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NBS TECHNICAL NOTE 567

Reactor Radiation Division:

Annual Progress Report for the Period Ending October 31, 1970

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Annual Progress Report for the Period Ending October 31, 1970

Robert S. Carter, Editor

Reactor Radiation Division
Institute for Materials Research
National Bureau of Standards
Washington, D.C. 20234

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FOREWORD

The reactor has completed its first year of full power operation so it is now appropriate to review the progress made during the last year. This report is the first annual progress report of the Reactor Radiation Division.

The reactor was built not only to serve the needs of the National Bureau of Standards but also those of the greater Washington Scientific Community and other government agencies. It is the responsibility of the Reactor Radiation Division to foster the wise scientific and technological use of the reactor as well as assure its safe and efficient operation. Toward this end, the Division has a small nucleus of scientists experienced in the use of reactors for a wide range of scientific and technical problems. In addition to pursuing their own research and developing sophisticated experimental facilities, they actively seek out and encourage collaboration with other scientists engaged in challenging programs, whose work can benefit from use of the reactor, but who as yet do not have the reactor experience necessary to take full advantage of the facilities available. The Division also provides irradiation services to a wide variety of users as well as engineering and other technical services.

The reactor operates at 10 Mw and is designed to provide a broad spectrum of facilities ranging from intense neutron beams to extensive irradiation facilities, making it one of the most versatile high flux Research Reactors in the country. Thus it is able to serve a large number of scientists and engineers in a broad range of activities both within and outside the NBS.

Use of the reactor by individuals and organizations outside the Reactor Radiation Division has increased significantly during the past year and continues to expand rapidly. Twenty-five scientists from other parts of the Bureau, representing 16 man-years of effort, use the reactor regularly either directly or in cooperation with scientists in the Division. An additional 32 scientists from other agencies, universities, and industry spent 12 man-years of effort using the reactor during the past year.

The purpose of this report is to summarize the activities of the large number of users outside the Division as well as the work in the Division. The summaries are divided into two groups. One is the research activities and the other is the operation and service activities. The research activities are further broken down into three groups: the first includes all the work of the Division and most of the collaborative work; the second describes the activities of our formal interagency collaboration based on long-term arrangements where scientists from other agencies are actually stationed at the reactor; and the third catagory is the summary of activities of other NBS divisions which use the reactor directly rather than through collaboration with Reactor Radiation scientists.

R. S. Carter, Chief
Reactor Radiation Division

REACTOR RADIATION DIVISION

Annual Progress Report

for period ending October 31, 1970

Edited by R. S. Carter

The reactor has completed its first year of full power operation so it is now appropriate to review the progress made during the last year. This report is the first annual progress report of the Reactor Radiation Division.

Key words: Crystal structure; diffraction; molecular structure; neutron; nuclear reactor; radiation; scattering.

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RESEARCH PROGRAMS

A. Division Programs

Time-Dependent Klein-Gordon Equation

V. W. Myers

The time-dependent Klein-Gordon equation is solved for the motion of a charged particle interacting with a traveling plane electromagnetic wave. The Klein-Gordon equation is the relativistic quantum mechanical equation for a spin zero particle and is appropriate for a π meson, for example. The electromagnetic field is treated classically, and the solutions of the Klein-Gordon equation correspond to the strong field limit. The solutions can be expressed in the form

$$f(z-ct)e^{i(\vec{k}\cdot\vec{r}-Wt)}$$

where z is the propagation direction of the wave, and k and W are constants. Expectation values for such quantities as the particle's kinetic energy and mechanical momentum are computed. A manuscript describing the details of this research is being prepared.

Thermal Neutron Flux Measurements

V. W. Myers and M. Ganoczy

Research has continued on a new technique for measuring thermal neutron flux which is the number of neutrons per second passing through a sphere of 1 cm cross section from any direction. Small glass beads ($\sim$ 1 millimeter radius) are made black to thermal neutrons by the inclusion of ^{10}B since this isotope has a very large capture cross section for thermal neutrons. An activator such as cobalt is incorporated in the glass. If the bead is placed in a neutron field for a known time interval, the resulting radioactivity can then be related directly to the neutron flux. In contrast, a thin foil with a 1/v capture cross section determines only the neutron density (the number of neutrons per unit volume).

