also just becoming operational and is one of the stages in the LASL pulsed-neutron-source program. The primary impetus for this program, which is supported by the Medium Energy Program and the Division of Military Applications, DOE, is weapons neutron research (WNR). It appears, however, that the WNR facility is available for an ancillary unclassified basic research program utilizing nine beam ports over 90 percent of the operational time. At the time of this report, a small materials research program is under way that utilizes two of the twelve beam ports.

The Oak Ridge Electron Linear Accelerator (ORELA) is operated as a pulsed neutron source for nuclear-physics measurements. A few exploratory condensed-matter experiments have been successfully performed even though the facility is not properly instrumented for this type of measurement.

5.3. NEUTRON INSTRUMENTATION IN THE UNITED STATES

A survey of the neutron instrumentation facilities in the United States operating at the above neutron-source centers shows the existence of 73 spectrometer units and 5 radiation-damage facilities. The latter facilities are concentrated at two reactor sources, four being located at the special-purpose reactor, the BSR at Oak Ridge, and the other at CP-5 at Argonne. Discussion of the radiation-damage facilities has been given earlier in Section 4.5.

The spectrometer units fall into different classes according to their function and operating characteristics, and Table 5.3 summarizes their distribution relative to the neutron flux classification of their sources. Generally speaking, these spectrometers can be said to be of the same age as their sources since they have been usually installed at the time of, or shortly after, new reactor commissioning. Great strides in the area of spectrometer design have been made over the last decade, yielding new varieties of older classes of units with improved operating efficiency as well as spectrometers of new concept for specialized use.

TABLE 5.3 Operational Neutron-Scattering Instruments Located at U.S. Neutron Sources

	Number of Spectrometer Units at Source with Flux (neutrons/cm^2 sec)		
Type of Instrument Facility	$<10^{14}$	$(1-3) \times 10^{14}$	$>5 \times 10^{14}$
Crystallographic (4 circle)	5	2	4
Powder diffraction (crystal monochromated)	11	5½	2
Powder diffraction (time-of-flight)	1	1	0
Small-angle scattering	1	5	4
Polarized beam	4	1½	1
Inelastic scattering (triple-axis)	9	4	8
Inelastic scattering (time-of-flight)	1	2	1
	32	21	20

It is apparent when one compares the spectrometer units in the United States with those that have been installed over the last five years at the relatively new high-flux ILL reactor at Grenoble that our instrumentation has lagged behind that available elsewhere.

A breakdown of the spectrometer distribution given in Table 5.3 into three broad, functional categories, (1) two-axis units for crystallographic and diffraction studies, (2) three-axis units for excitation-fluctuation studies, and (3) time-of-flight units for both types of study, shows a distribution of 46, 21, and 6 units, respectively. Equating spectrometer availability to research activity, this distribution suggests that about 60 percent of the research activity is associated with crystallographic or diffraction types of problems. The small number of time-of-flight units is of course correlated with the sparsity of suitable pulsed-neutron sources as discussed earlier.

It is interesting that the fractional distribution of inelastic-scattering instruments is fairly common across the different flux categories, implying that experimentalists at lower-flux centers are undeterred by the more stringent intensity problems associated with such studies. It should be stated, however, that the most prolific and most significant work in inelastic scattering has been accomplished at the two high-flux centers so that source strength is a very real factor.

All the spectrometer units located at sources in the two highest-flux categories of Table 5.3 are in full-time operation. This is not the case with the remaining units in the lowest-flux category, and it has been estimated that these units are operational only about 50 percent of the time because of staff limitations and insufficient program support. Such waste of spectrometer resource is a matter of concern to this Panel and, of course, also to those sources engaged in supporting reactor operation.

Of additional concern is the fact that a sizable number of available beam ports are uninstrumented even at some of the more powerful sources. Table 5.4 shows a tabulation of these with the most notable entries being those at ORR and at HFIR. It should be mentioned that the eight beam ports built into the HFIR assembly (four being high-flux radial variety and four of lower-flux vertical slant type) were added as appendages to this primarily isotope-producing

TABLE 5.4 Presently Unused Beam Ports at Neutron Sources

Number	Facility	Source Flux at Beam Port (neutrons/cm^2 sec)
4	HFIR	2×10^{14} (inclined ports)
4	ORR	1.5×10^{14}
9	WNR	$(0.5\text{-}3.4) \times 10^{14}$ peak at 120-Hz pulsed source
6	MITR	8×10^{13}
10	Georgia Tech	2×10^{13}
2	Rhode Island	2×10^{13}

reactor, and the science community is greatly indebted to the original designers for this foresight. The four slant ports (still with moderately high flux) have gone unused, again because of staff and budgetary limitations and partly because equivalent-flux ports of more acceptable configuration have been available at ORR in the same laboratory.

The large number of unused ports at the pulsed WNR source arises because this facility is just coming into operation. The instrumentation of these ports, in parallel with the instrumentation associated with the IPNS project (later discussion), could help to overcome the deficiency in pulsed-neutron science, with all of its potential advantages, which has been present in the United States. The remaining entries of Table 5.4 are those associated with the three university-based reactors of low- or low-intermediate-flux capability. Here again, one sees the effect of a common pattern of inadequate support for programs and staff. This is particularly serious in the cases of the Missouri and MIT university projects, where, even though a respectable and very useful neutron flux is available, there is incomplete usage of facilities, either ports or existing instruments, because of research program and staff support limitations.

5.4. POSSIBILITIES AND PLANS FOR BETTER UTILIZATION OF EXISTING FACILITIES

This Panel has considered the question of better utilization of existing facilities from a technical standpoint. Our conclusions can be logically grouped into three broad categories, which will be presented below.

5.4.1. Instrumentation Improvement

The United States has done reasonably well at equipping the major facilities with first-class instrumentation. While the conventional inelastic-scattering and diffractometer instruments in the United States are generally competitive with the best in the world, it is important that steps be taken to upgrade U.S. spectrometer instrumentation to include new powerful instruments developed elsewhere in recent years and as well to improve ancillary equipment associated with the spectrometers. This forms one of our recommendation items and is described in more detail in the following section. Detailed discussion of the scientific opportunities associated with these instrumentation improvements and extensions has been given earlier in various parts of Chapter 4.

Development of Special Spectrometers

Ultra-High-Energy Resolution Spectrometers We strongly recommend that special-purpose spectrometers be developed with energy resolution between 0.1 and 25 μeV as compared with the 100-μeV resolution generally attainable with present spectrometers. We believe that it will likely require two separate instruments to cover this energy range, each costing about $0.5 million, although we have not attempted a realistic design. These instruments could be used to study the diffusion of heavy metals (e.g., in superionic conductors), the inelasticity of central peaks associated with phase transitions, and the reorientation of molecules in solids, among many other experiments.

