The neutron reflectivity profile of a symmetric diblock copolymer of polystyrene and polymethylmethacrylate annealed on a silicon substrate. In the main figure, the open circles are the measured reflectivity, and the solid line is the calculated profile. The inset figure represents the depth dependence of the scattering-length density profile used to calculate the reflectivity. These results were obtained at a modified spectrometer on a thermal beam at the research reactor as part of a collaborative effort with scientists from IBM, San Jose. As a result of the success of these first measurements, a new dedicated instrument has been designed for installation in the CNRF in 1990. The new facility will provide an order of magnitude improvement in the capability for such measurements.

The neutron reflectivity profile of a symmetric diblock copolymer of polystyrene and polymethylmethacrylate annealed on a silicon substrate. In the main figure, the open circles are the measured reflectivity, and the solid line is the calculated profile. The inset figure represents the depth dependence of the scattering-length density profile used to calculate the reflectivity. These results were obtained at a modified spectrometer on a thermal beam at the research reactor as part of a collaborative effort with scientists from IBM, San Jose. As a result of the success of these first measurements, a new dedicated instrument has been designed for installation in the CNRF in 1990. The new facility will provide an order of magnitude improvement in the capability for such measurements.
ABSTRACT

This report summarizes all those programs which depend on the NIST reactor. It covers the period from October 1, 1988 through September 30, 1989. The programs include the application of neutron methods to the characterization of materials, neutron standards, trace analysis by neutron activation analysis, neutron depth profiling, nondestructive evaluation, and the production of radioisotopes.

Key words: Activation analysis; crystal structure; diffraction; isotopes; molecular dynamics; neutron; neutron radiography; nondestructive evaluation; nuclear reactor; radiation.

DISCLAIMER

Certain trade names and company products are identified in order to adequately specify the experimental procedure. In no case does such identification imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the products are necessarily the best available for the purpose.
# CONTENTS

Abstract .................................................. ii

Reactor Radiation Division .............................. 1

Organization Charts ..................................... 6

Research, Engineering and Technical Staff .......... 9

Description of Technical Activities .................. 12
  Reactor Operations and Services ..................... 13
  Neutron Condensed Matter Science .................... 14
  Cold Neutron Project ................................ 28
  Independent Programs ................................. 31

Outputs/Interactions .................................... 40
  Publications ........................................... 41
  Technical and Professional Committee Participation and Leadership ......................... 52

Industrial and Academic Interactions ............... 54

Associated Activities .................................... 55
The responsibilities of the Reactor Radiation Division are threefold: to operate the research reactor (NBSR) in a cost-effective manner while protecting the public safety; to conduct a program of materials research using neutron methods, while developing and maintaining state-of-the-art instrumentation to ensure the best utilization of the NBSR as a NIST and national resource; and to develop and operate the Cold Neutron Research Facility as a National User Facility, providing unique measurement capabilities to U.S. researchers.

In order to fulfill these responsibilities, the Reactor Radiation Division (RRD), in collaboration with other researchers in the Institute for Materials Science and Engineering (IMSE), other NIST centers, and many outside organizations, develops and applies neutron methods to a broad range of problems of national concern. These include the use of activation and other nuclear methods for chemical analysis, the characterization of the submicroscopic structure and dynamics of materials (e.g., hydrogen in metals, polymers, ceramics, alloys, magnetic materials, amorphous systems, micelles, microporous materials), the determination of residual stress and texture in materials, the use of neutron radiography to examine large components, the establishment of neutron flux and personnel radiation standards, and the development of a Cold Neutron Research Facility (CNRF).

The sections that follow are a summary of the technical activities of the Reactor Radiation Division over the past year. These activities are covered in much greater detail in a NIST Technical Note entitled NIST Reactor: Summary of Activities July 1988 through June 1989, which should be consulted for further details.

Major Activities:

In addition to operating the reactor, the Reactor Operations group is responsible for sample irradiations, both for Activation Analysis and isotope production. Once the materials are irradiated, other groups (either from NIST or outside organizations) are responsible for the analysis and scientific evaluation. The Center for Analytical Chemistry (CAC) is responsible for the majority of the Activation Analysis work, which provides a very sensitive technique for the determination of trace elements at very low concentrations. The method is extensively applied to the characterization of Standard Reference Materials (SRM's), to environmental studies, and to other problems such as stoichiometry in ceramic superconductors. In addition, CAC develops novel applications of nuclear methods for analytical chemistry needs, such as neutron depth profiling and prompt gamma activation analysis. Many outside agencies (e.g., FDA, FBI, Treasury, EPA) make use of the activation and analysis capabilities developed by RRD and CAC. As part of the CNRF, CAC is working with RRD to develop greatly improved Depth Profiling and Prompt Gamma capabilities.

RRD staff, in collaboration with many researchers from IMSE, NIST, other government agencies, universities, and industry, carry out an active program
of research on materials characterization, and develop and maintain a state-of-the-art suite of neutron scattering instrumentation. The emphasis is on the study of new materials of importance to science and technology such as polymer blends, composites, electronic and structural ceramics, chemical catalysts, novel magnetic systems, thin films and interfaces, biological materials, and new metal alloys. This work involves more than 200 researchers from outside the Division, working both collaboratively and independently. At the present time, the NIST reactor has more users than any other facility for neutron research in the U.S., even before the CNRF is completed. As an essential part of this base research program, the Division staff are also responsible for the ongoing development of new applications of neutron methods and new instrumentation necessary to maintain the NBSR at the forefront of neutron research.

RRD staff also develop and apply neutron methods to problems in Non-Destructive Evaluation (NDE), such as the determination of residual stress in bulk specimens, measurement of texture in formed metals, and other areas. The technique of autoradiography, in which activation of elements in an artifact are used to determine elemental composition as a function of position is applied to study rare paintings, both for research and restoration purposes.

The Center for Radiation Research (CRR) carries out a program in radiation standards and measurement using the facilities at the NBSR. Through the use of double fission chambers and a series of accurately calibrated fission foils, they provide the basis for reactor neutron flux and power density measurements needed in the U.S. reactor development program. The calibration and intercomparison of the series of standard fission foils makes use of standard reference neutron fields established in the thermal column of the NBSR. CRR also maintains well characterized filtered beams of 2, 25, and 144 keV in the reactor through-tubes for the calibration and development of personnel neutron dosimeters.

A major new element of the Division mission is centered in the Cold Neutron Project, with the goal of providing a critical new measurement capability for U.S. researchers. This multiyear project makes use of the large volume cold source installed into the reactor in 1987 by providing seven neutron guides (analogous to fiber optic light pipes, except for a difference in scale) to conduct large (6x15 cm) beams into a newly constructed experimental hall that provides 20,000 sq. ft. of new experimental space. In this hall, fifteen experimental stations will be installed over the next three years to provide new cold neutron experimental capability not currently available anywhere in the U.S. Several non-RRD groups are involved in this effort, including CAC, the Center for Atomic, Molecular and Optical Properties (CAMOP), Exxon Research and Engineering Co., Eastman Kodak Company, Sandia National Laboratory, the National Science Foundation, and several universities. The CNRF will be operated as a National User Facility, with a large fraction of the experimental time allocated by a Program Advisory Committee.

Reactor Utilization:

The NBSR is a National Center providing neutron-based measurement capabilities to researchers in IMSE, NIST and many other government
agencies, industries and universities all over the U.S. Interactions with other scientists and organizations take the form of both collaborative efforts and independent programs which rely on utilization of the reactor and facilities provided by the NBSR staff. The extent of such interactions for FY 89 are indicated in the tables below. The number of personnel shown in Tables 1 and 2 include many short-term collaborators as well as permanent government, university or industrial guest scientists. These numbers are constantly changing and so may not be exact.

Collaborative interactions are those in which workers from outside the RRD cooperate scientifically with RRD scientists on problems of mutual interest. These interactions are summarized in Table 1.

Many of the other agency, university, and industrial collaborators have worked with us regularly for many years. In many cases research participants provide research sample preparation or characterization, theoretical analysis or other special expertise to the neutron research activities. Most come from all over the world to spend a few days, weeks, or months to carry out specific experiments using the facilities available at the NBSR. Collaborative programs include measurements on new magnetic materials and superconductors, fast-ion conductors, polymers, and biological molecules, thin films, catalytic and microporous materials, hydrogen in metals, voids and precipitates in alloys and ceramics, etc.

Table 1. Collaborative Interactions

<table>
<thead>
<tr>
<th>No. of Personnel</th>
</tr>
</thead>
<tbody>
<tr>
<td>FY 89</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>RRD Permanent Scientists</td>
</tr>
<tr>
<td>Non-RRD Participants</td>
</tr>
<tr>
<td>Other NIST</td>
</tr>
<tr>
<td>Other Agency</td>
</tr>
<tr>
<td>University</td>
</tr>
<tr>
<td>Industrial</td>
</tr>
<tr>
<td>International</td>
</tr>
<tr>
<td>Total Non-RRD</td>
</tr>
</tbody>
</table>

Independent programs are those programs carried out independently of the Reactor Radiation Division scientific staff by other NIST Divisions and outside organizations. Table 2 summarizes these interactions.

Table 2. Independent Programs

<table>
<thead>
<tr>
<th>No. of Personnel</th>
</tr>
</thead>
<tbody>
<tr>
<td>FY 89</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>Other NIST</td>
</tr>
<tr>
<td>Other Agencies</td>
</tr>
<tr>
<td>Universities</td>
</tr>
<tr>
<td>Industrial</td>
</tr>
<tr>
<td>International</td>
</tr>
<tr>
<td>Total</td>
</tr>
</tbody>
</table>
These tables demonstrate the extensive utilization of the NIST reactor by scientists and engineers from outside the Division. They come from 14 NIST Divisions and offices, 18 other Federal organizations, 45 U.S. universities, 23 U.S. industrial laboratories, and 23 foreign laboratories.
FY 89 Representative Accomplishments:

- The CNRF buildings were completed, dedicated by Secretary of Commerce William Verity on Jan. 12, and occupied in March.

- The neutron guide installation was begun by boring the seven holes through the reactor confinement building.

- A large number of reactor maintenance and upgrade projects were completed during the shutdown to install the guides, thus minimizing overall shutdown time.

- The Center for High Resolution Neutron Scattering (CHRNS) was established with a grant from NSF.

- The recently developed NIST/Sandia/ICDD diffraction database has been integrated by several manufacturers into their electron microscopy instruments to greatly enhance capabilities for materials characterization. The new database is also being extended to allow identification by full lattice matching to increase the power of this materials identification resource.

- Our research efforts on neutron reflectometry and surface diffraction have clearly demonstrated the ability to distinguish between detailed models of interfacial polymer structures between lamellae microdomains and the feasibility of grazing-angle neutron diffraction with imperfect crystals.

- A new model has been developed which describes the critical role of diffusion and rearrangement of oxygen and metal defects in the evolution and movement of twin boundaries which are a key to the microstructural and related mechanical and electrical properties of 1,2,3 superconductors.

- New results in our application of maximum entropy procedures to macromolecular crystallography clearly suggest that this approach provides a powerful technique for extension and refinement of phases.

- A new capability for studying complex fluids under high shear rates has been developed in cooperation with the Thermophysics Division and successfully used to study shear-induced melting of colloidal crystals.

- A new cooperative effort has been initiated with Sandia Laboratories to develop and apply high resolution cold neutron spectroscopy to the study of macromolecular and microporous materials. Sandia will support instrumentation development and provide cooperative scientific staff.

- Antiferromagnetic long range order in two dimensions was found for the first time in multilayer specimens of alternate layers of magnetic semiconductors ZnSe and Mn Se.