The activity of a cobalt bead after irradiation for a time t small compared to the mean life τ (7.6 years) in a thermal neutron flux is

$$A = (n\ \bar{v})(\pi\ r^2)(\frac{\Sigma_{co}}{\Sigma})(\frac{t}{\tau})(1-f).$$

A is the disintegration rate, $n\ \bar{v}$ is the thermal neutron flux, and r is the bead radius. Σ_{co} is the macroscopic activation cross sections of cobalt (nuclear cross section times atomic density), and Σ is the total macroscopic capture cross section of the glass. The correction for neutron transmission and scattering is f and is $\sim$ 0.02. It is advantageous to have identical beads since a bead with a known disintegration rate could then be sent to other laboratories for calibration of counting equipment, and such complications as the dependence of Co^{60} gamma ray absorption and counting efficiency on bead size are eliminated.

Improvements were made in fabrication of the beads. The finishing technique involves a random grinding process for a single bead, and spherical beads can be produced with essentially identical radii. The initial grinding of the beads is achieved by driving them with an air jet inside a cup grinding wheel.

It is necessary to know the composition by weight of the major elements comprising the glass in order that Σ_{co}/Σ can be determined. Collaboration with the Activation Analysis Section is countinuing on attempting to obtain more precise measurements of the cobalt content so that the uncertainly in Σ_{co}/Σ can be reduced to 1% or less.

An alternative approach is to activate a bead in a known neutron flux. For this purpose, a bath consisting of manganese sulfate dissolved in water is being assembled at one of the beamports of the NBSR. A neutron beam of the order of a millimeter radius will be directed into the bath. The cylindrical bath is of sufficient size (length and diameter are each 0.45 meter) that less than 1 percent of the thermal neutrons are not captured in the bath. Continuous counting of the ^{56}Mn activity produced from neutron capture in manganese will be done using a NaI detector. The system is calibrated by the addition of a known ^{56}Mn activity into the bath, and the neutron beam flux can then be determined. A bead can then be calibrated by activation in this known neutron flux for a prescribed time.

The overall uncertainty in the measurement of thermal neutron flux by the bead technique is anticipated to be of the order of 2%.

Crystallographic Studies of Metal Organic Materials

A. Santoro

and

C. W. Reimann and A. D. Mighell
(Inorganic Materials Division)

In the past year a number of transition metal complexes have been studied by means of diffraction techniques. The metal ions used in these studies include Cu^{++}, Cd^{++}, Co^{++} and Co^{+++} and the ligands include imidazole, dipyridine, and pyridine-N-oxide.

The structure analysis of these complexes provides direct information on the stereochemistry and the utilization of the active sites of the ligand molecules and on the symmetry of the coordination polyhedra. Moreover, this type of analysis gives precise bond distances and angles and furnishes evidence on the role played by hydrogen bonding in the packing of the structures.

The results of these studies may be used as the structural basis for a meaningful interpretation of spectroscopic data and may clarify the role played by some ligand molecules, such as imidazole, in biological systems.

Most of this work has been carried out by x-ray diffraction techniques. More recently, however, x-ray and neutron diffraction studies have been combined in order to locate hydrogen atoms in hydrogen bonded systems. The use of neutrons will be extended also to those cases in which radiation damage prevents the collection of accurate x-ray intensities.

The following crystal structures have been analyzed in the past year:

1) Crystal structure of Hexakis(imidazole)cadmium(II) Nitrate, $[Cd(C_3H_4N_2)_6](NO_3)_2$,

and Hexakis(imidazole) cadmium(II) Hydroxide Nitrate Tetrahydrate, $[Cd(C_3H_4N_2)_6](OH)(NO_3)\cdot 4H_2O$.

Both these structures consist of discrete $Cd(C_3H_4N_2)_6^{++}$ complex cations and NO_3^- anions, but as result of the presence of water molecules and OH^- ions in the hydroxide complex, the packing of the two structures is considerably different. The structure determination of both compounds was carried out mainly to compare the same complex cation in two different environments. The bond distances and angles between the atoms of the imidazole molecules are practically identical in the two structures but, in order to accomodate hydrogen bonding, there is considerable difference in the orientation of the rings.