Small-Angle-Scattering Spectrometers In various parts of Chapter 4, we have repeatedly mentioned the usefulness of small-angle-scattering techniques in studying problems of biological and polymer structure, clustering in solutions and alloys, long-range domain structure in superconductors, and defect classification. In spite of the broad range of applications for this technique, the facilities in the United States for performing this type of research are very limited both in number and sophistication. The Panel sees this as an omission, and it is encouraging that the National Science Foundation is presently soliciting proposals for a national facility in this area. Although a high-performance national facility would be extremely valuable by itself, the Panel believes that there is justification for having other SANS facilities with specialized features useful in defined research areas at distributed centers.

New Monochromators and Polarizing Devices

The availability of good crystal monochromators and polarizers is clearly a critical problem in neutron scattering. There is a need for good monochromators operable at energies up to 200 meV. Use of beryllium as a monochromator is the best prospect for this application, but recent attempts to grow suitably large single crystals have been unsuccessful. Successful efforts to obtain good monochromators for high-energy neutrons would yield results as impressive as the addition of a hot source to a present reactor and would have the same effect on epithermal energy work as pyrolytic graphite has had at low energies. The crystal reflectivity problem is even more severe for polarizing crystals. The crystals ordinarily used to produce polarized beams have rather high absorption for thermal neutrons and so are not very efficient. The work at ORNL on the development of ^{57}Fe crystals as polarizing monochromators with their high reflectivity and low absorption should be re-emphasized, as should the work on multilayers and "super mirrors" at BNL. Layered, mirror assemblies when magnetized can act as polarizing devices by selective critical reflection of the two neutron polarization states, and their use has just begun to be explored. Finally, it is essential that white beam polarizers be developed if polarized beams are to be used effectively with the existing and proposed pulsed sources. Polarized proton filters (involving resonant proton polarization at low temperatures) are currently being developed for this, but other novel assemblies such as those exploiting matched magnetic-nuclear incoherent scattering may yet prove feasible.

Multidetectors and Associated Data Acquisition Systems

Dramatic gains in neutron-data-collection rates are possible with linear and area position-sensitive detectors. These detectors permit simultaneous data collection over a large solid-angle range as seen from the scattering specimen, and they have already been exploited in studies of small-angle scattering, in powder diffractometry, and in protein crystallography. There are currently strong programs in the development of these detectors at both Brookhaven and Oak Ridge National Laboratories. Further development and refinement of these detectors and the associated electronics may be needed to make them adequate for use with pulsed, time-of-flight techniques.

Instrumentation Associated with Extreme Sample Conditions

As the field of neutron scattering has matured, there has been a move toward application associated with more extreme sample environmental conditions--mK temperatures, 100-kG fields, 40-kbar pressures. The United States has often led the way and is presently well equipped in these applications. However, there is a need for continued effort to maintain a strong position.

5.4.2. Modification of Sources

Hot and Cold Sources within Reactor

The high-flux research reactor at the Institut Laue-Langevin (Grenoble, France) incorporates both hot (2000 K) and cold (20 K) moderator regions, which are necessarily thermally isolated from the main moderator. This means that the thermal neutron energy distribution emanating from these regions is shifted up or down and an enhancement of neutron flux in the beam ports viewing these regions is obtained at either the high (>200 meV) or low (<5 meV) energy end of the spectrum. It should be noted that these two sources were designed into the reactor from the very beginning of the reactor design--they were not "bolt-on" accessories.

The United States lags far behind Europe in this area. With regard to cold sources, there are three reactors at which such sources with their required cryogenic accessibility could be installed, HFBR, NBS, and MITR-II. Brookhaven expects to install a liquid H_2 cold source in 1978, while the National Bureau of Standards plans to install a D_2O-ice cold source by 1979. Because of reactor design considerations, neither of these two facilities will be able to support a large complement of neutron guide tubes such as is installed at the ILL reactor in Grenoble. Both laboratories are planning to install small-angle scattering facilities as well as more general types of instrumentation at these sources.

There are no present plans to install a hot source in any reactor in the United States. The only major reactors in which a satisfactory hot source can be installed are the Missouri University Research Reactor (MURR) and the ORR at Oak Ridge.

Full Utilization of Undeveloped Beam Ports and Multiple Instrumentation at Beam Ports Now in Use

There are a number of beam ports at various reactors that are not now instrumented or used for materials research. Table 5.4 contains a list of such beam ports at various reactors in the country. Clearly, these ports could be developed to give an immediate increase in the number of facilities available for neutron-scattering research. The source flux for these facilities is also shown in the table to allow an assessment of the relative utility of these ports.

Further, it would be possible to increase the number of instruments now installed at various reactors through multiple usage of individual beam ports. However, it should be noted that each such multiple use implies a compromise with the use of existing instruments. There are currently several examples of such multiple usage of instruments (HFBR, HFIR, MURR, NBSR, CP-5) at single-beam ports, and there are plans to do more of this at several facilities where the radiation shieldwork is adaptable and compatible with spectrometer space requirements. This trend is expected to continue and should be encouraged.

Power Increase of Existing Reactor Sources

For a given reactor, source flux is nearly proportional to operating power. Thus, the source flux can be increased by increasing power where the reactor design (most noticeably the features of radiation shielding and cooling capacity) allows this. There are several reactors where this is planned or being considered, as listed below:

1. HFBR--The operating power of HFBR at Brookhaven is being increased from 40 to 60 MW. This modification will be completed in 1979 for a total cost of $2.7 million and a projected increase in operating cost of 15 percent per year.
2. NBSR--The reactor at the National Bureau of Standards presently operates at 10 MW. This power could be increased to 20 MW for a total cost of $0.6 million and an increase of 20 percent in operating costs.
3. MURR--The reactor at the University of Missouri currently operates at 10 MW. Power could be increased to 20 MW with a minimal capital cost, but relicensing is required.

It appears to this Panel that all of these improvements would be well worth the capital expenditure and the relatively small increase in annual operation cost that is required. All beam ports at these centers are currently instrumented with fully active programs of use at HFBR and NBSR but with support-limited programs at MURR. Encouragement would be given to MURR for reaching its design-power limit if the augmented program support included as one of the Panel recommendations were to be effected. In assessing the relative desirability of having twice the intensity versus twice the number of spectrometers, it should be pointed out that experimentalists would favor the former, since that would permit new forms of present intensity-limited experimentation.

5.4.3. Other Problems Faced by Research Reactor Centers

Some difficulties faced by U.S. research reactor centers in maintaining efficient and productive operation are due to increasing government regulations that have arisen because of concerns about the safety and environmental impact of nuclear-power facilities. Existing and potential problems include uncertainty in the cost and supply of enriched uranium fuel and the increased operating costs necessitated by new regulatory policies. The Panel hopes that a sympathetic viewing of these increased costs of source operation will be taken by the federal and other agencies who are supporting research programs at the centers.

5.5. POSSIBILITIES AND PLANS FOR NEW SOURCE FACILITIES

Table 5.5 summarizes the characteristics of three pulsed-neutron sources that have been proposed for development in the United States. IPNS I and IPNS II are stages of the Argonne National Laboratory program; two stages in the development of the Los Alamos WNR facility as a pulsed neutron source are shown; and, in addition, potential uses of the Oak Ridge Electron Linear Accelerator (ORELA) are summarized. The table entries are estimates of performance using consistent and feasible targets and moderator configurations. Design figures published elsewhere may assume different configurations tailored for special purposes. The capital costs associated with the construction of these facilities and their anticipated annual operating expenses are those developed after careful study by the project laboratories.