- In the ceramic high $T_c$ superconductors, the effect on $T_c$ of charge transfer from chain to plane sites was analyzed using samples with oxygen deficiencies and atomic substitutions. A magnetic ordering enhancement in Co samples was also found.
ORGANIZATION CHARTS
AND
RESEARCH STAFF
REACTOR RADIATION DIVISION
J. M. Rowe, Chief
T. M. Raby, Deputy
H. J. Prask, Sci. Ass't
E. C. Maxwell, Admin. Officer
S. L. Neal, Admin. Asst.
E. S. Altman, Sec'y

REACTOR OPERATIONS & ENGINEERING
T. M. Raby, Chief
J. F. Torrence, Deputy
J. H. Nicklas, Chief Eng.
G. C. Harrison, Sec'y
C. M. Sweeney, Sec'y

COLD NEUTRON PROJECT
J. M. Rowe, Manager
I. G. Schroder, Tech. Leader
C. L. O'Connor, User Coord.
N. P. Repetti, Sec'y

NEUTRON CONDENSED MATTER SCIENCE
J. J. Rush, Leader
J. K. Spillman, Sec'y
L. K. Clutter, Sec'y
Research and Engineering Staff

N. F. Berk
- Condensed matter theory
- SANS theory for microstructure analysis
- Computer software for graphics and data analysis

R. C. Casella
- Theory of neutron scattering from light-atom defects in metals
- Group theory analyses of neutron scattering from condensed matter
- Elementary particle theory, especially as related to reactor generated experiments

R. Conway
- Electronic engineering
- Nuclear instrumentation

R. W. Erwin
- Magnetic materials
- Phase transformations
- Spin echo techniques
- Cryogenics

C. J. Glinka
- SANS microstructure of metals and porous media
- Magnetic materials
- Cold neutron instrument development

J. Gotaas
- Low temperature phase transformation
- SANS microstructure studies
- Magnetism

V. Himes
- Crystal database development
- X-ray crystallography

P. A. Kopetka
- Mechanical engineering
- Cold Source design
- Electro-mechanical systems

S. Krueger
- Small angle neutron scattering instrumentation
- Microstructure of materials
- Biological problems

J. LaRock
- Mechanical engineering
- Neutron instrumentation design

H. Layer
- Electronics and data processing
- Advanced instrumentation
- Fundamental physics

C. F. Majkrzak
- Condensed matter physics
- Polarized neutron scattering
- Polarizing and monochromating devices

A. Mighell
- Crystallographic database development
- Single crystal diffraction
- Theory of crystal lattices
<table>
<thead>
<tr>
<th>Name</th>
<th>Research Interests</th>
</tr>
</thead>
</table>
| B. Mozer      | o Structure and microstructure of metallic glasses  
                 o Dynamics of liquids  
                 o NDE of alloys |
| D. Neumann    | o Two-dimensional materials  
                 o Solid state physics  
                 o Neutron and x-ray scattering instrumentation |
| J. H. Nicklas | o Mechanical engineering  
                 o Reactor fuel design  
                 o Reactor engineering support |
| D. Pierce     | o Mechanical engineering  
                 o Neutron instrumentation design |
| H. J. Prask   | o Residual stress measurement  
                 o Neutron NDE of hardware  
                 o Neutron NDE instrumentation |
| E. Prince     | o Structural properties of alloys, catalysts and minerals  
                 o Advanced crystallographic refinement methods  
                 o Software for materials structure analyses |
| T. M. Raby    | o Reactor operations  
                 o Nuclear engineering  
                 o Reactor standards |
| J. J. Rhyne   | o Properties and transformations of high technology magnetic materials  
                 o Structure of amorphous solids  
                 o Data acquisition and analysis system |
| J. M. Rowe    | o Orientationally disordered solids  
                 o Hydrogen in metals  
                 o Cold neutron research and instrumentation |
| J. J. Rush    | o Catalysts and molecular materials  
                 o Hydrogen in metals  
                 o Two-dimensional systems  
                 o Inelastic scattering methods |
| A. Santoro    | o Structure of electronic and structured ceramics  
                 o Theory of crystal lattices  
                 o Powder diffraction methods |
| S. Satija     | o Low-dimensional molecular systems  
                 o Fractal aspects of microporous media  
                 o Neutron reflectometry |
| I. G. Schroder| o Cold neutron instrumentation development  
                 o Nuclear and engineering physics  
                 o Optical devices for neutron transport |
F. Shorten  o Physical measurement
           o Safety

J. Stalick  o Neutron and x-ray diffraction
           o Inorganic chemistry
           o Crystal database development

J. Sturrock o Design engineering
           o Nuclear systems and components

M. Suthar  o Design engineering
           o Nuclear systems and components

J. F. Torrence o Reactor supervision
               o Reactor maintenance
               o Nuclear engineering

T. J. Udovic o Neutron time-of-flight instrumentation
               o Properties of catalysts
               o Spectroscopy of surfaces

Technical Staff

Reactor Operations

Richard D. Beasley
Myron H. Bell
Nathan A. Bickford
Mark G. Cassells
Forest C. Clark
Howard W. Dilks
Daniel J. Flynn
Larry T. Lindstrom
Michael J. McDonald
William W. Mueller
Thomas J. Myers
John H. Ring
Ricky P. Sprow
Robert F. Stiber
Attila L. Toth
Daniel P. Wilkison

Neutron Research

George M. Baltic
Donald H. Fravel
Wayne C. Knill
Hugh L. McConaghy
Michael J. Rinehart
William H. Rymes
Thuan Thai
Robert H. Williams

Reactor Engineering

James A. Beatty
Richard P. Hayes
DESCRIPTION OF TECHNICAL ACTIVITIES

The technical activities of the Division are summarized in this section. A more detailed description of each project can be found in the NIST Technical Note "NIST Reactor, Summary of Activities July 1988 through June 1989."
Reactor operation over the past year was uneventful and routine. The few unscheduled shutdowns were all due to interruption of outside electrical power and resulted in no loss of operating time. Accordingly, essentially 100% of all scheduled operating time was achieved. There was, however, considerable scheduled downtime for construction of the cold neutron facility aside from that regularly scheduled for refueling and maintenance. The extra shutdown time was used to accomplish overdue major maintenance and reactor modification and upgrade.

Among the major projects that were carried out was retreatment of the thermal shield cooling system. To date approximately one third of the 200 cooling tubes have been sealed using a unique method developed by the National Nuclear Corporation of England. Both the size of the leaks and their frequency have gotten progressively smaller with each application, and reactor operation personnel became proficient in the use of the method thereby reducing costs substantially.

As another step to assure long-term uninterrupted operation, we are in the process of procuring new heat exchangers. The first request for this generated limited response from vendors. The specifications are being revised and resubmitted with continued emphasis on long term reliable performance.

Backups were designed or installed to several major engineered safety systems to guard against extended shutdowns since the reactor cannot be operated without them. These include the emergency power supply systems, the reactor stack radiation monitoring system and the secondary coolant nitrogen-16 detection system. Other major improvements include a separate auxiliary and experimental cooling systems, a new fuel element design that will greatly enhance fuel utilizations (already the best in the country), and an advanced design of the reactor automatic control system.

The small operating and engineering staffs continue to carry out multiple duties including operations, maintenance, repair, testing, design, installation and quality control as well as providing significant technical assistance to users and experimenters. The NIST reactor is the only reactor in the U.S. to have its entire operating staff hold a senior level reactor operator license.
NEUTRON CONDENSED MATTER SCIENCE

1. Microscopic Properties of Chemical Materials
   R.C. Casella, D. Neumann, T. Udovic, J.J. Rush, J. Nicol¹, and
   S.F. Trevino²

¹Guest Scientist, University of Maryland
²Guest Scientist, Army Armament Munitions and Chemical Command

Catalysts and H in Metals

A variety of areas were pursued in our research on the vibrational dynamics and molecular motions of hydrogenous adsorbates/absorbates on and in technologically interesting materials.

A study (with the Center for Analytical Chemistry) of trimethylsilyl ((CH₃)₃Si-, TMS) species anchored to surface oxygen on silica, which constitute a bonded alkyl phase used in reversed-phase liquid chromatography, has revealed TMS and methyl torsional vibrations of 2.5 and 20 meV. Scattering data also indicate surprisingly rapid TMS rotational motion (around the Si-O axis) on a neutron timescale (>10¹⁰ rot./sec) even at 80K, with methyl rotational motions already virtually frozen out. Further hydrogen adsorption studies have been carried out (with Auburn U) on RuS₂, a catalyst for hydrodesulfurization (HDS) reactions, to determine the origin of the complex peak shape of the Ru-S-H bending modes. The dissociative adsorption of HD onto the S-S anion pair sites without significant isotopic mixing with neighboring sites yielded the same Ru-S-H vibrational peak structure as for H₂ adsorption, indicating that the complex shape is due to a distribution of bending mode energies rather than a dispersion effect induced by dynamic coupling interactions between hydrogens on the surface. Spectroscopic studies of a model HDS compound, thiophene (C₄H₄S), adsorbed on RuS₂ are also in progress.

In our research on adsorbed species inside porous frameworks, we have continued our investigations with zeolites and analogous molecular sieves. Studies of the interaction of molecular hydrogen with exchangeable cations in zeolites (with LASL and Imperial Chem. Ind.) have been extended to H₂ adsorbed in ZnNa-A zeolite. Scattering data reveal two transitions at 4 and 8.4 meV. The temperature dependence of the peak intensities and frequency shifts are being analyzed to establish their origin as rotational or vibrational transitions of the H₂ molecules interacting with the framework cations, along with the strength and symmetry of the force field. As another example, studies (with UCSB) of p-nitroaniline (p-NA) adsorbed in the molecular sieve ALPO-5, a novel nonlinear optical material, have indicated a concentration-dependent vibrational spectrum. Spectra at 3 and 13 wt % p-NA, representing the onset of and the maximum in the nonlinear optical coefficient, respectively, display differences in the regions of 30-60 and 100-150 meV, which arise from differences in the alignment, ordering, and hydrogen bonding of the p-NA molecules inside the confines of the ALPO-5
one-dimensional channels. It is believed that the nonlinear optical properties are related to the alignment of the p-NA molecules as ordered chains stabilized by hydrogen bonding inside these channels. Further neutron studies of the dynamics and structure of this system vs. concentration are planned using quasielastic scattering, SANS, and diffraction techniques.

In our hydrogen-in-metals research, we have continued our investigation (with Paul Scherrer Institut and Ames Lab) of $\alpha$-Sc$H_x$ $(x<0.33)$, a solid solution of tetrahedrally-bound hydrogen inside hcp scandium which is stable down to very low temperature. Similar to $\alpha$-YH$_x$, the H vibrational mode along the c-axis is both substantially lower (30%) in energy and intrinsically much broader than the basal plane modes, and possesses considerable anharmonicity as reflected by the ratio (1.76) of the second-to-first-excited-state energies. Unlike the c-axis doublet previously observed for $\alpha$-YH$_x$, high-resolution spectra of the c-axis peak as a function of H concentration reveal no observable splitting due to H-pairing (even at the highest measured concentration, $x=0.25$), even though the overall peakwidth for $\alpha$-ScH$_x$ is broader than for $\alpha$-YH$_x$. This suggests that the local ordering of H pairs along the c-axis is less extensive than in $\alpha$-YH$_x$.

Quasielastic neutron scattering measurements have been performed on ScH$_x$ to probe the transport between nearest neighbor tetrahedral sites along the c-axis, which since out-of-chain motion is much slower, provide an interesting analogue of a double-well system in an otherwise pure metal. The temperature dependence of the measured hopping rates shows a striking minimum near $T=100$ K, with a power law rise at low temperatures. Kondo has predicted such behavior for tunneling defects in metals as a result of weak coupling to the conduction electrons. Our attempts to achieve a consistent analysis of the quasielastic data using available theory have not yet been satisfactory, and proper theoretical treatment of local site asymmetry, as well as accounting of defect-defect interactions, may be required to fully interpret these systems.