2) Crystal structure of Nitratobis(dipyridine)cobalt(III) Hydroxide Nitrate Tetrahydrate, $[Co(C_{10}H_8N_2)_2(NO_3)](OH)(NO_3)\cdot 4H_2O$.

In this structure the Co atom is octahedrally coordinated by two molecules of dipyridine and by a intrate group acting as a symmetric bidentate ligand. This is one of the few nitrate complexes in which the role of the nitrate group has been clarified crystallographically. The water molecules, the nitrate groups and the OH^- ions are involved in a continuous network of hydrogen bonds.

3) Crystal Structure of Hexakis(imidazole)Cobalt(II) Nitrate, $[Co(C_3H_4N_2)_6](NO_3)_2$.

This compound has been analyzed in collaboration with Dr. E. Prince by x-ray and neutron diffraction techniques in order to locate the hydrogen atoms and define without ambiguity the hydrogen bonding in the structure. Another purpose of this work is to compare x-ray and neutron structural data.

4) Crystal structure of the 2:1 Dimer$[(\text{Pyridine-N-Oxide})_2CuBr_2]_2$.

A large number of aromatic N-oxides of Cu^{++} have been classified in terms of 12 structure types in an effort to generalize their magnetic behavior. $[(\text{Pyridine-N-Oxide})_2CuBr_2]_2$ was classified as probably belonging to the same structural type of the chloride analog in spite of its anomalous magnetic moment. The completed structure determination of the complex has shown that, although the basic molecular unit is a dimer as in the chloride analog, it differs markedly from the chloride complex.

The Structure of Potassium Silicotungstate

E. Prince

and

P. M. Smith and J. V. Silverton
(Dept. of Chemistry, Georgetown University, Washington, D.C.)

Potassium silictungstate is one of a series of compounds containing anions that are designated "heteropoly" because they are large, complex groups containing more than one kind of cation. The silicotungstate ion has the composition $SiW_{12}O_{40}$. The potassium salt crystallizes in 2 different forms, a hexagonal α form containing from 16 to 18 waters of hydration per formula unit, and a monoclinic β form containing 12 to 14 waters of hydration. A neutron diffraction investigation was undertaken with two aims, first to determine accurately the configuration of the silicotungstate ion and, second, to attempt to elucidate the role of water of hydration in the crystal structure. Complete, 3-dimensional data have been collected from a crystal of the α form crystallized from normal water, and hk0 data have been collected from a crystal grown from heavy water. Analysis of the data, which is still in progress, has yielded accurate parameters for the anion, but the search for water has been less successful. A difference Fourier synthesis has revealed a large hole, with a radius of nearly 6 Å extending all the way through the crystal along a 6_2 axis, in which there is apparently no ordered scattering material. One hypothesis is that this void is occupied by water which is virtually free, thus accounting for the instability of these crystals in an unsaturated atmosphere. It is hoped that in the future it will be possible to study the β form, which is somewhat more stable.

The Structure of Dimethyl Sulfone Diimine

E. Prince
and
J. Bevan
(Procter & Gamble Co., Cincinatti, Ohio)

Dimenthyl sulfone diimine, $(CH_3)_2S(NH)_2$ is a simple compound which is chemically related to the basic constituents of synthetic detergents. A neutron diffraction study was undertaken to determine the positions of the hydrogen atoms. The imino hydrogens form a system of hydrogen bonds. 261 independent diffraction intensities were measured. Analysis of the data is still in progress.

The Structure of Apophyllite

E. Prince
and
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(Dept. of Geology, California State College, Los Angeles, Calif.)
and
G. Donnay
(Geophysical Laboratory, Carnegie Institute of Washington, Washington, D.C.)

Apophyllite is a hydrated, fluorosilicate mineral with the ideal chemical formula $KCa_4(Si_4O_{10})_2F\cdot 8H_2O$. It has an unusual sheet structure, with 4-membered rings of SiO_4 tetrahedra linked together to form an infinite, two-dimensional network. Fig. 1 is a stereoscopic pair showing this structure clearly. If apophyllite is heated, gas is evolved in two distinct stages. A neutron diffraction investigation was undertaken in order to determine the structural role of the water of hydration, in hopes that the reason for the two-stage evolution of gas would become apparent.