TABLE 5.5 Proposed Pulsed-Neutron Source Projects in the United States[a]

Project	Accelerator and Target	Repetition Rate (pulses per sec)	Source Pulse Width (μsec)	Time Average Neutron Production Rate $(\text{sec})^{-1}$	Peak Thermal Flux $(\text{cm}^2\ \text{sec})^{-1}$	Peak Epithermal Flux Density at 1 eV $(\text{cm}^2\ \text{sec eV})^{-1}$	Number of Ports	Proposed Operation Date	Incremental Capital Cost ($M Actual)	Annual Operating Cost of Source (1979 $M)
IPNS I	Booster II 500-MeV, 5×10^{12} protons/pulse uranium	30	0.1	3×10^{15}	1×10^{15}	2.5×10^{15}	12	1981	6.4	2
IPNS II	High intensity synchrotron 800-MeV, 5×10^{13} protons/pulse uranium	60	0.2	9×10^{16}	1.5×10^{16}	3.5×10^{16}	12	1984	62	5
WNR	LAMPF at 1-mA 800-MeV, 1×10^{12} protons/pulse uranium	120	10	4×10^{15}	3×10^{14}	2×10^{14}	12	1979	0	1.3 Primarily supported by DOE (DMA)
WNR + Storage Ring	LAMPF at 1-mA 800-MeV, 3×10^{12} protons/pulse at 40 Hz uranium	1-720	0.25	4×10^{15}	1×10^{15} at 40 Hz	2.5×10^{15} at 40 Hz	12	1983	17	2.0 Primarily supported by DOE (DMA)
ORELA + Booster	140-MeV e^- (1-2) × 65 kW ^{233}U booster	½ × 1000	0.2	5×10^{15}	--	--	5	--	12.5	0.6

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[a] The fluxes quoted in this table have been estimated on a standard basis to obtain a uniform comparison of different sources and may differ slightly from values given in design reports.

5.5.1. The Intense Pulsed Neutron Source

The Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory has been proposed as a national facility for condensed-matter research using neutron scattering and radiation-damage methods. High-energy protons will produce intense pulses of neutrons by spallation in heavy-element targets. Two experimental facilities are to be provided. For slow-neutron scattering, three hydrogenous moderators slow down neutrons to the energies of interest (<10 eV), to provide short, intense pulses at the neutron source that would service 12 beams. The moderators can be easily changed and differently tailored by choice of material, poisoning, or cooling to optimize the spectral characteristics of the source as seen by different instruments. Beryllium reflectors around the moderators serve to increase the available flux by factors of 5-10. For radiation effects studies, a separate target will provide fast neutrons in an array of ten flexibly arranged, easily accessible, vertical irradiation thimbles close by the source. The source, like all spallation neutron sources, will have an extremely low-gamma-ray flux.

The IPNS program is proposed to proceed through two phases. In the first phase, the existing 500-MeV Booster II accelerator will deliver 5×10^{12} protons during each 100-nsec pulse to a uranium target at a repetition rate of 30 pulses per sec. Peak thermal neutron fluxes of 10^{15} neutrons/cm^2 sec will be produced, accompanied by a peak epithermal flux density 2.5×10^{15} neutrons/cm^2 sec eV at 1 eV. In the second phase, a new, dedicated high-intensity synchrotron will provide 5×10^{13} protons during each 200 nsec pulse, 60 pulses per sec. Peak thermal and epithermal neutron fluxes in the neutron scattering source will be 10^{16} neutrons/cm^2 sec and 2.5×10^{16} neutrons/cm^2 sec eV. The time-average fast-neutron flux will be 2×10^{14} neutrons/cm^2 sec.

A request for detailed design and construction funds for IPNS, beginning in fiscal 1979, has been made to DOE, which would make the first phase available in fiscal year 1981 and the second phase in fiscal year 1984. This schedule, in conjunction with the two prototype stages already completed, represents a logical and orderly development leading to a "new-generation" pulsed-spallation source with unprecedented high peak flux in both thermal and epithermal regions, to be operated as a national research center.

5.5.2. The Weapons Neutron Research Facility (WNR)

The WNR is an intense pulsed neutron source that utilizes a fraction of the 800-MeV proton beam of the Clinton P. Anderson Meson Physics Facility, LAMPF, located at Los Alamos, New Mexico. Up to 10 percent of the LAMPF 500-μsec pulse, and corresponding 10 percent of the proton current, can be diverted to the WNR facility through a beam transport tunnel.

Operations at the WNR will begin in the fall of 1977. At that time, the LAMPF proton beam will be 300 μA (time-averaged) delivered on a four weeks on-one week off schedule. The initial allocation of 2 percent (10 μsec in length) of the LAMPF 500-μsec pulse will permit production of 6 × 10^{14} neutrons/sec (time-averaged) from a tantalum target. The source strength will increase in steps to the final figure of 4 × 10^{15} neutrons/sec in 1979 as the LAMPF average proton current reaches the design goal of 1000 μA and by using targets of actinide isotopes.

The proton storage ring, proposed for operation by the summer of 1983, would reduce the proton pulse length to 0.25 μsec, improving energy resolution and increasing peak neutron fluxes. The repetition rate for discharge of the stored, circulating proton beam may be varied from 1.2 to 720 Hz, providing, respectively, a peak thermal neutron flux ranging from 4 × 10^{16} to 0.6 × 10^{14} neutrons/cm^2 sec for the different discharge rates. This flexibility will permit optimization of signal-to-noise ratio, adjustment of repetition rate to that of pulsed environment experiments, and extension of the useful neutron energy range to either higher- or lower-energy regions. In addition, with modest improvements to source cooling and shielding, the WNR could utilize the full allocation of the LAMPF beam (10 percent), and this would provide a peak thermal flux of 5 × 10^{15} neutrons/cm^2 sec at a 40-Hz discharge rate. Any further expansion of the WNR source strength would be formidable.

The WNR facility is expected to produce by 1979 nine neutron beams, which will be accessible to the national materials research community, making it competitive with those at our present reactor centers. With the exception of the U.S.S.R. IBR-11 flashed reactor facility, it will represent the most intense pulsed neutron research facility available anywhere in 1979. One of the most important areas needing development is the generation of new instrumentation capable of making use of high-flux pulses, and the WNR could clearly play an important role in answering this need.

The availability of this existing pulsed source at Los Alamos presents opportunity both for the development of instrumentation for high-flux pulsed-neutron spectroscopy and the accessibility of state-of-the-art facilities for the West Coast scientific community. There are at present no instrumented high-flux sources near this materials-research community.