Finally considerable progress has also been made in our cooperative research with KFA, Jülich and ILL on the study of H potentials and unusual multiple site occupancies in Nb$_x$V$_y$H$_z$ alloys over a wide range of concentrations, phases and temperatures.

**Two-dimensional and Layered Materials**

During the past year, we have continued our research on the structure and dynamics of intercalation compounds of both clays and graphite in an attempt to elucidate details of the guest-guest and the guest-host interactions.

Clays are a naturally occurring class of expandable layered silicates in which the two-dimensional oxyanions are separated by layers of intercalated cations. Permanent porosity with a wide range of pore sizes can be induced in these minerals by exchanging large cations, such as tetramethylammonium,
for the naturally occurring ions, e.g. Na. The resulting material is known as a "pillared clay" and the cations which prop the layers apart are referred to as "pillars". In collaboration with scientists from Michigan State University and the Colorado School of Mines, we have undertaken a series of experiments using the BT4 filter analyzer spectrometer and the cold neutron time-of-flight spectrometer in order to elucidate details of the clay-pillar interaction thereby providing a crucial first test of current models of the template potential within the gallery of a clay. Our results have yielded important information on the orientation of pillars within the galleries, on the overall torsional motions of the pillars, and on the torsional modes of the methyl groups within individual pillars which have proven to be very sensitive to the surrounding environment. In addition, we have discovered that inelastic neutron scattering provides a very sensitive test of the octahedral site occupancy within the clay itself via the OH librations. This information, when combined with measurements of the ion exchange capacity and a simple chemical analysis of the metal ions which are present, allows the accurate determination of how much of the fixed charge on the clay layer is due to substitutions on in the tetrahedral sites which causes a disruption of the average template potential and how much is due to substitutions on the octahedral sites which cause little such disruption. The understanding of this distinction and it role in the pillaring process may be crucial for the creation of regular arrays of pillaring ions necessary for fabricating highly selective catalysts, catalyst supports, and molecular sieves. We have also performed elastic scattering experiments which have shown that the ion-exchange may not be complete in these systems, a fact which, if verified, would create serious doubts about the ability to form a regular array of pillars in these materials.

Work has also continued on the low temperature rotational properties of the ammonia molecules in K-ammonia intercalated graphite in collaboration with scientists from the University of Illinois and the Institut Laue-Langevin. Here recent results on a sample intercalated with deuterated ammonia have shown that neither a six-fold nor a three-fold potential is capable of explaining the data. Work is continuing in order to find a satisfactory description of the low-temperature rotational potential in these materials. We are also just beginning work on the Cs-ammonia system in order to determine the origin of the phonon-libron coupling observed in the K-ammonia compound. Some current models depend on the fact that the K ion is small compared to the gallery height. This is not the case for Cs thereby allowing one to distinguish between the competing models.
2. Neutron Scattering in Superconducting, Layered, and Amorphous Magnetic Materials


1University of Notre Dame
2Consultant, University of Maryland
3Guest Scientists
4Visting Professor, University of Illinois

Multilayer Magnetic Systems

Recent techniques for growing epitaxial rare-earth superlattices and films have afforded a unique opportunity to study the interactions responsible for the complex magnetic orderings in these materials. In particular, it has been possible to measure the range, strength and nature of rare-earth exchange interactions by interleaving dissimilar layers of rare earths with and without local magnetic moments, thereby fine-tuning the already well developed theoretical framework for rare-earth magnetism.

We have been focusing (with U. of Illinois scientists) on investigations of the role of magneto-elastic energetics in the magnetic structure. These energetics are modified by the constraints imposed by epitaxy of the magnetic lattice to nonmagnetic interlayers and the substrate. The magnetic structure of Dy is a basal-plane helix modulated along the c-axis of the hcp structure. This structure is also found in the films and superlattices. This order is thus described by the single parameter \( \omega \), the precession angle of the basal-plane moments from one atomic plane to the next. In bulk Dy, the discontinuous decrease of the magnetoelastic energy at \( T_C \) drives the transition by overcoming the exchange energy difference between the ferromagnetic and helimagnetic states. The \( \gamma \)-mode associated with an orthorhombic distortion is the part of the magnetoelastic energy primarily responsible for the ferromagnetic transition, and this magnetoelastic energy can only be gained when \( \omega = 0 \) (the clamping effect). In the superlattices and epitaxial films, the interaction with the substrate and non-magnetic layers inhibits the \( \gamma \)-mode magnetostriction and suppresses the ferromagnetic transition. The energy difference between the helimagnetic and ferromagnetic states in the superlattice is calculated to be roughly 2 K/atom at 10 K, compared to the 4 K/atom jump at \( T_C \) in the bulk. This difference corresponds to a critical field of 2 K/atom \( \approx 3.5 \) kOe on 10 \( \mu_B \).

In a related area, the dependence of the magnetic structure on growth direction has been investigated in superlattices made from alternating layers of Dy and Y metals (again, with U. of Illinois). The Illinois group has grown high quality single crystal materials with structural coherence lengths > 500 Å and with the growth direction along the a, b or c crystallographic axes. In earlier work we showed that, in c-axis samples, Dy layers order coherently via RKKY-like interactions propagated through Y layers up to about 150 Å. Diffraction scans on multilayers in which the b crystal direction was parallel to the growth direction revealed the existence of helical order below the Neel temperature. The width of the magnetic peaks, however, confirmed that the order was confined to a single
Dy layer with no long-range propagation, in contrast to the ordering observed in the c-axis multilayers. This remarkable result finds its explanation in the anisotropy of the conduction electron susceptibility.

We have also begun neutron diffraction studies of new diluted magnetic semiconductor (DMS) films and superlattices. Molecular beam epitaxy (MBE) makes possible entirely new DMS systems that do not form in the bulk. One of the latest achievements has been the growth of zinc-blend (ZB) type Cd$_{1-x}$Mn$_x$Te epilayers on (100) GaAs substrates. In samples with $x = 0.70$ and 0.75 we have observed additional diffraction maxima at low temperatures clearly indicating the Type III antiferromagnetic structure (AF III). This ordering, which is associated with doubling of the unit cell cube axis, also occurs in two other alloys from the A$_{II}$B$_{VI}$ DMS series, Cd$_{1-x}$Mn$_x$Te and Zn$_{1-x}$Mn$_x$Te, which form bulk zinc-blend type crystals. However, in contrast with the short-range nature of the spin correlations seen in those materials, data from Cd$_{1-x}$Mn$_x$Te epilayers indicate long-range ordered AF III domains. Such domains have the doubling direction parallel to the epilayer surface, whereas for the third domain orientation the order remains of short range. This anisotropic behavior is not yet explained.

Another example of new DMS systems are ZnSe/MnSe multilayers. The MnSe and ZnSe layers both grow in the ZB structure, although bulk MnSe forms only NaCl-type crystals. Superlattices were studied containing ZnSe/MnSe bilayers MnSe layers. Nuclear diffraction patterns from the multilayers exhibit a number of well resolved satellites from the GaAs substrate, reflections, in good agreement with theory. At low T we have observed additional peaks, indicating an AF III spin structure in the MnSe layers. However, in contrast to the behavior observed in the Cd$_{1-x}$Mn$_x$Te epilayers, the tetragonal axes of the domains are oriented perpendicular to the layers. The magnetic reflections were strongly broadened in the growth directions, showing that the AF III order does not propagate through the non-magnetic ZnSe layers, consistent with short-range nature of the exchange interactions in a semiconducting crystal.

**Magnetic Structures of Alloys and Compounds**

As nonmagnetic Y is substituted for Tb, the cubic intermetallic compound TbAg changes from a conventional antiferromagnet into a complex magnetic system with components of both commensurate and incommensurate spin structures and magnetic properties typical of spin glass behavior for Tb<0.5. We have carried out (with U. of Chicago and ANL) diffraction experiments on Tb$_x$Y$_{1-x}$Ag with $x = 0.15$, 0.5, and 1.0 to complement our earlier studies for $x = 0.3$. For $x = 1.0$, a ($\pi \times 0$) antiferromagnetic structure is observed as in many CsCl-type RA$\bar{g}$ (R = rare earth) compounds, with ferromagnetically aligned [110] planes of moments which are antiferromagnetically coupled. $T_N$ is found to be approximately 106 K, and there is no evidence for a change in magnetic structure on cooling. For $x = 0.5$, the system orders at $T_N \approx 58$ K into the mixed structure observed for $x = 0.3$, with a peak corresponding to the commensurate ($\pi \times 0$) magnetic structure at the (1/2, 1/2, 0) position and
satellites arising from an incommensurate modulation of the basic structure. In this case, however, the (1/2, 1/2, 0) peak is much sharper, and on cooling below 40 K the incommensurate peaks almost disappear. For x = 0.15, the system orders at \( T_N \approx 18 \) K into the two component structure, but with the (1/2, 1/2, 0) peak much broader than the incommensurate satellite peaks, indicating that the commensurate spin correlations are an order of magnitude shorter than that of the modulated structure. The two components have an approximately constant intensity ratio and show no evidence of hysteresis, similar to the behavior found for \( x = 0.3 \). Thus, as the Tb concentration decreases, the relative stability of the commensurate and incommensurate structures appears to change.

We have also recently studied (with NSWC) the magnetic ordering in a single crystal of \( \text{Tb}_{0.5}\text{Dy}_{0.5} \). The rare earth elements Tb and Dy both order magnetically with decreasing temperature into a basal plane helical state, followed at lower temperatures by a transition to a basal plane ferromagnetic state. In the low-temperature ferromagnetic phase, the b-axis is the magnetic easy axis for Tb, and the a-axis is the easy axis for Dy. In the mixed alloy system, the competition of the different basal plane anisotropies leads to temperature-dependent changes in the easy-axis direction. Measurements were performed with the b-axis oriented perpendicular to the scattering plane, primarily on the (0002) peak and the satellites observed along the c-axis in the helical phase. On cooling from the paramagnetic phase, the crystal orders into the helical phase with magnetic peaks observed at \( (0002 \pm \delta) \) at \( T_N \approx 206 \) K. Near 170 K, second harmonics begin to appear at \( (0002 \pm 2\delta) \), arising from bunching of the moments prior to the ferromagnetic ordering at \( T_C \approx 152 \) K. At this temperature, there is an abrupt reduction in the satellite peaks at \( \delta \), while the harmonics at \( 2\delta \) effectively vanish, as the ferromagnetic intensity increases sharply at the (0002) position. We also observe a Lorentzian peak underlying the (0002) which appears as the moments order into the helical phase, whose intensity peaks at \(-152 \) K. This suggests the presence of critical scattering in the helical-ferromagnetic phase transition, which would be quite unusual. Work will continue to clarify this situation and to study expected changes in the alignment of moments in the ferromagnetic phase.

As a final example, neutron scattering experiments (with CNRS and BNL) were used to study the magnetic state observed at low temperatures in the icosahedral phase of the alloy \( \text{Al}_{73}\text{Mn}_{21}\text{Si}_6 \). Measurements were performed on the BT-1 powder diffractometer, at room temperature and below the magnetic transition to observe any changes in the relative intensities of the diffraction peaks. Preliminary data analysis indicates no significant changes in the peak intensities associated with the magnetic ordering nor any evidence of new peaks due to a possible magnetic transition. Polarized neutron experiments were then performed on the triple-axis spectrometer H8 at BNL. Spin-flip measurements were taken between 9 K and 150 K at different wave vectors from 0.40 to 2.84 \( \text{A}^{-1} \). The observed form factor is the same as that observed previously for \( \text{Al}_4\text{Mn} \) but very different from that for \( \text{Mn} \) in alloys or compounds. The higher temperature form factor is much different
than would be expected for a paramagnetic scatterer and still seems to show some magnetic ordering.