A least-squares refinement of three-dimensional data (671 reflections) with anisotropic temperature factors led to an R index ($R=\Sigma|F_o-F_c|/\Sigma F_o$) of .046. The structure parameters were in excellent agreement with previous x-ray determinations, and the configuration and environment of the water molecule (Fig. 2) were very reasonable. A difference Fourier

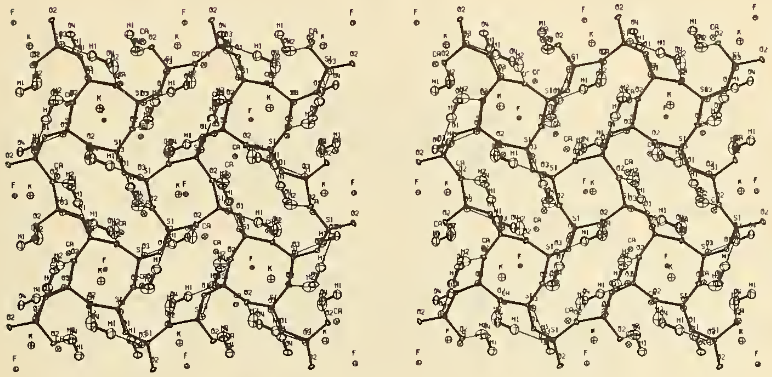

Fig. 1. Stereoscopic pair showing the infinite silicate sheets in apophyllite.

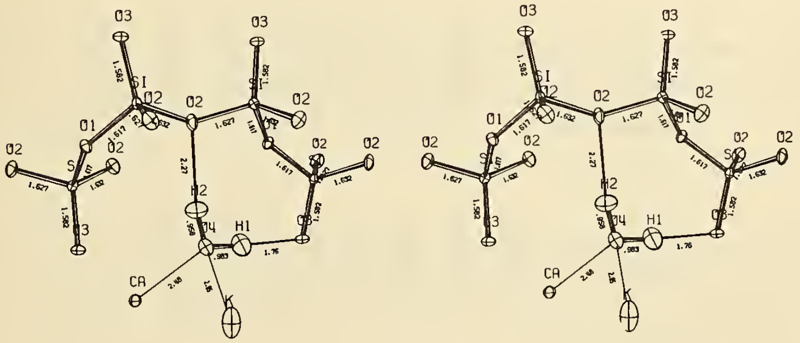

Fig. 2. The environment of the water molecule in apophyllite.

Table 1

Atom	*x*	*y*	*z*	B^*_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
F	0	0	0	0.56(2)	0.56(2)	1.61(2)	0	0	0
K	0	0	1/2	1.4(2)	1.4(2)	3.8(4)	0	0	0
Ca	.1110(3)	.2473(3)	0	0.83(9)	0.79(9)	0.79(8)	-.08(6)	0	0
Si	.2267(2)	.0866(2)	.1896(1)	0.42(6)	0.57(6)	0.66(6)	-.02(6)	-.02(5)	.06(6)
O(1)	.3636(3)	.1364(3)	1/4	0.64(5)	0.64(5)	0.85(6)	-.02(5)	-.20(4)	-.20(4)
O(2)	.0846(2)	.1897(2)	.2169(1)	0.59(5)	1.07(5)	1.38(5)	.34(4)	-.11(7)	-.26(4)
O(3)	.2657(2)	.1018(2)	.0924(1)	1.09(5)	1.05(5)	0.43(5)	-.01(4)	-.03(4)	-.02(4)
O(4)	.2154(2)	.4500(2)	.0893(1)	2.45(7)	1.13(7)	1.47(8)	-.06(5)	.11(6)	-.29(5)
H(1)	.4490(4)	.1730(4)	.0889(2)	2.0(1)	2.6(1)	2.6(1)	-.1(1)	.36(9)	-.06(9)
H(2)	.2244(5)	.4255(4)	.1481(2)	5.0(2)	2.6(1)	1.3(1)	0.0(1)	-.5(1)	.1(1)
H(3)	0	0	.065(1)	7.1(8)	7.1(8)	2.8(8)	0	0	0

*Units are Å^2.

synthesis revealed an additional hydrogen atom near the fluorine atom, forming a molecule of hydrogen fluoride. A further refinement, replacing the water molecules by hydroxyl ions and attaching the remaining hydrogen atom to the fluorine atom gave an R index of .037. Table 1 shows the final parameters. A paper describing these results is being submitted to The American Mineralogist.