5.5.3. Pulsed Neutron Booster at ORELA

The Oak Ridge Electron Linear Accelerator (ORELA) is now in use as a neutron source for nuclear cross-section measurements. Bursts of electrons of 140-MeV average energy are incident on a Ta target, producing intense bremsstrahlung and subsequent photoneutrons. A preliminary study (ORNL-TM-4987) has examined the possibility of broadening the use of ORELA to include low-energy neutron-scattering experiments. This can best be accomplished by constructing a new target facility devoted to condensed-matter studies employing a ^{238}U target in a neutron booster consisting of a subcritical assembly of ^{233}U. In the proposed facility, electron pulses could be switched from the present target to the new one in any desired time sequence, thus accommodating simultaneous experiments in nuclear physics and condensed matter. The neutron booster would increase the total number of neutrons per pulse by a factor of about 50. In the tentative plans for the new facility, flight-path lengths, shielding, and overall design would be optimized for about five neutron beams.

ORELA can produce pulses at any rate up to 1000 per sec. The entry in Table 5.5 is based on sending every other pulse at a maximum rate to the condensed-matter target and sharing operating costs equally between the nuclear-physics and condensed-matter programs. The resulting repetition rate of 500 per sec would cause frame overlap troubles for experiments employing thermal neutrons, but it is not too high for many problems requiring relatively fast neutrons. It is anticipated that this facility would be devoted to those experiments requiring high incident energies that are now impossible with reactor sources.

It should be noted that the neutron booster introduces an undesirable delayed neutron background between pulses which need not be present with a spallation source. Because the neutron booster is nearly critical, it would have many of the same licensing and containment problems as a reactor. Aside from these undesirable features, the ORELA + booster performance should be roughly comparable with that of IPNS I in the epithermal region.

5.5.4. Advanced Research Reactors

While there are no current formal proposals for new research reactors, the age of our current reactors (the youngest being 10 years old) and the long lead time from conception to operation (about 10 years) suggests that it is prudent to plan now to satisfy our needs in the latter part of the next decade. We list in Table 5.6 some characteristics of three reactors that could be constructed based on existing technology.

The Advanced HFIR-Type II would employ a cylindrical flux-trap core with D_2O as coolant and reflector with a power density about twice that of the HFIR. The substitution of D_2O for Be and the higher power density should lead to an increase in thermal flux in the reflector region of a factor of 4 over that now obtained in the HFIR. This reactor is probably very close to the upper power density limit possible with existing technology. The Advanced HFIR-Type I would operate at the current HFIR power but would achieve an increase in thermal flux in the reflector of a factor of 2 by using D_2O as coolant and reflector. Either of these reactors could be equipped with a large number of beam tubes and cold and hot sources. The central flux trap region could be designed for accommodating radiation-damage experiments with adequate instrumentation and/or target rods for isotope production.

Also listed in Table 5.6 is a medium-flux reactor, highly instrumented and with hot and cold sources. This reactor has the merit of lower capital and operating costs

TABLE 5.6 Contemplated Advanced Research Reactors

	Power (MW)	Construction Time (years)	Flux $(\text{cm}^2\ \text{sec})^{-1}$	Ports	Capital Cost (1977 $M)	Operating Cost (1977 $M)
Advanced HFIR I	100	10	3×10^{15}	17	125	5.5
Advanced HFIR II	200	10	6×10^{15}	17	150	6.0
Medium-flux reactor	25	5	3×10^{14}	13	45	2

and would provide a number of instruments useful for problems for which the highest possible flux is not necessary. It is similar in concept to the reactor now being constructed at Saclay.

The capital costs for all three of these reactors are highly uncertain in the absence of design studies. At present, there is no effort being made within the United States on design of new research reactors. It would seem wise to establish a small group to perform conceptual design studies so that a decision among the various options could be made based on more fully developed plans.

5.6. COMPARISON OF PULSED AND STEADY-STATE TYPES OF EXPERIMENTATION

In order to assess the importance of new pulsed sources it is necessary to compare their potential uses in LEN scattering research with the known capabilities of steady-state sources. Such a comparison must remain somewhat speculative at this time since the pulsed-source characteristics are estimates not yet reduced to practice, while the characteristics of steady-state reactors are well known.

With a steady-state source, measurements must be made in the presence of an unwanted background because of high-energy neutrons and decay products from their interactions with the surroundings. In the case of a pulsed source, the slow neutrons of interest arrive after the fast neutrons, so in principle it is possible to discriminate against them.

In the case of a pulsed source, both targets and moderators can be changed relatively easily to suit varying experimental requirements. In contrast, the flux density of a thermal reactor may be modified in local regions by means of hot or cold moderators, but the overall structure is relatively inflexible. Pulsed-spallation or bremsstrahlung sources do not necessarily involve any fissionable materials and therefore avoid complications arising from the safety regulation of these materials. In what follows we first discuss experiments involving thermal and lower-energy (cold) neutrons and then discuss the essentially unexplored epithermal energy area. For simplicity, we have assumed equivalent instantaneous thermal neutron flux for all sources.

5.6.1. Elastic Scattering

Powder Diffractometry

In this type of experiment the sample is in the form of a powder consisting of many small crystallites with presumed random distribution of orientation. Recently, it has been demonstrated that a great deal of structural information can be obtained even in the case of complex unit cells by modeling the positions of atoms within the unit cell and adjusting the parameters of the model until a best fit to the data is obtained. This method is called powder profile analysis, and it has been discussed in Section 4.2.2. For experiments involving approximately 1 percent resolution, sample fluxes are comparable for pulsed and steady-state sources. Pulsed sources appear to have a greater potential for improved resolution. An additional advantage of pulsed-source diffraction patterns is that the fractional width of the Bragg peaks and hence the uncertainty in the derived unit cell parameter is independent of position in the pattern. This is not true for conventional powder patterns.

Crystallography

A double-axis crystal spectrometer (the first axis being associated with monochromatization) is currently the most popular technique for crystallographic studies with steady-state reactor sources. This method involves point-by-point scanning of the momentum transfer by the specimen crystal. If the crystal structure is simple with only a few atoms distributed in the unit cell (the fundamental cell which by repetition forms the macroscopic crystal) only a small number of Bragg peaks are needed to classify the structure. In the case of more complex crystal structures (for instance in biological materials), a very large number, perhaps many thousand, are needed for this purpose and the conventional method becomes very time-consuming and cumbersome. For such cases, use of a large-area, high-resolution position-sensitive detector offers dramatic improvement for both steady-state and pulsed sources. In particular, with a pulsed source a large range of momentum transfer can be sampled so that many Bragg reflections can be observed simultaneously.

Small-Angle Scattering

Small-angle-scattering studies usually involve low-energy neutrons whose diffraction gives information about the spatial variation of the sample cross section over the range 10-1000 Å. What counts in this type of experiment is the time-average flux at the sample position. Many possibilities exist for improved experimental conditions both at reactors and with pulsed sources, making use of cold moderators, correlation choppers, and large-area position-sensitive detectors. Although pulsed sources offer advantages for certain types of studies in this area, most small-angle-scattering experiments are better adapted to a steady-state source.

Diffuse Scattering

In a diffuse-scattering experiment, such as the study of short-range order in liquids, the important scattered intensity is distributed quasi-continuously over a wide range of momentum transfer. To carry out such measurements efficiently, a large-area position-sensitive detector is needed. If comparable detectors are used in conjunction with either a pulsed or steady-state source, the two should be competitive.