Magnetism and High Temperature Superconductors

A complete understanding of high temperature superconductivity must include the magnetic properties of the Cu oxide based superconductors, which are either superconducting or antiferromagnetic depending on the composition. An important tool for probing the basic physical properties, in particular the magnetic properties, of these systems is the isomorphous substitution of magnetic ions such as Co for Cu. Previous work has shown that Co preferentially occupies the "chain" site in these materials and that two antiferromagnetic transitions exist for \( y = 0.45 \) corresponding to magnetic ordering first on the "plane" sites followed by ordering on the "chain" sites as well as the "planes". At higher oxygen concentrations the samples become superconducting. Thus, this system exhibits all of the essential magnetic and superconducting features of the parent compound \( \text{YBa}_2\text{Cu}_3\text{O}_{6+y} \).

We have carried out (with Bellcore) neutron scattering studies to determine the oxygen dependence of these transitions in \( \text{YBa}_2\text{Cu}_{2.8}\text{Co}_{0.2}\text{O}_{6+y} \).

Powder diffraction measurements allowed the determination of the 0 content to \( \pm 0.02 \). The magnetic structures were determined using triple-axis spectrometers. The oxygen dependence of the two magnetic phase transitions was determined from the temperature dependence of the whole and half integer \( l \) reflections. Both transition temperatures increase with decreasing oxygen content until \( y = 0.32 \) where both chains and planes order at a single temperature. We have also performed a similar experiment on a sample with a Co concentration of 0.8 which displayed only half-integer \( l \) peaks for all 0 concentrations. The effect of adding Co is to raise the chain site ordering temperature \( T_{N1} \) until \( T_{N1} = T_{N2} \). These results provide a direct test for theoretical predictions of the electronic and magnetic interactions in these materials.

We have also studied the magnetic properties of the rare earth ions \( R \) in the \( \text{RBa}_2\text{Cu}_3\text{O}_7 \) superconductors, which are of interest because the superconductivity is not significantly affected by the substitution of several rare earths. The \( \text{RBa}_2\text{Cu}_3\text{O}_7 \) materials order antiferromagnetically, where the system is orthorhombic and superconducting. The magnetic interactions are weak, yielding ordering temperatures \( (T_N - 1 \text{ K}) \) well below the superconducting transition temperature \( (T_C - 293 \text{ K}) \), making these materials good examples for investigating the competing interactions of superconductivity and magnetism. The first neutron experiments (University of MD, University of Cal., NIST) on \( \text{ErBa}_2\text{Cu}_3\text{O}_7 \) powder showed a magnetic phase transition at \( T_N = 0.6 \text{ K} \) with specific heat. The Er moments exhibited a two-dimensional character in the a-b plane, while there were no significant magnetic correlations detected along the c-axis down to the lowest temperatures \( (0.33 \text{ K}) \). Recently we have made measurements on a high quality single crystal. Above the 3-d ordering temperature \( (T_N = 0.618 \text{ K}) \)
K) we observe a rod of scattering along \((1/2,0,\ell)\) which demonstrates the expected strong anisotropy of the magnetic interactions and the concomitant 2-d character of the system. Scans through the rod have a width which is limited by the instrument resolution, while the scan along the rod shows no significant spin correlations between adjacent a-b layers. Below the 3-d Néel temperature we find that two magnetic structures can occur at low \(T\), one characterized by a wave vector \((1/2,0,0)\) and the other by \((1/2,0,1/2)\). The spin structure for the \((1/2,0,1/2)\) configuration consists of ferromagnetic chains of spins along the b direction, with adjacent chains in the a-b plane coupled antiferromagnetically. Along the c-axis the spins are antiparallel. We believe that the \((1/2,0,1/2)\) structure represents the true ground state configuration. Both previous specific heat and neutron scattering reveal that the ground state is a doublet, so that \(S = 1/2\) is appropriate. In addition, in an orthorhombic structure with such a low Néel temperature we might expect the anisotropy to be large compared to the magnetic interactions, thus defining a unique spin direction. This corresponds to a 2-d Ising model, which is the only one exhibiting long range order in 2-d. A fit to the Onsager’s exact solution for \(S = 1/2\) 2-d Ising model \([9]\) provides an excellent description of the data. Thus, we believe that the physics of the rare earth ordering in these materials is controlled by the 2-d like character, with the 3-d ordering being induced as a consequence of the onset of long range order within the a-b planes.

The paramagnetic-antiferromagnetic ordering temperature of the \(\text{RBa}_2\text{Cu}_3\text{O}_{6+x}\) (\(R = \text{rare earth}\)) compounds is sensitive to the oxygen concentration x in the chain layers which is also sensitive to pressure. As a final example, we have studied (with the U. of Maryland and Army scientists) the magnetic order of the Cu spins in two single crystals of composition \(\text{NdBa}_2\text{Cu}_3\text{O}_{6.35}\) and \(\text{NdBa}_2\text{Cu}_3\text{O}_{6.1}\) as a function of pressure. At zero pressure the ordering temperature \(T_{N1}\) increases linearly at a rate of \((23 \pm 3)\) K/kbar up to an applied pressure of 4 kbars. This rate is more than two orders-of-magnitude higher than the rate of \(0.05\) K/kbar observed for the superconducting transition temperature \(T_c\) for \(\text{YBa}_2\text{Cu}_3\text{O}_7\). The pressure dependence of the Bragg scattering associated with the lower transition temperature \(T_{N2}\), where the Cu chain spins order, shows only a small shift in \(T_{N2}\). We believe that this weak pressure dependence is due to the fact that when the chain ions order, the spacing between the Cu ions in the a, b, and c directions is about equal and we have a fully 3-d magnetic structure with simple antiferromagnetic near neighbor interactions, in contrast to the situation at \(T_{N1}\).
3. Neutron Diffraction Methods and Applications
E. Prince, A. Santoro, J. Stalick, B. Mozer, C. Choi, H. Prask and F. Beech

1 Guest Scientist, University of London

During the past year, structure and modelling studies of high $T_C$ superconductors, including work on new classes of these materials and the effects of metal and oxygen defects, have been the largest single component of our crystallographic research. Studies on superconductor type materials with substitutional magnetic atoms are discussed above. Diffraction efforts have been focused on several new "classes" of high $T_C$ materials. Work with AT&T Bell Labs on $\text{Pb}_2\text{Sr}_2\text{LnCu}_3\text{O}_8$, a prototype of a family of superconductors with a complex (Sr,Pb,Cu,0) block separating electronically active CuO$_2$ pyramidal layers, has revealed the origin of the orthorhombic distortion as the irregular coordination requirement of the Pb$^{2+}$ ion within its square pyramidal oxygen environment. Another study with Dupont on thallium-based (Tl$_2$Ba$_2$CuO$_6$) superconductors has provided structural information which addresses possible models to explain the observed superconductivity (the basic composition would not be expected to superconduct). Our results show that neither Tl-vacancies nor tetragonal symmetry are necessary conditions for superconductivity in this material and suggest that oxygen non-stoichiometry is a key in producing superconductive behavior. Similar research is underway (with Los Alamos and U. of Grenoble). $\text{LaTl}_{1-x}\text{Sr}_x\text{CuO}_4$ materials of another new class of superconductors whose complex tetragonal structure is a combination of $\text{K}_2\text{NiF}_{4}$-type and Nd$_2$CuO$_4$-type blocks. As another part of the superconductor effort, a model has been developed which can describe the critical role of rearrangements and diffusion of oxygen and metal defects in the evolution and movement of twin boundaries in 1,2,3 type superconductors. The extent and behavior of twins is a central factor in determining the microstructure of these materials, and thus directly impacts their mechanical and electrical properties. This model, which can reproduce all existing structural and microstructural observations and crystal-chemistry considerations, can be a valuable aid in interpreting and predicting these properties.

Another area of crystallographic research has involved the application of our previously developed maximum entropy procedures to phase refinement in macromolecular crystallography. Recent work (with Chalmers Institute and NRL) has yielded promising and exciting results on a ribonuclease A complex and two other macromolecules, which have clarified important structural features, e.g., by providing a clear picture of uridine vanadate complex (in ribonuclease). The results so far suggest that this maximum entropy approach could be a very powerful technique for extention and refinement of phases. Work has also been continued with MIT on the ceramic fast ion conductor system $\text{Y}_2(\text{Zr}_x\text{Tl}_{1-x})_2\text{O}_7$, a series of compounds with ionic conductivities ranging over two and one-half orders of magnitude. Attention
has been focused on superlattice intensities which track the anion disorder associated with conductivity changes in the florite type structures. Research on molecular systems include studies (with MIT) of the structure of metal-dichloride graphite intercalation compounds and of highly complex energetic (explosive) molecules (Army, U. of Maryland). In another area, work on metal and alloy systems has included a detailed study (with ORNL and U. of Missouri) of the martensitic transformation in sodium metal and continuing studies (with the Metallurgy Division, CNRS, Saclay and Iowa State) on the structure of new alloys with a stable icosahedral phase. Finally, we have made a substantial improvement in the treatment of background contributions in Rietveld Structure refinement, including the use of Chebychev polynomials for more accurate description of Q dependence of the background. These improvements considerably enhance the ability to probe site occupancies of substitutional elements and to extract contributions from up to 5 impurity phases.

Aside from crystallographic research, applications of neutron diffraction residual stress and texture measurements have been extended (with Army scientists) to a number of materials problems. Bulk texture studies have been extended in several areas: for texture/mechanical-properties correlations in cold-worked depleted-uranium alloys (with Battelle Labs); to confirm the effectiveness of magnetic orientation of high-T_c superconductors (with the Ceramics Division); and to characterize new shaped-charge liners prior to performance testing (with LANL). Residual stress studies of the failure region of 7075-T6 Al nose pieces ("ogives") for the M483 155mm projectile continued. Earlier examination of end-items from two manufacturers--one of which produces high fail-rate ogives--identifies residual stress differences consistent with the failure mode. New studies of the stress distributions in ogives from the high fail-rate group before and after a possibly critical cold-sizing step have been completed. Efforts to characterize sub-surface stresses in highly-textured samples has continued. A new program (with the Fatigue Design and Evaluation Committee of the Society of Automotive Engineers) has been initiated to evaluate sub-surface stresses in a steel axle fatigue-test specimen. Also, initial measurements to determine the role of whisker size and processing conditions on mechanical properties of alumina-SiC composites have been completed (with Ceramics Division and U. of Missouri).

NIST Crystal Data Center

Alan D. Mighell and Vicky L. Himes

The NIST Crystal Data Center is concerned with the collection, evaluation and dissemination of data on solid-state materials. The Data Center maintains a comprehensive database with chemical, physical and crystallographic information on all types of well-characterized substances. These materials fall into the following categories: inorganics, organics, organometallics, metals, intermetallics, and minerals. The data are made available to the scientific community through computer oriented modes of dissemination and through printed products. Two major database are produced: NIST CRYSTAL DATA and the NIST/Sandia/ICDD Electron Diffraction Database. These databases have many uses including materials design (e.g.,
During the year, work in the Data Center has focused on the following areas:

a) the development of a new analytical tool by integrating the Electron Diffraction Database into the analytical electron microscopes that are marketed by several different commercial companies; b) the initiation of technology transfer procedures for the development and implementation of new algorithms for phase identification based on matrix analysis; c) the expansion of the database and the development of new procedures so that biological and organic materials can be identified using limited powder data; d) the publication of a new book for phase identification ("Elemental and Interplanar Spacing Index"); e) the design of new algorithms and software to be used in the automation of diffractometers with respect to accurate symmetry determination; f) the production of a Crystal Data CD-ROM and accompanying software; g) the preparation of the annual update to the master database (currently 142,000 entries); and h) the preparation of 1000 new entries for the forthcoming Minerals Crystal Data Book.