The Structure of Durene

E. Prince, L. W. Schroeder, and J. J. Rush

As part of a study of the structure, dynamics and phase transitions in methylbenzene compounds, we have extended our experiments to a crystal structure study of durene. Durene (1,2,4,5-tetramethyl benzene) is a methylated, aromatic hydrocarbon. When methyl groups are attached to adjacent carbon atoms on a benzene ring, they are "over-crowded", and the structure of the molecule must have some distortion from an idealized structure containing 120° angles all around the ring and tetrahedral (109° 28') angles around the methyl groups. Durene, in contrast to hexamethylbenzene, was determined by previous neutron-inelastic-scattering experiments, to have a comparatively high barrier to rotation of the methyl groups (2 Kcal/mole vs 1 Kcal/mole). A neutron diffraction study of a single crystal of durene was undertaken in order to determine the configuration of the methyl groups. It is hoped that it will be possible to subject the diffraction data to a segmented-rigid-body analysis, and thereby to correlate the various amplitudes of molecular vibration with spectroscopic data. Analysis of the data is still in progress.

Proton Dynamics and Phase Transitions in Transition Metal Hydrides

L. A. deGraaf and J. J. Rush

During the past year work has continued in collaboration with scientists at Argonne National Laboratory on the study of the vibration and diffusion of hydrogen atoms in transition-metal hydrides by quasielastic and inelastic neutron scattering measurements. The dynamics of the hydrogen atoms and the changes in motional behavior that occur at phase transitions in polymorphic hydrides are related to the thermodynamic, electrical and other physical properties of these materials.

Earlier this year the analysis was completed of quasielastic scattering spectra measured previously for the α and β phases of vanadium hydride using an incident beam of cold neutrons (λ 4.07Å, FWHM=0.7Å). These spectra for $VH_{0.20}$, $VH_{0.40}$ and $VH_{0.57}$ covered a range of momentum transfers from 0.5 to 2.3Å^{-1} and a temperature range of 23° to 250°C. The analysis of the quasielastic peak widths vs. momentum transfer was used to determine the concentration dependence of the diffusion in the β-phase(b.c.c.). In addition an activation energy of 4570 ±415 J/mole was derived from the temperature dependence of the hydrogen diffusion in the $VH_{0.20}$ sample. The neutron spectra for the β-phase of $VH_{0.570}$ indicate an abrupt decrease in the diffusion rate at the $\alpha \rightarrow \beta$ transition.

One principal aim of these experiments was to determine, if possible, the diffusive jump mechanism (and indirectly the sites occupied in the lattice) for the hydrogen atoms. Theoretical calculations were performed of the dependence of the neutron quasielastic line broadening on momentum transfer (κ) for hydrogen diffusion between various tetrahedral and octahedral interstitial sites in the b.c.c. lattice (α phase). Unfortunately, however, it was determined that the range of κ covered in the cold neutron experiment was not great enough for a meaningful comparison of theory and experiment to be made. We have thus recently expanded the quasielastic scattering measurements on $VH_{0.20}$ and $VH_{0.57}$ from $\kappa=2$ to $\kappa=4$ using a crystal monochrometer, neutron chopper time-of-flight facility at the NBSR. For these measurements we used an incident neutron wavelength of 2.43Å and studied the scattered spectra over an angular range of 45° to 106° at temperatures from 23 to 215°C. These data are presently being analyzed so that detailed comparisons can be made with the theoretical predictions for various diffusion models.

Neutron studies of the vibration spectra in two crystal phases of $PdH_{0.6}$ and of hydrogen diffusion in the TaH_x system are also being planned. Special sample holders have been designed for these measurements and are presently being constructed.