Polarized-Beam Experiments

The usual ways of producing polarized neutron beams in the low-energy range are (a) Bragg scattering from a ferromagnetic single crystal, (b) spin-dependent scattering or capture by polarized nuclei (e.g., protons) in a filter, and (c) critical reflection from magnetized mirror surfaces. Bragg scattering is only suitable for a monoenergetic beam, and this method is well developed. Polarization filters (see Section 5.4.1) are effective for both monoenergetic and continuous sources but have not yet been used as extensively. At present, therefore, steady-state sources have the advantage, but a practical white-radiation polarization filter could make the two competitive.

5.6.2. Inelastic Scattering in Thermal Energy Region

Discrete Spectra

The study of well-defined elementary excitations such as phonons and magnons will certainly remain an important mainstream application of thermal neutron scattering. In this kind of experiment, the important information comes from a narrow range of values of $\vec{q},\omega$. This situation is well matched to the capabilities of a triple-axis spectrometer, which selects a single value of $\vec{q}$ and a corresponding single value of ω. A TOF spectrometer can provide the same information, but only a small fraction of all the recorded events would be of interest since a wide range of values of both $\vec{q}$ and ω are measured with such a device. Therefore, the equivalent efficiency of the TOF spectrometer as compared with the triple-axis device is jeopardized in this case, and steady-state source methods appear to be the most efficient approach to this type of problem.

Continuous Spectra

Conversely, any type of problem in which the scattering events of interest are spread over a wide range of values of $\vec{q}$ and ω, such as single particle excitations and Stoner bands, the TOF spectrometer is much better matched to the experimental requirements. Very large increases in total counting rates are then possible because of the ability to use large arrays of detectors.

Quasi-elastic Scattering

Some quasi-elastic scattering experiments involve spectra well localized in $\vec{q},\omega$ space, such as critical scattering, and are therefore better matched to a triple-axis spectrometer than to a TOF spectrometer. Other experiments of this type, for example, the study of diffusion in superionic conductors, involve spectra spread over a wide range of $\vec{q}$ values and are therefore better matched to a TOF spectrometer.

5.6.3. Specialized Experiments

Limited-Geometry Experiments

In some experiments, the sample is nearly surrounded by other parts of the apparatus necessary for controlling sample environment, such as in high-pressure, high- or low-temperature, and high-magnetic-field work. In such cases, a TOF spectrometer is much preferred since a single angle of scattering can be used to obtain information covering a range of values of the momentum transfer.

Transient Phenomena

A pulsed source is more naturally suited to study the reponse of a system subjected to time-varying conditions. Examples include pulsed magnetic fields, pulsed electric fields, pulsed laser excitation, acoustic pulses, and electrochemical stimulation for physical, chemical, and biological systems.

Neutron Interferometry

These experiments, in which individual neutron wave packets are split coherently and then recombined for studying quantum phase effects, are just now being developed. In present form they seem to be more suitably studied using steady-state reactor sources, although there may very well arise applications with pulsed sources.

Ultra-Cold Neutron Beams

Pulsed and steady-state sources are equally useful in carrying out neutron storage container (bottle) experiments with very-low-energy neutrons because the relevant quantity is the maximum instantaneous phase-space density available at the source.

Energy-Resolved Radiography

Direct shadowing of a sample onto a position and time-sensitive detector can reveal the positions of various nuclear isotopes in the sample. This technique is possible only with pulsed-source radiation.

5.6.4. Epithermal Neutrons

One of the exciting new areas that will become available with the advent of a pulsed neutron source is the use of epithermal neutrons ($E \gtrsim 0.2$ eV), which are not presently available from reactor sources. Such energetic neutrons could be used to study all forms of excitations in solids, liquids, and gases that are above the thermal energy range.

Figure 5.1 illustrates the region of energy-momentum space made available with 5-eV neutrons compared with those covered by x ray, electron, and thermal-neutron inelastic-scattering techniques at energies and resolutions currently available. It can be seen that new classes of measurement will become possible requiring either large

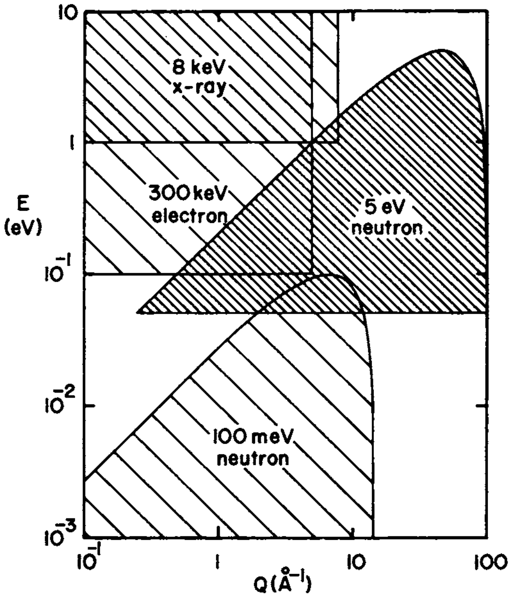

FIGURE 5.1 Regions of energy-momentum space currently accessible with various inelastic-scattering techniques.

energy transfer, large momentum transfer, or intermediate-energy transfer simultaneous with low momentum transfer.

The general experimental arrangement will probably involve a chopper synchronized with the source pulse placed some distance from the moderator, which allows neutrons of a selected velocity to fall on the sample. Some distance after the sample, a band of detectors records the flight time and angle of scattering, thereby determining both the energy loss and momentum transfer suffered in the collision with the sample. Although instrumentation for this energy range has not been refined, some performance characteristics can be obtained simply from the estimated pulse width of the epithermal neutrons and the requirement that energy and momentum be conserved in the scattering process.

Suppose, for example, the neutron incident energy is 1 eV and we wish to study excitations in the sample with a wave vector of 1 Å^{-1}. The incident neutron has a wave vector of 22 Å^{-1}, to which we must add the momentum transfer of interest. The largest possible energy change of the scattered neutron corresponds to orienting the scattering wave vector oppositely to the incident wave vector, so that the scattered neutron has a wave vector of 21 Å^{-1}. This corresponds to a maximum energy transfer of 90 meV. For a momentum transfer of 4 Å^{-1}, the maximum energy transfer is 320 meV. Therefore, the fact that momentum as well as energy must be conserved in the scattering process means that for reasonably small momentum transfers only a small fraction of the incident neutron energy is available to be transferred to the sample.

The situation improves with higher-energy neutrons. For example, if the incident neutron has an energy of 10 eV, the maximum energy transfers for $q = 1$ Å^{-1} and 4 Å^{-1} are, respectively, 280 meV and 1 eV. However, at such high beam energies the finite time resolution of the TOF spectrometer is a significant problem. If we assume an overall time resolution of 1 μsec, which is optimistic, and moderator-sample and sample-detector distances of 30 m, the energy resolution is comparable with that obtainable in inelastic electron scattering but compares unfavorably with that available from infrared absorption and Raman scattering. In assessing the value of experiments using epithermal neutrons, these important restrictions must be kept in mind.