As the databases now contain large collections of evaluated data, the development of efficient dissemination methods is critical. Recently, it has become apparent that three principal methods for the computerized delivery of scientific data are rapidly evolving. All three are now used by the Data Center. The first is a desk top method in which the database on CD-ROM is used in conjunction with a PC-computer. The second is the incorporation of the database into a scientific instrument where the combination represents a new analytical tool. The third is the distribution of data through international online systems.

The online method permits the Data Center to provide rapidly the general scientific community with up-to-date data and with extensive software tools to retrieve and manipulate the information. To build and maintain the online system in the most efficient manner, the National Institute of Standards and Technology (US) and the National Research Council (Canada) have formally agreed to pool their resources. The objective is a longterm collaboration not only to broaden the scope of the existing CRYSTDAT System (=online version of NIST CRYSTAL DATA) but also to expand the system to include materials databases in related fields.

**Autoradiography Study of Paintings**

F. J. Shorten, Y. T. Cheng and J. S. Olins

Research Associate, Smithsonian Institution

This past year the first phase of autoradiography study on paintings of Thomas Wilmer Dewing has been completed. A manuscript titled "Thomas Wilmer Dewing: An Ebauche or Sketch of His Painting Technique" has been prepared and accepted by the "Smithsonian Studies in American Art" for publication. The work comprises the study on 11 of Dewing's paintings and is the result of a close collaboration among scientists, art historian and painting conservators. The autoradiographs suggest the technique that the artist employed in his creative process by registering brush marks, pigment distributions and form changes. Extensive literature research and personal
interviews have also contributed to the interpretation of the autoradiographs. They often provided information about changes in the painting which were subsequently revealed in the autoradiographs. Dewing varied his paint applications from picture to picture, blending his individual style with a repertory of images that he drew from diverse sources. We find that throughout his career, Dewing continued to employ the classical ebauche that he had learned while a student in Paris. This technique ordained the method whereby he laid in his work, establishing its light and dark passages which were clearly revealed by the autoradiographs.

Work to examine paintings of another American artist Albert P. Ryder by neutron activation autoradiography continued. Thus far, 10 of his works from the collections in the National Museum of American Art (NMAA), including his master work "Jonah", have been studied. In addition, to more accurately analyze the radiographic information generated in the study, calibration references are being established. Finally, we have received funding of $100 K to upgrade shielding, irradiation and analysis systems to allow more timely autoradiography of more and larger paintings.

4. Materials Microstructure

Small Angle Neutron Scattering

C. J. Glinka, J. A. Gotaas, N. F. Berk, and J. LaRock

Both as a result of the improved measurement capability provided by the cold source and the continued shutdown of the SANS facility at Oak Ridge National Laboratory, demand for beam time on the 8 M SANS spectrometer was greater than ever during the past year. A wide range of experiments was performed, encompassing both traditional and novel applications of the SANS technique for characterizing the submicron structure of materials. The major areas of research were polymers (60%), chemistry (15%), ceramics (7%), metallurgy (4%), biology (5%), and magnetism (5%), where the percentages represent the distribution of beam time in each area. Here we summarize selected examples of these diverse research efforts.

Much of the SANS research effort by the Polymer Division focused on various aspects of the microstructure of polymer blends. C. Han and coworkers from Kyoto University and the Gen Corp. carried out a systematic study of the miscibility of polybutadiene blends (mixtures of 1,2- and 1,4-butadiene) which are widely used in the rubber industry because their properties can be tailored for various applications. In this system the use of deuterium labeling, which is essential for determining chain conformation in the blend, also has an effect on the miscibility limits. By using random copolymer theory in analyzing an extensive set of data, Han and coworkers were, for the first time in any blend system, also able to separate the effects of isotope labeling from those due to the interactions between each type of isomeric monomer in the chains. They were able to extract values for the individual interaction parameters between monomers and to predict specific chain microstructures that would favor miscibility.
Han along with A. Nakatani and H. Kim (Polymer Division) and Y. Takahashi (Nagoya University) are also studying the effects of shear on the phase separation of blends using a temperature controlled shearing apparatus that they designed and built for the SANS spectrometer. In measurements on blends of deuterated polystyrene and normal polybutadiene, they have observed that shear depresses the spinodal temperature. This finding supports certain theories that predict that shear can stabilize a blend in a region of temperature and concentration where it would normally phase separate, a result that relates directly to the processing of blends. In related work on blends, R. Briber and B. Bauer (Polymer Division) have mapped out the phase behavior of blends in which crosslinks have been introduced, either chemically or by gamma radiation, to determine how such geometrical constraints modify the phase separation conditions and the resulting microstructures.

The microstructure of model polymer networks, formed by crosslinking polymer chains exclusively at their ends, has been another focus of recent research initiated by the Polymer Division. W. Wu and L. Coyne (Polymer Division) have analyzed the structure of homopolymer networks formed from a population of chains with a bimodal distribution of chain lengths. Such networks are known to be considerably stronger than those formed from a unimodal distribution. Wu and Coyne observed for polydimethylsiloxane (PDMS) bimodal networks that the short and long chains, although homogeneously mixed before crosslinking, tend to segregate into molecular-sized regions in the fully cured state.

SANS research on polymers by outside groups included: a determination of the first-order character of the order-disorder transition in symmetric diblock copolymers by F. Bates and J. Rosedale (AT&T Bell Labs); an analysis of the growth and internal structure of latex particles by L. Sperling, S. Yang and J. Yu (Lehigh University); and a study of the effects of solvents on the structure of synthetic membranes by B. Abeles, J. Huang and M. Lin (Exxon Research and Engineering Co.).

A versatile couette-type cell for subjecting low viscosity fluids and suspensions to high shear rates has been designed and built for the SANS instrument by H. Hanley and G. Straty of the NIST Thermophysics Division. The apparatus has a dedicated computer for controlling the shear rate and cell temperature and will serve the growing interest in the microstructure of complex fluids and its relation to rheological properties. In their initial tests of the apparatus, Hanley and Straty observed the shear-induced melting of charge-stabilized colloidal crystals of spherical polystyrene particles suspended in water. They observed that low shear rates actually stabilize the orientational order in the suspension and that this hexatic order persists to rather high shear rates before finally "melting" to form a more liquid-like state.

The theoretical methods for analyzing multiple small angle scattering from micron-sized inhomogeneities in materials developed by N. Berk have been successfully applied by G. Long (Ceramics Division), S. Krueger (Polymer Division) and R. Page (Southwest Research Institute) to characterize the pore structure of alumina powder compacts at various stages of densification by sintering. They observed at intermediate densities (ranging from 50% to 90% of theoretical density) that while the number of pores decreases, their
average size remains fairly constant. At densities above 90%, however, the average pore size increases steadily as the porosity decreases indicating a transition from one type of sintering mechanism to another.

Other recent experiments that illustrate the diversity of applications that continue to develop for the SANS technique include: the first measurements of capillary condensation in a microporous glass as a function of vapor pressure by B. Abeles, J. Huang and M. Lin (Exxon R&E); measurements characterizing the aggregates that form in the sol-gel processing of high-Tc superconductors by G. Kordas, G. Moore and M. Salamon (University of Illinois); and the use of magnetized ferrofluid to align anisotropic biological macromolecules by J. Trewella, T. Sosnick (Los Alamos) and D. Gray (University of Houston).

Neutron Reflectivity and Grazing Angle Neutron Diffraction
C.F. Majkrzak, D. Neumann, S. Satija, J. Ankner

We have continued our measurements of neutron reflectivity on thin polymer films of diblock copolymers. A comprehensive study with IBM of interfacial structure of polystyrene, PS, and Polymethylmethacrylate, PMMA, block copolymers as a function of molecular weight and temperature has been completed. The power of the neutron reflectivity technique to differentiate between various detailed models of interfacial structure between the lamellar copolymer microdomains was clearly demonstrated. For example, a hyperbolic tangent function describes the interfacial structure most closely as opposed to a linear or a cosine squared function. In addition we performed neutron reflectivity measurements from a novel cell arrangement, in which neutron beam is passed through a large single crystal of Quartz (or silicon) and reflected internally at the quartz/liquid interface with the reflected beam coming out through the quartz on the other side. This technique opens up the possibility of study of solid-liquid interface in a variety of problems. For example we have studied the segment density profile (with the Metallurgy Division) of polymers adsorbed from solution onto a quartz surface as well as in situ density profile of deuterium in thin films of palladium in an electrochemical cell.

The possibility of measuring a (111) diffracted beam excited under conditions of total external reflection from the surface of (110) surface of a perfect silicon crystal was demonstrated last year. This experiment with the University of Illinois was the first demonstration of the grazing-angle diffraction technique using neutrons. We have subsequently studied kinematically-scattering Cr/Nb/sapphire films grown by molecular beam epitaxy (MBE) and thereby demonstrated the feasibility of grazing-angle neutron diffraction with imperfect crystals. We have now the added capability of using polarized neutron for reflectometry as well as grazing-angle diffraction. Using polarized neutrons we have determined the magnetization of the interface between Y(0001) and Gd(0001) single crystal films grown by MBE.
COLD NEUTRON PROJECT

The major goal of this project is the construction of a Cold Neutron Research Facility (CNRF) to meet National needs for cold neutron measurement technology. The CNRF comprises a network of eight totally reflecting neutron guides, all of which originate at the cold neutron source in the reactor, and seven of which extend into a large new experimental hall. Fifteen new instruments will be installed on these guides, and of these, 1/3 will be funded by groups not associated with the project. The construction of the buildings associated with this project (guide hall, office/laboratory, and compressor building) was completed, and the facility was dedicated by Secretary of Commerce William Verity on January 12, 1989. The neutron guide installation began in June with the boring of seven holes through the reactor confinement building wall. This part of the work was completed in August, and the actual guide installation was begun. For the first phase of the project, only three guides will be extended into the guide hall, allowing installation of the first complement of instruments. The beam that terminates in the existing confinement building will also be installed as part of the first phase of construction. A layout of the first stage of installation is shown in Figure 1.

Several instruments are at various stages of completion, and will be described in more detail below. As of September, commitments for instrumentation construction by non-NIST groups totals in excess of $3.0 M for three instruments, and several other agreements are being actively negotiated now, so that the project is close to meeting its commitments with respect to outside support. A new second generation cold source is being designed as part of the project, building on the experience gained from the successful installation and operation of the heavy ice cold source. This new source will utilize liquid hydrogen, and be cooled by a new helium gas refrigerator, which is in the final contracting stages and should be ordered early in the new fiscal year. The new refrigerator will have a maximum cooling capacity of 3.5 kW at 14 K, but will be capable of operating at lower capacity with high efficiency for interim use with the current source. Acquisition of this refrigerator will alleviate the operational problems that have been experienced with the current refrigerator, which is nearing the end of its useful lifetime. The new system should be built, installed, and available for use in October, 1990. The new source is being planned for installation at the same time as the four remaining guides, in eighteen months to two years in order to minimize radiation exposures. The hydrogen source will improve cold neutron intensities for all instruments by a factor of at least two.

The status of the instruments currently under active design and construction are summarized below. Details of the instrument parameters and designs can be found in the NIST Technical Note on the Summary of Activities in the Reactor Radiation Division, July 1988 through June 1989.

- Two 30 m small angle neutron scattering instruments are under construction, the NIST/Exxon and National Science Foundation (NSF) instruments. The former is in the final stages of construction and assembly in the guide hall, for operation by summer, 1990. The latter, which forms a major part of the Center for High Resolution Neutron Scattering (CHRNS) funded by a grant from NSF, will be essentially
identical to the former, and several components have been ordered. This instrument is expected to be operational within two years.