Spectroscopic Investigations of Dynamics and Hydrogen Bonding in Sodium and Potassium Bifluoride

J. J. Rush and L. W. Schroeder

The structure, vibration spectra and hydrogen bonding of sodium and potassium bifluoride have been investigated in the past by a variety of spectroscopic, diffraction and thermodynamic techniques. The lattice and internal vibrations of these crystals and their related properties are of particular interest because of the very strong symmetric hydrogen bond of the bifluoride (HF_2^-) ions. The strength of the F-H-F$^-$ bond is indicated in part by the large shift of the asymmetric stretching frequency from the value for HF (3700 down to ~1500 cm^{-1}) and by the significant broadening of the IR stretching band, which accompanies this shift. Smaller shifts and broadenings are observed for the F-H-F$^-$ bending mode. Such spectral changes are analogous to those observed due to the hydrogen bonding in O-H-O and other systems, but no generally satisfactory explanation for these effects has been established.

In the experiments recently completed, we have studied the dynamics of $NaHF_2$ and KHF_2 in two sets of measurements using complementary spectroscopic techniques. In the first of these we have applied the isotope dilution technique to an infrared study of the characteristics of the stretching and bending modes of the bifluoride salts. We have measured infrared spectra of samples containing various mole concentrations (2 to 80%) of DF_2^- ions. These conditions enable us to study the vibrations of deuterium atoms either uncoupled, or coupled in varying degrees, while the ionic environment and lattice dynamics are essentially unchanged from those of the undeuterated crystal. The results of our measurements clearly indicate that the considerable width of the IR absorption bands assigned to the asymmetric stretching (ν_3) and bending (ν_2) modes of the HF_2^- ion is associated with the coupling of near neighbor HF_2^- (or DF_2^-) oscillators having similar frequencies or energy states. This is illustrated clearly in the spectra in Fig. 1, which show the changes in the infrared absorption band for the DF_2^- bending mode in $NaHF_2$ as the DF_2^- concentration changes. Thus the results observed for the 2% sample (in which the great majority of DF_2^- ions are surrounded by HF_2^- ions with different ν_2 and ν_3 frequencies) may be thought of as representing the "natural" frequency and width for an isolated DF_2^- ion in the sodium bifluoride crystal field. The broad distribution of frequencies observed for the ~80%D sample, however, would seem to reflect a coupling of oscillators of states of similar energy in the IR absorption spectrum. These results also show that in this case at least, not only the width, but the IR peak position appears to be considerably influenced by the coupling effects. Similar spectra effects have been observed for the stretching vibrations in $NaHF_2$ and for both ν_2 and ν_3 modes in KHF_2.

The second part of our study of the dynamics of the bihalide salts involved the study of the lattice vibrations and symmetric stretching (ν_1) vibrations by Raman, far infrared, and neutron inelastic scattering techniques. The Raman spectrum for pure $NaHF_2$ is shown in Fig. 2. The peaks at 145 and 630.5 cm^{-1} are assigned, respectively, to the librational lattice E_g mode and the HF_2^- symmetric stretch. It is interesting to note that the ν_1 frequency of 630.5 cm^{-1} for $NaHF_2$ is 30 cm^{-1} higher than the average ν_1 frequency for KHF_2 indicating a strong effect of the crystal field on the HF_2^- internal modes. A publication is in preparation showing the correlation of these spectra with our neutron and IR results is in preparation.