Some examples of measurements that will be made possible by the availability of high fluxes of epithermal neutrons are the following:

(a) Vibrational spectroscopy of molecular solids, especially hydrogen-bonded systems and organic molecules involving large-amplitude hydrogen vibrations, including molecules adsorbed on surfaces.

(b) Studies of molecular vibrations in fluid phases.

(c) Studies of optic modes and their higher harmonics in metal hydrides, which allow one to map the potential in which the hydrogen atoms reside.

(d) Electronic band spectroscopy at nonzero wave vector, e.g., of transitions between bonding and antibonding levels in semiconductors.

(e) Spectroscopy of electronic transitions that are optically forbidden but observable in neutron scattering.

(f) Studies of high-energy magnetic excitations in ferromagnetic metals (Stoner modes).

(g) Measurements of local structure with high spatial resolution in amorphous solids and molecular liquids.

(h) Measurement of atomic momentum distributions (neutron Compton scattering) in many types of systems, e.g., superfluid helium, metal hydrides, hydrogen-bonded ferroelectrics.

Finally, because epithermal neutron scattering represents a totally new technique, it will almost certainly lead to new discoveries that we cannot now predict.

6

USERS AND EDUCATION TOPICS

6.1 CHARACTERISTICS OF THE U.S. USER COMMUNITY

The introduction of new techniques, such as small-angle neutron scattering (SANS) and profile analysis, as well as the prospects of significantly more intense and higher-energy sources, make neutron scattering an important technique to an ever-widening community of scientists. Moreover, in the area of neutron radiation damage, a sizable community has been involved in a wide range of effort spanning the complete spectrum from basic to highly applied work. It can be anticipated that this effort and its associated community will be expanded as the intense national effort to develop advanced energy sources with concomitant radiation-damage problems continues. Particularly promising areas for greatly expanded use of neutron-scattering techniques include microscopic studies of polymer structure and elasticity, metallurgy, biological structure and organization, and molecular spectroscopy.

This potential users community is very much larger than the traditional neutron users community in this country, which has consisted mainly of solid-state physicists and crystallographers. For example, approximately half (450) of the experiments performed at ILL last year were metallurgical and polymer small-angle studies. The SANS technique has enabled direct study of clustering in alloys and the configuration of individual polymer chains both in solution and in solid samples. Likewise, the ability to locate hydrogen atoms and to study selectively positions of a complex biological assembly have caused a rapid growth in usage, which is still in its initial stages. At BNL, the major biological neutron facility, the growth in the user community over the recent past has been 25 percent per year.

Molecular spectroscopists contribute a major scientific effort within the physical-chemistry community, and the most useful neutron technique to them is TOF spectrometry, in both thermal and epithermal regions. Development of pulsed sources, which must naturally utilize TOF and which will increase the available epithermal flux by about three orders of magnitude, should stimulate significant usage by chemical spectroscopists, who are at present underrepresented at U.S. facilities.

It thus seems appropriate at this stage to assess present and future user policies with a goal of developing methods to encourage optimum use of the facilities by both specialists and nonspecialists. Three aspects of the users question that have evolved in our deliberations are those related to education, funding, and operational procedures, and we shall discuss these in turn.

Before addressing these issues, it will be helpful to identify the users community in this country, that is, the type and number of scientists who are presently exploiting the neutron facilities and aspects of their activity. It is convenient to classify user scientists into two groups, full-time equivalent users (FTE) and part-time users (PTU) according to the scale of their preoccupation in neutron science. The former group would include all the in-house scientists at the DOE-NBS centers who have dedicated interest in the field and additionally those at academic or industrial centers whose dominant research interest is that of LEN-scattering radiation damage whether or not their physical location is at a neutron facility. On the other hand, PTU's would be identified as those scientists who occasionally exploit the facilities, generally in collaboration with in-house staff, to obtain some packet of structural or dynamical information on materials that they are studying as part of a more general research program. Such classification would generally be associated with a grouping of specialists and nonspecialists, since in many cases the PTU's would not be experienced in details of either the facilities or the technique.

Despite a historical position of leadership in the field, the neutron-scattering community in the United States is relatively small. A survey of scientist personnel in this community for the past year shows numbers of 102 FTE and 284 PTU in total, with 70 FTE and 255 PTU located at or associated with DOE-NBS centers. The high fraction of FTE users at the DOE-NBS centers arises, of course, because that is where most, and the best, facilities are to be found, and the even higher fraction of PTU activity there arises because this group is naturally attracted to the best facilities. In addition, there are practical roadblocks for PTU activity at not centrally supported facilities (as at the universities), which are as mundane as "who is to pay for the beam charges and liquid helium." Numerous cases have come to our attention wherein a scientist has transferred his research contacts and problem execution away from even home-based and adequate facilities or has abandoned the research prospect for just such reasons. In this community of users, the key group has been the dedicated FTE personnel at the major centers. The scientists who have been attracted there are top grade, and many have achieved worldwide distinction. Much of the research progress, both fundamental and applied, in this country's effort has originated with this group and their uncompromising dedication to the very best science. The research management of our major laboratories is due high commendation for its unwavering support of this group, often in the face of mission-oriented pressures on the laboratories. This Panel is not unaware of the importance and necessity of mission-related, more applied research activity, and this must be pursued with vigor at all obvious points of contact with neutron technology. However, it would be indeed tragic if our zeal to attain mission goals were to lessen, let alone force abandonment of, our science objectives. A healthy balance between the two is obviously needed and should be struck.

Our key group of FTE scientists has produced systematic, in-depth studies (as reviewed earlier) in such important areas as phase transitions (BNL), liquid dynamics (ANL), magnetic properties (ORNL), and hydrogen in metals (NBS) with important support from the other group of users. A survey of the research publication activity of this group of scientists over the past three years shows publications numbering 204, 217, and 313 per year for 1974-1976. Since the FTE staff size at the centers has been essentially unchanged during this period, this growth in productivity is presumably associated with a growth in the number of part-time users, which is confirmed in reports from the centers. *This growth trend in the number of outside users of the neutron facilities is an encouraging one since the Panel believes that there should and could be enlarged activity in this direction.* The usefulness of LEN techniques in contributing to both fundamental and technological developments has been amply documented, and there is a strong feeling that a larger community should profit from the existence of the neutron facilities.

It is important that this expansion of outside use should in no way interfere with in-house programs whose continuing high quality must be maintained. Since the highest-flux sources are already heavily utilized, this may require additional instrumentation and support staff at these facilities. Additionally, our intermediate-flux centers should be considered as playing an important role in servicing an expanded users community. It has been noted in Section 5.3 that there is incomplete usage of the facilities, either ports or instruments, at all the university centers. MURR, although fully instrumented, experiences support-limited usage of spectrometer units.