Two cold neutron triple axis spectrometers are being designed and constructed, with the first scheduled for installation in of 1990. The second will be similar to the first conventional triple axis in basic design, but will incorporate neutron polarization devices and a Drabkin flipper to achieve unique resolutions capabilities at high intensity. The latter instrument, the Spin Polarized Inelastic Neutron Scattering spectrometer (SPINS), is being constructed as a joint effort by the Cold Neutron project and CHRNS (NSF grant).

A very high resolution cold neutron time of flight spectrometer (TOF) is in the early stages of design and construction. The TOF will incorporate six or seven neutron choppers to allow a wide range of neutron energies and resolution for inelastic scattering and optimal intensity. This instrument is scheduled for installation in two years.

Two instruments for analytical chemistry are being constructed for installation in early 1990 as a collaboration between the Cold Neutron Project, the Center for Analytical Chemistry, and the Eastman Kodak Corporation. The first is a prompt gamma activation analysis facility that will be installed at one of the first guides, and will provide entirely new capabilities as a result of reduced backgrounds and increased neutron flux in the guide hall. Tests of the concept at the KFA in Jülich indicate at least a ten fold increase in sensitivity. The second is an improved depth profiling facility to be installed at the guide in the existing confinement building, which will improve signal to noise capabilities over the present thermal neutron facility by one to two orders of magnitude, and provide a broad range of opportunities for new types of measurements and applications for near-surface trace analysis.

Two experimental stations for research into the fundamental properties of the neutron are being developed in a collaboration between the Cold Neutron Project, the Center for Atomic, Molecular and Optical Physics, the Center for Radiation Research and several outside organizations. The first facility makes use of the neutrons at the end of a guide to perform various experiments, the first of which will be a measurement of the neutron lifetime. The second will be a neutron interferometer facility that will be used to explore the wave properties of the neutron, both to verify concepts of matter waves, and to measure other fundamental properties of matter, such as neutron scattering lengths.

A neutron reflectometer is being designed and constructed for installation in the guide hall by the summer of 1990 as a collaborative effort between the Cold Neutron Project, IBM and other outside organizations. This instrument was given a high priority as a result of a workshop held at NIST, and will permit study of surfaces and interfaces.

The existing 8 m SANS and TOF are being modified for installation in the guide hall in early 1990.
In addition to the instrument design and construction work, several development projects are underway in this Project, including the improvement of "supermirror" devices for use in instruments and possibly in guides, the development of new computer codes for instrumentation design and modelling, the testing and development of new shielding materials, and the development of new techniques for data analysis.
INDEPENDENT PROGRAMS

The three major independent (non-collaborative) NIST programs using the reactor are nuclear methods group in the Center for Analytical Chemistry, standard neutron fields for neutron flux calibration and materials dosimetry in the Center for Radiation Research, and the Fundamental Physics and Quantum Metrology Program of the Center for Atomic, Molecular and Optical Physics. Overviews of these programs are presented here. The major non-NIST independent programs were summarized in the RRD Annual Report.

1. Nuclear Methods Group

R. R. Greenberg

The development and application of nuclear analytical techniques for greater accuracy, higher sensitivity, and better selectivity are the goals of the Nuclear Methods Group. A high level of competence has been developed in both instrumental and radiochemical neutron activation analysis (INAA and RNAA). In addition, the group has the unique capability of using neutron beams as analytical probes for both prompt gamma activation analysis (PGAA) and neutron depth profiling (NDP). NDP determines elemental (isotopic) concentrations versus depth profiles within the first few micrometers of a surface by the energy analysis of the prompt charged-particles emitted during neutron bombardment. PGAA, on the other hand, measures the total amount of an analyte present throughout a sample by the analysis of the prompt gamma-rays emitted during neutron capture. These techniques (INAA, RNAA, PGAA and NDP) provide a powerful combination of tools to address a wide variety of analytical problems of great importance in science and technology.

The group has continued to contribute to the Standard Reference Material (SRM) certification effort; this year's efforts include multielement determinations performed on a number of SRMs including: Total Diet, Apple Leaves, Peach Leaves, Oyster Tissue, Coal, and Marine Sediment. Group members are serving as Technical Champions for many of these new SRM's, and as such are responsible for scientific decisions made throughout the production and certification processes of these materials. Additionally, RNAA measurements are currently under way to determine Hg in Bovine Serum and Se in Buffalo River Sediment. This will allow the certification of two additional, highly-important elements in these existing SRMs. A homogeneity study has been completed on the new Apple and Peach Leaves SRMs which showed that many elements were homogeneous to about 1% in 100 mg samples. In collaboration with members from other groups in the Inorganic Analytical Research Division, a study of these leaf materials has been undertaken to identifying problems in the dissolution of these materials which can lead to analytical errors during the certification analyses. A new ability to quantify nitrogen in biological samples will result from this year's research in developing an RNAA-liquid scintillation beta counting technique which exploits the thermal neutron reaction \(^{14}\text{N}(n,p)^{14}\text{C}\), and measures the activity of (radioactive) \(\text{CO}_2\) upon sample processing. This project is being done in collaboration with the University of Illinois.
The International Conference on Nuclear Analytical Methods in the Life Sciences, held at NIST from April 17-21, 1989, was organized by members of the Nuclear Methods Group. At this conference, 144 participants from 32 countries presented papers and posters detailing recent advances of nuclear analytical methodology in the life sciences. The conference pointed to the continuing, essential role of nuclear methods in the life sciences, and provided new emphasis for the group's research program in this field.

Recent applications of Neutron Activation Analysis (NAA) in the life sciences by the group include "nonclassical" detection and determination of "tags" associated with biological macromolecules such as proteins, enzymes, and antibodies. In one application, an activatable tag has been attached to an antibody used in a medical diagnostic test. The INAA determination of the activatable tag is then used as the detection step of the diagnostic analysis. Research is continuing on the polyacrylamide gel electrophoresis neutron activation analysis technique (PAGE-NAA). The activation and subsequent determination of phosphorus in phosphoproteins has already been demonstrated. Research to extend this technique to the determination of other elements in specific macromolecules is under way. Such advanced uses of NAA in the life sciences may open a wide variety of applications for nuclear analytical chemistry.

The Biomonitoring Specimen Bank Research Project continued its research support for other agencies' monitoring programs. These programs included the Environmental Protection Agency's human liver project, the National Oceanic & Atmospheric Administration (NOAA)'s National Status and Trends (NS&T) program, the NCI micronutrient program, the IAEA/NIST/FDA/USDA Total Diet Study, and most recently, the NOAA Alaska Marine Mammal Project. Research has centered on specimen banking protocols and improved analytical methodology. The group's participation in intercalibration exercises with the project participants and the development of QA materials for various marine analyses helped to enhance the quality of the analytical results used in the assessment of the environmental health of the nation. Of major importance this year has been the analysis of samples from 30 NOAA NS&T sites. Both essential and pollutant elements have been studied to assess the effects of anthropogenic inputs to the environment.

The joint NIST/FDA/USDA study of trace elements in human diet, sponsored by the International Atomic Energy Agency, has completed its fifth year. A total of 120 diets from different countries have been analyzed to date for minor and trace elements. Nine diets from different regions of the United States have also been collected, and thus far, six have been analyzed. The data obtained from the U.S. diet composites have confirmed daily intake results for some of the elements investigated by the FDA (based on individual food analysis), as well as established reliable daily dietary intakes for additional elements including B, Cr, Cs, Li and Sn. In the case of Li, the observed intake values are considerably lower than earlier published results. The group has taken an active role in the NIST program on high-temperature superconductivity research, both in the measurement of impurities in starting materials and final products, and the determination of the actual stoichiometry of the metallic constituents. The effort this year has centered on the development of accurate, rapid measurements of these materials by both NAA and PGAA. An effort to establish an accurate,
monitor activation analysis technique for further study of high temperature superconductors at NIST is under way.

Nearly a decade ago, a systematic error for PGAA was observed in which elemental sensitivities (counts/μg-s) increased with increasing hydrogen concentration. Sensitivities in hydrogenous samples have been observed to increase by up to approximately 25% relative to samples with little H. This error was handled previously by matching H concentrations in samples and standards without any real understanding of the problem. During the last year, substantial progress has been made in understanding the problem on a theoretical level. In addition, systematic, experimental solutions to this problem are currently under investigation. The sensitivity enhancement seems to be caused predominantly by neutron scattering. This scattering can increase the neutron path length within the sample for some sample geometries, and thus may enhance the probability of neutron absorption. Measurements have been made which demonstrate this sensitivity enhancement as a function of H concentration for 11 elements in samples of constant size and shape. In a second set of measurements made on a series of disk-shaped pellets with constant H concentration but varying thicknesses (shapes), elemental sensitivities were observed to increase with decreasing sample thickness. These general trends have been predicted by theoretical calculations. Our current understanding of this problem indicates that effects of scattering on sensitivities can be minimized or perhaps eliminated by irradiating spherical samples. A final series of measurements made on spherical samples with constant H concentration, and varying diameters (and thus masses), has indicated that the use of a spherical geometry greatly reduces, and perhaps eliminates, this problem.

A novel technique of PGAA analysis of gas samples has been developed using both the currently available thermal neutron beam at the NBSR and the ELLA Cold Neutron Facility at KFA, Jülich. Feasibility studies involving noble gases and gas mixtures indicate that sensitivities in the sub-ppm range for a number of elements should be achievable using the new NIST cold-source PGAA instrument which will be operational in FY 90. The development of a nuclear technique for gas analysis will establish an independent analytical method for reference material certification.

During the past year, a large part of the group's efforts has been directed at the exploitation of the analytical applications of cold neutrons. These efforts are needed to take full advantage of the guided beams of cold neutrons soon to be available at the new Cold Neutron Research Facility (CNRF). Plans are under way to include both PGAA and NDP experimental systems in the first phase of implementation. The group's involvement includes the design and construction of state-of-the-art instruments for both PGAA and NDP. Last year, the cold source itself was installed and became operational, and construction of the guide hall was completed during the first part of this year. The group's efforts have been aided substantially by its ability to use the existing cold neutron beams at the KFA Jülich facility. A major grant from the Eastman Kodak Company is contributing to the timely development of the NDP cold-neutron instrument. In order to introduce these new instruments to the potential user community outside of NIST, the Nuclear Methods Group organized a workshop on Analytical Measurements with Cold Neutron Beams, which was held May 22-23,
1989. This was the third in an ongoing series of workshops on the uses of cold neutrons organized by NIST.

Research on the new recoil-nucleus time-of-flight NDP technique has focused on the problem of detecting (with good energy resolution) the low energy heavy nuclei which are emitted along with the light nuclei used in conventional NDP. These heavy nuclei can be used to provide very high resolution depth profiling capabilities in the sample region very close to the surface. Initial measurements appear to have validated this approach, and efforts are under way to determine how closely the actual depth resolution measured will approach the calculated values.

A long-range program to explore and develop the analytical applications of focused beams of cold neutrons has been initiated within the group. The ultimate goal of this research is to produce beams of neutrons which have intensities several orders of magnitude greater than previously available. Such beams will greatly enhance the capabilities of both PGAA and NDP, and may ultimately lead to a neutron milliprobe or even microprobe. The neutron microguide appears to be the most promising approach to achieve analytically useful focused neutrons, and as a first approach, a device will be developed and tested using a stack of nickel-coated single-crystal silicon wafers as focusing elements.