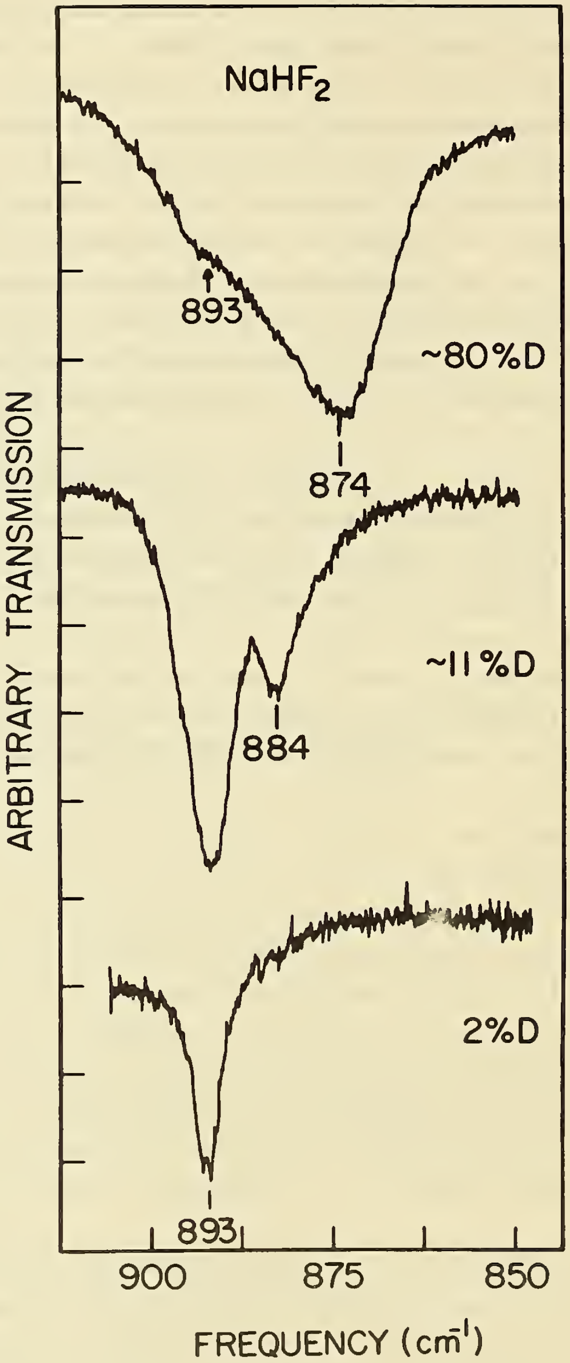

Fig. 1. Infrared spectrum in the region of $DF_2^- \nu_2$ mode for sodium bifluoride containing 2, 11 and ≈80% DF_2^-