6.2 THE EUROPEAN EXPERIENCE

It is interesting to compare these communal statistics for neutron scattering with those appropriate to activities at the Institut Laue-Langevin in Grenoble, France, where a high-flux reactor has been in operation since 1972. At this one location in 1976 there were present 80 scientists as in-house staff, and the laboratory attracted a total of 900 outside users. Research publications in the area of this study have been growing rapidly, and in 1976 they numbered approximately 250. It is clear that the outside user community contributes there, relative to U.S. centers, much more significantly to the laboratory activity, with a broader range of topics being pursued. The new instrumentation there is highly sophisticated, and many new special-purpose spectrometers of novel design have been developed. Because of the large number of scientists involved in the program, the scheduling of spectrometer time is very tight, and this sometimes has meant that studies are performed in less depth than desired. Proposals for experimentation are acted upon by the Scientific Council, and their number has been growing dramatically, as shown in Figure 6.1, which is extracted from the ILL 1976 Annual Report.

While discussing the European facilities and their use in this field of science, it should be noted that a sizable number of scientists from the United States has spent residency periods at European centers. These periods have been either of longer variety (associated with leaves of absence or sabbattical leave) or of short-term character in which a specific problem (with specimens and perhaps

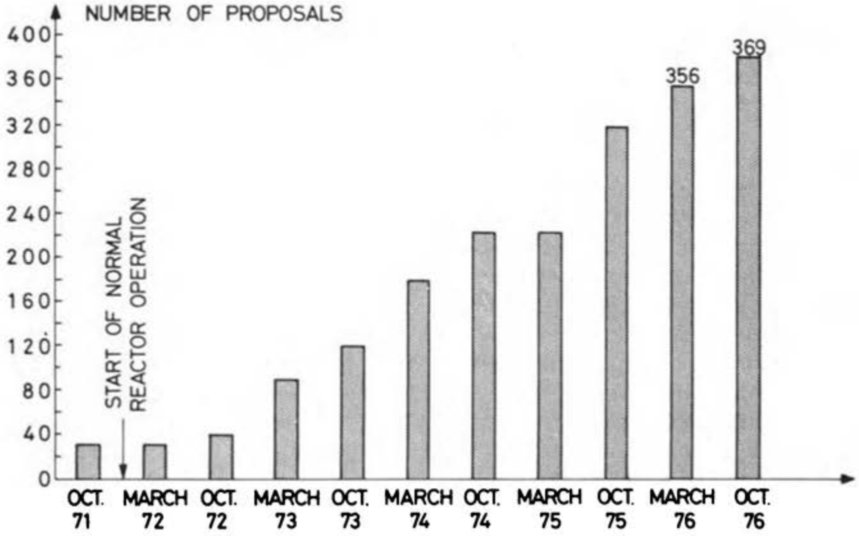

FIGURE 6.1 Evolution of research proposals submitted at Institut Laue-Langevin, Grenoble, France.

specialized equipment being transported from the United States) has been scheduled on a particular instrument having unique features pertinent to the problem study. This scientific interplay is obviously fruitful, but it is also apparent that longer lead times and heavier travel expenditures are involved, let alone a greater disruption in the scientist's schedule, than would be the case if the facilities were available nearby.

The European experience demonstrates that scientists find the neutron technique a superior tool and suggests that, if properly stimulated, a larger community of outside users would develop in this country. Such development should be pursued in a manner that maintains the excellence in current U.S. research standards, while at the same time encouraging nonspecialists to learn about the potential of neutron techniques in their areas of interest, what facilities are available, and how opportunities can be developed for their use.

It is useful also to understand something of the European philosophy of neutron scattering, which is seen as a field of research that impacts strongly on every area of condensed-matter science, including physics, chemistry, and biology. It seems to have been clear in the European experience that an active neutron community does not spring full-born from the universities--that it requires an infusion of funds and a commitment of resources at both the centers and to outlying research groups. This has been done at the highest government levels, the Science Research Council in the United Kingdom, for example, and is a long-term commitment. The three-country Grenoble activity is one manifestation of this basic commitment to neutron scattering. The new spallation source recently approved for the Rutherford Laboratory is a more current example, as is the medium-flux reactor at Saclay, France, and a similar installation being discussed for Germany.

Given all of this activity, it may still fairly be said that the most sophisticated neutron scattering in the world has consistently been done at the U.S. reactor centers. This is due largely to the will and talent of the scientists at these facilities, their flexibility of operation, and their uncompromising dedication to the very best science. There has, concomitantly, not been in the United States an attempt equivalent to that at Grenoble to seed and develop a large user community. Again, a statistic from the 1975 ILL Annual Report shows a median age of their neutron staff of 31 years. This is substantially lower than the U.S. figure. It reflects the fact that we have not been engaged in an aggressive training program mainly because positions for neutron scatterers do not generally exist at universities, and the national laboratories have been staffed to budget limitation. Thus the issue is further complicated by questions of support and funding for both inside and outside use to which we shall presently direct our attention.

6.3 EDUCATIONAL ASPECTS

The educational issue has three aspects:

1. Education and training of professional neutron scatterers and radiation-damage experts;
2. Education and encouragement of potential science collaborators;
3. Education of the technological and applied science community.

In the first instance, the smaller university reactors play an important role and, indeed, the MIT reactor center, for example, has trained several of our prominent neutron scatterers. The informal and slower-paced atmosphere in these centers is more conducive to the training of an experimentalist, providing opportunity for trial and error, and offers a more intimate research environment. Oftentimes experiments that have been initiated at a small reactor will be transferred to a high-flux facility when appropriate. For this and other reasons it seems clear to this Panel that university reactor projects should be maintained.

In addition, however, the educational function of the national facilities should be emphasized. Currently, for example, the national laboratories have an active postdoctoral program, in addition to the occasional, albeit significant, training of PhD's in which scientists trained in both neutron and nonneutron work participate. These programs bring new people into the field and permit the broadening of interests of trained personnel. Even within this group, however, a difficulty ultimately exists with the limited availability of permanent positions for young scientists who have developed expertise in neutron science. The issues then become coupled. A healthy community consists of university training centers, along with predoctoral and postdoctoral positions at national laboratories, and with positions on the faculties of universities and the staffs of industrial laboratories devoted to neutron science. These latter groups must, in turn, feel welcome at the national laboratories and have minimal barriers to their participation in the laboratory program. Again this impacts on funding policy.

The second group of users should be classified as (currently) nonprofessional neutron scientists. They may consist of x-ray diffractionists or light scatterers or be even further removed, having an interest in neutron methods only (initially) as as adjunct to their major research. Such a person might be, for example, a specialist in superconductivity or magnetism in metals, who has a research problem susceptible to neutron study and yet who has little expertise with the neutron technique. This larger group of scientists also embraces a great many people who would like to do neutron scattering in a more routine fashion--one in which triple-axis spectrometers are not needed as an example, or who would like to apply neutron methods to more applied problems. Through the encouragement of these people, we stand to benefit from the new science that will emerge. *This group of scientists will play an especially important role as we develop our major facilities as true* *national facilities*. For example, among the largest and most successful contingents in the European neutron-science field are the chemists who perhaps perform the largest volume of research work. This is in contrast to the situation in the United States where this group has played a relatively minor role in neutron research.