The strong interaction with industrial scientists using NDP, PGAA, and NAA has continued this year with a growing number of guest workers, research associates, and joint publications. An important development has been the demonstration of two-parameter coincidence spectroscopy (in NDP) for thin samples, in collaboration with the NDP group at the University of North Carolina and with Eastman Kodak. For appropriate samples, this technique provides an increase in sensitivity equivalent to more than a tenfold increase in reactor neutron flux. The measurement of distributions of lithium and boron in metals, glasses, and polymers continues to produce important results.

The Nuclear Methods Group is collaborating with scientists from the NIST Center for Atomic Molecular and Optical Physics, the NIST Center for Radiation Research, and the University of Sussex in a new measurement of the neutron lifetime. The goal of this research is to provide a new value for the neutron lifetime with an uncertainty of less than 0.3%. The current uncertainty for the neutron lifetime is about 0.6%. The technique used involves detecting the protons which decay in a neutron beam passing through a superconducting-magnet proton trap. Such measurements have been made with a cold neutron beam at the Institut Laue-Langevin in Grenoble, and additional measurements will be made at the new NIST CNRF next year.

During the coming year the group will continue to improve the accuracy, productivity, and sensitivity of nuclear methods as applied to elemental measurements. Problems to be addressed include those inherent in sample preparation, irradiation, radiochemical separation, counting, and data reduction, with the goal of minimizing and quantifying various sources of random and systematic errors in analysis by nuclear methods. Maintaining full accuracy at high count rates using current-generation data-acquisition electronics is the goal of our count-rate-dependent studies. Accurate quantification of gamma-ray self-absorption and measurement of the shape of
the efficiency curve are required for accuracy in monitor activation analysis (since matching of sample with primary standards is not done). The development of monitor activation analysis is part of the high temperature superconductivity effort to achieve rapid sample turnaround with minimum sacrifice of accuracy; however, the approach will also provide good quality control when used in parallel with traditional primary standard NAA.

2. Neutron Interactions and Dosimetry Group

J. Grundl

The Neutron Interactions and Dosimetry Group develops and applies well-characterized neutron fields and related capabilities as permanent irradiation facilities for neutron dosimetry methods evaluation and standardization, for neutron detector development and calibration, and for reaction cross section measurements. Related calculational efforts focus on neutron transport and fundamentals of radiation exposure at the microscopic level. Strong involvement with outside organizations in the federal and private sectors include many types of research and technology assistance projects as well as leadership roles on national and international standards and radiation policy making bodies.

A selection of accomplishments for FY-89 with emphasis on NIST reactor related activities are outlined below in titled paragraphs grouped under three of the five projects that make up the group. More complete summaries may be found in the Reactor Division Annual Report.

Dosimetry for Material Performance Assessment

Dosimetry methods for monitoring the degradation of materials in high fluence neutron exposures are diverse. This project provides some form of measurement assurance, standardization, or methods development for nearly every approach to materials dosimetry employed in the United States. Interlaboratory measurement cooperation including substantial international participation are an important feature of this project.

1. **NIST/Nuclear Regulatory Commission Contract.** The 1977 Rancho Seco Reactor incident demonstrated that it was possible for a nuclear pressure vessel subjected to thermal shock to also experience significant pressure loading. Later studies showed that relatively small flaws subject to pressurized thermal shock (PTS) could lead to failure of the pressure vessel. The need for improved surveillance of pressure vessel embrittlement damage was clear. NIST has participated in a long program of benchmark dosimetry measurements and calculations, and continues as consultant to the Nuclear Regulatory Commission in this area. In particular this year, NIST has been a major participant in the formulation of regulatory guide for evaluating pressure vessel neutron fluences.

2. **NIST/Westinghouse Cooperative Agreement.** Measurement assurance activities with respect to Solid State Track Recorders (SSTRs) developed by Westinghouse Science & Technology Center (formerly Westinghouse Research) continues. The trend in the nuclear power industry to implement SSTR dosimetry in ex-vessel cavities is growing. NIST is
specifically interested in problems associated with establishing reliable masses for the SSTR fissionable deposits which are in the pico-
gram to nano-gram range.

3. **Calibration of BELGIAN Cavity Fission Source.** NIST personnel traveled to Mol, Belgium in December 1988 to re-establish the tie between the NIST and CEN/SCK fission neutron fluences in their respective 235U Cavity Fission Sources. The basic calibration measurements were made with an NIST dual fission chamber containing fissionable deposits identical to those used in the calibration accomplished at Mol by NIST (then NBS) in 1983. Measured fission rates agreed to about ±1 % (1σ). The most active use of the Belgian Cavity Fission Source has been to calibrate miniature fission chambers for active dosimetry in the Belgian LWR-PV Benchmark Experiment, VENUS, at the CEN/SCK Laboratory in Mol, Belgium. The VENUS series of PV benchmark experiments confirm neutron transport calculations of dosimetry measurements in the vicinity of fuel corners and out through the pressure vessel into the ex-vessel cavity.

4. **Dosimetry Methods Development for Reactor Support Structures.** An NRC priority effort in the Heavy Section Steel Technology (HSST) Program this year was evaluation of low-temperature, low-fluence-rate embrittlement on reactor vessel support structures. This issue arose from findings of higher than expected embrittlement in the High Flux Isotope Reactor (HIFR) at Oak Ridge, and in the reactor shield tank from the recently decommissioned Shippingport Reactor. The HSST staff selected two specific plants for detailed review. NIST will provide consultation and measurement assurance for planned dosimetry measurements in these plants.

5. **Energy Response of Innovative Electronic-hardware Dosimeter.** Nuclear Effects Directorate (NED) at the Aberdeen Proving Ground has been evaluating a new personnel dosimetry system for battlefield use by the Army. Special NIST neutron-field facilities were used to establish the neutron energy sensitivity of this semiconductor device. Multiple irradiations were performed for NED in a thermal neutron beam, in the 2, 24 and 144-keV filtered beams at the reactor and at the 252Cf Irradiation Facility.

**PUD Neutron Sensors for Out-of-Core Reactor Dosimetry.**

Paired Uranium Detectors (PUDs) is a new fission activation detector technique developed at NIST. Its main feature is to circumvent the conventional need for highly depleted 238U material (now becoming scarce) and to obtain an integrated measurement of the 235U/238U fission ratio, an important neutron spectrum index. PUDs were first used in low-level leakage core measurements at the H.B. Robinson commercial power plant, and more recently have become used as ex-vessel cavity fluence dosimeters in commercial power plants. All of this is carried out as part of the NIST/Westinghouse cooperative agreement.
Personnel Dosimetry

Standard neutron fields are used to calibrate radiation protection instrumentation and to investigate and test new types of dose-measuring techniques. Responsibilities in national and international dosimetry methods research focuses on tissue dose modeling, and tissue equivalent proportional counter (TEPC) measurements, and participation in the development of written standards by the ICRU and ISO.

1. Calibration Service. Approximately 60 neutron radiation protection instruments were calibrated this year. Although the majority of these calibrations are done for commercial nuclear power plants, also included are institutions as diverse as Redstone Arsenal and the M. D. Anderson Cancer Center.

2. Performance tests of "Bubble Dosimeter". The new "bubble", or superheated drop, detector is a new and promising approach to neutron dosimetry. We are involved in a joint project to determine the relevant properties of these detectors. While much data remains to be analyzed, it appears that the "bubble dosimeter" (supplied by Bubble Technology, Inc. (BTI)) has high sensitivity, a good dose equivalent response, and is quite linear up to the point where the bubbles can no longer be accurately counted.

3. TEPC Dose Measurement for Neutron RBE. The Armed Forces Radiobiology Research Institute (AFRRI) recently obtained surprising results in experiments which examined the relative biological efficiency (RBE) for lethality in mice exposed to reactor neutrons. In brief, the experiments indicated an anomalously high RBE for neutrons. In an effort to shed some light on this result, an independent measurement of the neutron and gamma-ray fields was carried out using a tissue-equivalent proportional counter (TEPC). The measurement disagreed with conventional ion chamber dosimetry in a manner that removed the anomaly. Efforts will be made to understand the disagreement between the two dosimetry techniques.

4. ICRU Publication on Practical Determination of Dose Equivalent. In a landmark publication, the International Commission on Radiation Units and Measurements (ICRU) recommended a new system for determining the dose equivalents resulting from exposure to external radiation sources. The advantages of the new system include a uniform approach to dose equivalent determination and reporting for gamma-rays, electrons, and neutrons, and a closer link with the fundamental limiting quantity: effective dose equivalent.

5. Radiation Protection Policy Committee Work. C. Eisenhauer serves as chairman of a subpanel of the Science Panel of the Committee on Interagency Radiation Research and Policy Coordination (CIRRPC). The sub-panel has produced a report on the desirability of planning for research on human health effects in the event of a nuclear accident. The Subpanel recommended a follow-up effort which would recommend ways of implementing such planning.
Research and Technology Assistance

Research and technology assistance are strongly coupled in neutron dosimetry. A multiplicity of institutional involvements, drawn to the group by the availability of unique irradiation facilities and measurement capabilities, encourages a variety of attractive projects and unavoidable responsibilities.

1. Neutron Lifetime - Absolute Neutron Counting. An overview of the NIST involvement in measurement of the free neutron lifetime is given in the Fundamental Physics section of the NON-RRD program reports. One aspect of the experiment will be mentioned here: improvements in the absolute neutron counting under development for the upcoming new phase of the project at the NIST guide hall. In the next phase of the project at NIST, calibration of the neutron counter will be made by two new methods which do not depend on either the B-10 sample mass nor the B-10 reaction cross section. The new methods are Alpha-Gamma Coincidence and Calorimetry. Both of the calibration methods are believed to have the potential for approaching the 0.1 % accuracy level.

2. Benchmark Measurements for Criticality Safety. In collaboration with Department of Energy an experimental program has been undertaken to improve the understanding of neutron leakage from aqueous systems. The problem addressed is in the field of criticality safety analysis where sub-critical aqueous assemblies are misrepresented by existing calculational techniques. The present efforts will measure fission reaction rates in the neutron leakage from water spheres driven by Cf-252 fission neutron sources. Fission chambers with fissionable deposits from the NIST collection of Fissionable Isotope Mass Standards (FIMS) are employed in the measurements. Calculation and experiment are in good agreement for U-238 and Np-237 reaction rates; for U-235 and Pu-239, experiment is 5% to 8% lower than calculation.

3. Radiation Shielding Calculations for NIST Cold Neutron Facility. Calculations have been made to determine shielding necessary for a shutter to be installed at the Cold Neutron Facility. Various combinations of iron, tungsten, borated polyethylene, and lead were explored to determine an optimum configuration. The shutter will be placed at the wall of the reactor room. Calculations were made for I. Schroeder using both the discrete ordinates and Monte Carlo methods. Special areas of concern are the regions near the shutter when the shutter is closed and the other side of the wall, which is the interior face of the Guide Hall. Monte Carlo calculations were used when the geometry was critical and discrete ordinate calculations for thick-shield calculations.

3. Fundamental Physics

Dr. Geoffrey Greene  
(Center for Atomic, Molecular and Optical Physics)

The Fundamental Neutron Physics Program at the NIST Cold Neutron Research Facility includes research in particle physics, basic nuclear physics, and
neutron interferometry. It is a collaborative effort involving personnel from the Reactor Radiation Division, as well as from other organizational units within the National Measurement Laboratory, including the Quantum Metrology Division, Center for Atomic, Molecular, and Optical Physics; and the Neutron Interactions and Dosimetry Group, Center for Radiation Research. This work has a strong collaborative character with contributions from other institutions including the University of Missouri, Los Alamos National Laboratory, the University of Sussex (U.K.), and Harvard University. In addition, this work receives support from the Department of Energy and NATO.

This year considerable effort has gone into the design and initial construction of the Neutron Interferometry Position at the Cold Neutron Facility. This work has included detailed analysis of the vibration isolation system as well as the completion of the design for the first stage of seismic isolation.