The third group of users comes mainly from industry and from the applied-science community. In this category, there are a large number of potential users not only of scattering and radiation-damage facilities but also of other facilities for activation analysis, transmutation doping, radiography and so forth. Some of these users are already being quite successfully handled at the medium-flux reactors at NBS, MURR, and MIT and where the highest neutron flux is not required; this should be continued. But there is an educational mission to be accomplished in bringing neutron scattering and radiation-damage potentialities to the attention of a larger community.

We should note here the paradox of an active, vigorous, and successful neutron-scattering and radiation-damage community, which is also undersupported and insufficiently extended into, and utilized by, the larger scientific and academic community. Our objective is to preserve those modes of operation that have brought U.S. neutron science to its position of preeminence while extending and expanding the entire community. It is clear from the U.S. and the European experience that neutron research can have an immense impact on all of condensed-matter science and engineering--from protein biology to alloy development. It is also equally clear that a full realization of this impact has not been attained in the United States, which ultimately returns to funding pattern restrictions.

6.4 FUNDING AND OPERATIONAL PATTERNS

There have developed four different modes of meeting the operational costs of our neutron sources: (1) the DOE national laboratories (BNL, ORNL, ANL, and LASL) receive multiple program support from federal agencies; (2) using the neutron source, the NBS reactor is supported by the Department of Commerce with backup support from other agencies or organizations using the neutron source; (3) the state-supported facilities (MURR, RI, and Georgia

Tech) are supported by state allocation plus user contribution to the university centers; and (4) the two private university facilities (MIT and Michigan) are supported by user contribution and general university resources. We shall address these in turn.

At present, the national laboratories have been more than willing to make their experimental facilities available cost-free to responsible outside scientists with proposals of scientific merit within the limitations placed by their own research activity and the necessity for their particular facility characteristics, be it intensity or specialized instrumentation or collaborative staff expertise. This truly befits their designation as *national centers.* A university scientist, or an industrial scientist with a nonproprietary problem, is faced only with his own ancillary expense responsibility such as salary support, travel, and special equipment needed for the experimentation, and this must be parceled from his general program support derived in the main from government agencies. Roughly speaking, the neutron-source expense will amount to perhaps 50 percent of the total program costs for a particular experiment, and thus the national center contribution is a significant one. *The Panel believes that this procedure is a proper one within the national center concept and that it should be continued and developed more fully.* It requires a sympathetic and enlightened view from the agencies (not necessarily the same as that supporting the center) that are supporting individual research programs at universities of their obligations to share in the neutron-related program. Moreover, this Panel believes and recommends that outside proposals to the national centers should not be restricted to merely those pertinent to the agency mission, but rather the criterion should be based on the good, useful science that can result.

Considering the other end of the source funding spectrum in categories (3) and (4), where individual users, either in-house or outside, make proportional contributions to source operational costs, we propose and recommend a continuation of the present mode with the proviso that funding agencies, such as the NSF, supporting individual research projects drawing on the neutron source take an enlightened, broadened view of their obligation to share in meeting source operating costs. In this way the distributed centers can see the mechanism for meeting the threat to their continued existence arising from present fiscal insecurity. Proceeding in this direction can mean that the university centers will also be national in concept, with accessibility open to all good science. The Panel also recommends that stringent reviewing of outside proposals by center review groups ensure that usage of facilities be distributed in the most effective manner, for example, if high-flux and other features are not paramount, the problem should not be handled at high-flux national centers.

6.5 INFORMATION DISSEMINATION ON NEUTRON FACILITIES

Another important factor related to future LEN science concerns the dissemination of information on the field and its techniques along with the education of the potential user community. Support should continue to be available for summer schools and workshops on neutron methods at the university and national laboratory centers on topics of wide relevance and applicability, such as molecular biology, polymer chemistry, metallurgy, surface science, and profile analysis in powder crystallography.

6.6 USER POLICY QUESTIONS

Finally, we come to the question of user policies at the national facilities. Policy statements already exist at the national laboratories, but either they have not been actively implemented or they have not been instituted because of unresolved questions (such as beam port charges for nonmission work and distribution of inside-outside activity). It is clear that a users committee of both inside and outside members should exist at each national facility to review proposals and to establish policy for use of the instrumentation. Working together with the laboratory research administration or the laboratory representation on the committee, the committee should arrange for an equitable distribution of instrumentation time between inside and outside programs, with consideration being given to the type of instrumentation available and to the interests and size of the resident staff. It is important that the laboratory interests, whether they be mission-oriented or of basic science variety, be safeguarded. The activities of the users committee at different laboratories should be correlated so that proposals are properly directed to the most effective neutron source.

There should be available from each laboratory a good description of all of its facilities and instrumentation. Proposals for beam time should cover briefly the intended research and the requisite equipment, special requirements, period of data collection, and other factors that will be needed in performing the research. Submission of the proposal could follow informal contacts exploring the research possibilities, and, in appropriate cases, the potential user might be encouraged and assisted to attend summer workshops or symposia or to take other means in acquiring familiarity with the technique and its potentialities.

In short, there should be a flexible policy that encourages both an active in-house and outside user program. A clear necessity of any such procedure should be the preservation of the excellent science activity at our national facilities. These are first and foremost scientific establishments, and they must be judged on that basis and not simply by the number of workers who pass through their doors.

One aspect of the accessibility of neutron sources to users is the question of geographic proximity of the source to the users' home base. We note the paucity of neutron sources in the western parts of the nation. In Chapter 5, we discussed the prospects for developing such facilities at the WNR with all of its potential capabilities. If first-class facilities for performing LEN research were made available at WNR, it would alleviate an imbalance in the current distribution of neutron sources in the United States.

6.7 SUMMARY OF RECOMMENDATIONS

1. The major neutron sources should be treated as national facilities rather than as strictly mission-oriented research centers. This implies central funding of general operating costs, as is now done both at ILL and at medium- and high-energy nuclear physics and synchrotron physics facilities in the United States.

2. In order to realize the scientific opportunities in broadly distributed areas of science that the report

has emphasized, there is a strong need to broaden the group of users of the existing and potential facilities. This wide class of new users must be first of all identified, educated, and encouraged. Topical workshops and summer schools should be held, and funding support for short-term consultation visits and exploratory experimentation should be made available. User policies already formulated at some of the national laboratories and university centers should be formalized and widely publicized.

User committees at the centers composed of both in-house and outside scientists should review brief written proposals for outside work, which should be scheduled in a flexible manner. Interchange between various review committees should be developed so that research problems are distributed in the most effective manner. In-house consultants or collaborators and technical support should be available to part-time users of the facilities. It is important that the present excellence of in-house and collaborative work at our centers be maintained and not be jeopardized by this broadened use of the facilities.

3. University neutron research centers should receive continuing support in their role of educating scientists and engineers in this field; in their making accessible intermediate-flux facilities to the national community; and in their housing of independent, home-based research groups.

4. Attempts should be made to make neutron scattering and radiation-damage facilities more widely accessible to scientists working in different geographical regions of the country.