In the area of nuclear and particle physics, significant work has gone into an experimental effort to determine the half-life of the neutron. Very successful preliminary runs were carried out at the Institut Laue Langevin in Grenoble, France. This experiment will be installed on guide #6 at the NIST facility as the initial instrument on the fundamental physics end position.
OUTPUTS/INTERACTIONS
PUBLICATIONS: Reactor Radiation Division


Giebultowicz, T. M.; Rhyne, J. J.; Seehra, M. S. and Kannan, R. "Neutron Diffraction in Co\textsuperscript{p}Mg\textsubscript{1-p}O Solid Solutions," Journal de Physique Colloque C8, 49, 1105 (1988).

Giebultowicz, T. M.; Rhyne, J. J.; Furdyna, J. K. and Galazka, R. R. "Neutron Diffraction Studies of Cd\textsubscript{1-x}Mn\textsubscript{x}Te and Zn\textsubscript{1-x}Mn\textsubscript{x}Te Single Crystals," Journal de Physique Colloque C8, 49, 1199 (1988).

Giebultowicz, T M.; Rhyne, J. J.; Ching, W. Y.; Huber, D. L; Furdyna, J. K.; Lebech, B. and Galazka, R. R. "Harmonic Magnons in Zn\textsubscript{1-x}Mn\textsubscript{x}Te and Cd\textsubscript{1-x}Mn\textsubscript{x}Te," Phys. Rev. B 39, 6857 (1989).


Gotaas, J. A.; Said, M. R.; Kouvel, J. S.; Brun, T. O. "Magnetic Structure of Cubic Tb\textsubscript{0.3}Y\textsubscript{0.7}Ag", J. de Physique (Paris) 49, C8-345-346 (1988).


Mangin, P.; Boumazouza, D.; George, B.; Rhyne, J. J. and Erwin, R. W. "Static and Dynamic Properties of the (Fe_{1-x}Cr_x)_{75}Si_{15}Mo_{10} Reentrant Spin Glass Amorphous Alloy," Physical Review B (to be published).


Spano, M. L., Gotaas, J. A. and Rhyne, J. J. "Neutron Scattering Study of Magnetic Order in a Tb$_{0.5}$Dy$_{0.5}$ Single Crystal," J. de Physique J. de Physique, Colloque C8, Suppl. #12, 345 (1988).


Torardi, C. C.; Reiff, W. M. and Prince, E. "Topochemical Li-ion Insertion into FeClMoO$_4$ and Fe$_2$(MoO$_4$)$_3$: Structure and Magnetism of LiFeClMoO$_4$ and Li$_2$Fe$_2$(MoO$_4$)$_3$". Matls. Sci. Forum 27/28, 223-228 (1988).


Technical and Professional Committee Participation and Leadership

American Crystallographic Association
A.I.P. Crystal Data Committee, A.D. Mighell, ACA Representative

American National Standards Institute
N-17: Research Reactors, Reactor Physics, and Shielding
T. M. Raby, Chairman, J. Torrence, alternate member.

American Nuclear Society
Standards Steering Committee, T.M. Raby, Member
Standards Subcommittee, ANS-15, Operations of Research Reactors,
T.M. Raby, Member
Working Groups ANS-15.1, ANS-15.11,
T.M. Raby, Chairman, J.F. Torrence, Member

Basic Energy Sciences Advisory Committee (DOE) (appointed by the Secretary of Energy), User Facilities Subcommittee
J.M. Rowe, Member and Subcommittee Chairman.

BNL Neutron Scattering Facility Scheduling Committee,
J.J. Rush, Member

Committee Refereeing Neutron Scattering Proposals for the High Flux Isotope Reactor at Oak Ridge National Laboratory.
C.F. Majkrzak, Member

French National Center for Scientific Research (C.N.R.S.), Louis Neél Laboratory, Scientific Advisory Panel
J.J. Rhyne, Member

HFBR (Brookhaven) Neutron Beamlines and Facilities Upgrade, Review Committee
J.J. Rhyne, Member

Intense Pulsed Neutron Source (Argonne) and Los Alamos Neutron Scattering Center, Program Review Committees
J.J. Rhyne, Chairman

Intense Pulsed Neutron Source Scientific Advisory Committee (ANL)
J.M. Rowe, Member

International Centre for Diffraction Data
Data Base Subcommittee, J.K. Stalick, Member

International Symposium on Magnetoelasticity and Electronic Structure of Transition Metals, Alloys, and Films, International Advisory Board
J.J. Rhyne, Member

International Union of Crystallography
-Commission on Crystallographic Nomenclature, Subcommittee on Statistical Descriptions in Crystallography, E. Prince, Member
-Commission on Journals, E. Prince, Member
JCPDS—International Centre for Diffraction Data
-Crystal Data Management Board, L.D. Mighell, Member
-Crystal Data Task Group, A.D. Mighell, Member
-Database Subcommittee, A.D. Mighell, Member
-Phase Identification by Electron Diffraction Subcommittee,
  A.D. Mighell, Member

Journal of Applied Crystallography
E. Prince, Co-Editor

Journal of Applied Physics, Editorial Board
J.J. Rhyne, Member

Los Alamos Neutron Scattering Center
Scientific Advisory Committee, J.M. Rowe, Chairman

National Organization for Test, Research, and Training Reactors
T.M. Raby, Member of Executive Committee and Past Chairman
J.F. Torrence, Member and past Co-Chairman.

National Steering Committee for the Advanced Neutron Source
J.J. Rush, Chairman

Program Advisory Committee for the High Flux Beam Reactor at Brookhaven
National Laboratory
C.F. Majkrzak, Member

Working Group on SANS/Reflectometry, ORNL/BNL Workshop on Neutron Scattering
Instrumentation at High Flux Reactors, June 5-7, 1989, Oak Ridge, TN.
C.F. Majkrzak, Member
Industrial and Academic Interactions

As a national center for the development and application of neutron methods in condensed matter and materials science, chemical analysis and radiation standards, the Reactor Radiation Division and other NIST organizations have direct interactions and cooperative programs with 45 universities, 22 industrial and 23 foreign laboratories. A few examples of the many interactions of the Neutron Scattering Group are:

o An agreement is in place between NIST and Exxon Research and Development Corporation to jointly develop and operate a world-class small angle neutron scattering spectrometer at the NIST cold neutron source. This facility is in the final days of construction. Cooperative research efforts on the existing SANS instrument at the reactor include work on wetting in microporous media and micellar systems.

o Work has been initiated on a new NIST/NSF Center for High Resolution Neutron Scattering at the CNRF which will offer U.S. Scientists access to world-class capabilities in high resolution small angle neutron scattering studies of materials microstructure and in very high resolution spectroscopy of condensed materials using polarized neutrons beams.

o An intense cooperative research effort was organized early in 1987 with several groups at AT&T Bell Laboratories on the systematic studies of the structure and properties of the new class of high-temperature superconductors. This joint research continues to grow and produce important new results.

o A joint research effort is underway with IBM for the development and application of neutron reflectometry for unique studies of surface and interfacial structure of polymer films, liquids and other materials. This research has thus far led to the best results worldwide in neutron studies of polymer surfaces.

o The Reactor Radiation Division’s Crystal Data Center is engaged in a number of interactive links including joint development and distribution to U.S. science and industry of evaluated crystal data with the International Center for Diffraction Data. The Data Center also has long-term agreements in place with crystal data programs in Canada, Great Britain, and Germany to jointly develop and share critical data on the structure of materials. An expansion of database systems into electron diffraction and microscopy has recently been achieved, which several instrument companies are preparing to integrate into their products.

o The Neutron Scattering Group has in place wide collaborative research with the University of California (Santa Barbara) involving neutron inelastic scattering, neutron diffraction, and SANS studies of catalysts, non-linear optical materials, and radiation damage.
Extensive cooperative research efforts with the Physics Department and Materials Research Laboratory at the University of Illinois and with Michigan State have continued during the past year including neutron diffraction studies of a new class of layered magnetic materials and research on new kinds of metal-molecular complexes created within the layers of pillared clays and oriented graphite. Joint NIST/Illinois efforts have recently been extended to the first use of near-grazing angle neutron diffraction in surface structure studies and to SANS studies of sol-gel processing of high \( T_c \) superconductors.

Cooperative Research Program with the Department of Physics and Astronomy of the University of Maryland. Under this program RRD staff are engaged with Maryland scientists in joint research on magnetic materials, superconductors and catalysts. Some of this research is carried out jointly with scientists from industrial labs.

An extensive collaborative research effort is in progress with Bell Communications Research this past year focussing on magnetic substitutions and interactions in high \( T_c \) superconductors. Associated research is also underway with Johns Hopkins on the effect of atomic substitution on superconductivity.

**Associated Activities**

During the past year scientists in the Reactor Radiation Division delivered over thirty-five invited lectures in the U.S. and abroad. The Neutron Scattering Group also hosted a workshop on "Applications of Neutron Methods for High Temperature Superconductors", and Group members were invited participants in several other workshops in the neutron field. Neutron Group Scientists continue to share several NATO grants with French and German colleagues in different areas of condensed matter physics. Joint research continues with ORNL and BNL in the development of supermirrors and monochromators.
**BIBLIOGRAPHIC DATA SHEET (See instructions)**

<table>
<thead>
<tr>
<th>1. PUBLICATION OR REPORT NO.</th>
<th>NISTIR 89-4152</th>
</tr>
</thead>
<tbody>
<tr>
<td>3. Publication Date</td>
<td>October 1989</td>
</tr>
<tr>
<td>4. TITLE AND SUBTITLE</td>
<td>Institute for Materials Science and Engineering, Reactor Radiation Division's Technical Activities 1989</td>
</tr>
<tr>
<td>5. AUTHOR(S)</td>
<td>J.M. Rowe, (460) x6210</td>
</tr>
<tr>
<td>6. PERFORMING ORGANIZATION (If joint or other than NBS, see instructions)</td>
<td>NATIONAL BUREAU OF STANDARDS U.S. DEPARTMENT OF COMMERCE GAITHERSBURG, MD 20899</td>
</tr>
<tr>
<td>7. Contract/Grant No.</td>
<td></td>
</tr>
<tr>
<td>8. Type of Report &amp; Period Covered</td>
<td>NISTIR</td>
</tr>
<tr>
<td>9. SPONSORING ORGANIZATION NAME AND COMPLETE ADDRESS (Street, City, State, ZIP)</td>
<td></td>
</tr>
<tr>
<td>10. SUPPLEMENTARY NOTES</td>
<td>Document describes a computer program; SF-185, FIPS Software Summary, is attached.</td>
</tr>
<tr>
<td>11. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)</td>
<td>This report summarizes all those programs which depend on the NIST reactor. It covers the period from October 1, 1988 through September 30, 1989. The programs range from the use of neutron beams to study the structure and dynamics of materials through nuclear physics and neutron standards to sample irradiation for activation analysis, isotope production, radiation effects studies, neutron radiography and nondestructive evaluation.</td>
</tr>
<tr>
<td>12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)</td>
<td>activation analysis; crystal structure; diffraction; isotopes; molecular dynamics; neutron; neutron radiography; nondestructive evaluation; nuclear reactor</td>
</tr>
<tr>
<td>13. AVAILABILITY</td>
<td>□ Unlimited</td>
</tr>
<tr>
<td></td>
<td>✔ For Official Distribution. Do Not Release to NTIS</td>
</tr>
<tr>
<td></td>
<td>□ Order From National Technical Information Service (NTIS), Springfield, VA. 22161</td>
</tr>
<tr>
<td>14. NO. OF PRINTED PAGES</td>
<td></td>
</tr>
<tr>
<td>15. Price</td>
<td></td>
</tr>
</tbody>
</table>