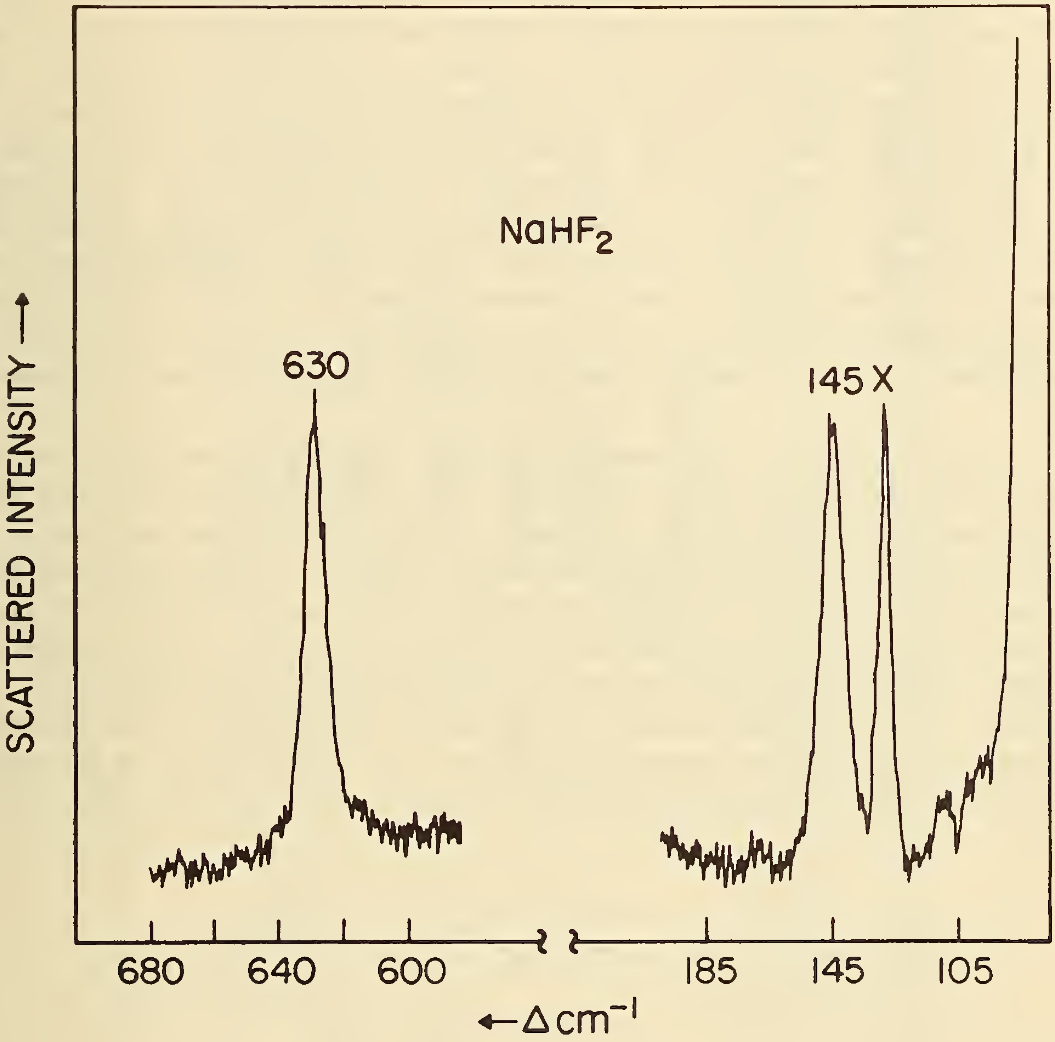

Fig. 2. Raman spectrum of $NaHF_2$. The peak marked (X) is a background line.

Neutron Diffraction Study of the Structure and Thermal Motion of Ph_4Br

L. W. Schroeder and J. J. Rush

As part of a program investigating molecular structure and dynamics of a complex ionic salts, a neutron diffraction study of phosphonium bromide has been completed. This (study) complements previous work in that accurate structural information is now available PH_4Br, PH_4I and the ammonium halides. This information together with the extensive spectroscopic data on these salts makes it possible to perform calculations with various force models to help understand their structural and dynamical properties and related interionic forces.

The tetragonal structure of PH_4Br is similar to that found previously for PH_4I and can be thought of as a distorted CsCl structure. The phosphonium ion, PH_4^+ is surrounded by four nearest-neighbor and four next nearest neighbor bromide ions. The PH_4^+ ion is orientated so that the P-H bond is directed toward the next nearest neighbor bromide ions. This is in contrast to the analogous phase III NH_4Br structure in which the N-H bond is directed towards its nearest neighbor bromide ion. The NH_4Br structure may be visualized from the PH_4Br structure shown in Fig. 1 by imagining the PH_4^+ ion to be NH_4^+ and rotating by 90° about the C axis.

A "rigid-body" analysis of the motion of the PH_4^+ in the phosphonium halides was performed using the anisotropic thermal parameters of the phosphorus and hydrogen atoms derived from PH_4Br and PH_4I diffraction data. As shown in Table 1, the resulting mean-square-amplitudes of translational and torsional motion of the PH_4^+ ion lead to frequencies which are in good agreement with spectroscopic observations. This indicates that the thermal amplitudes from diffraction data, combined with the rigid-body model can give physically meaningful information even under less than ideal circumstances. Having established descriptions of the rotational motion of the PH_4^+ ion in PH_4Br and PH_4I, we are presently attempting to account for this behavior and the somewhat different behavior of the ammonium halides by combined calculations of electrostatic, van der Waals, and short-range repulsive force.

Table 1 Mean-Square-Amplitudes of Translational and Torsional Motion of PH_4^+ in PH_4Br and PH_4I. Corresponding torsional frequencies are also presented for comparison with spectroscopic results at 23°C.

	PH_4Br			PH_4I		
		Torsional Frequency			Torsional Frequency	
Component	Mean-square amplitude[b]	Rigid-body	Spectroscopic	Mean-square amplitude[b]	Rigid-body	Spectroscopic
T_{11}	.035(2)Å^2, [.032(2)]			.043(3)Å^2, [.037(3)]		
T_{33}	.031(3)Å^2, [.027(2)]			.036(4)Å^2, [.030(5)]		
L_{11}	.019(3)rad^2	290 cm^{-1}	343 cm^{-1}	.020(4)rad^2	280 cm^{-1}	326 cm^{-1}
L_{33}	.011(4)rad^2	380 cm^{-1}	N.A.	.013(7)rad^2	360 cm^{-1}	N.A.
S_{12}	-.0031rad-Å			-.0023rad-Å		

[a] Amplitudes given here were calculated from the thermal parameters of Hamilton and Sequeria, and except for L_{33} are similar to the values given by them.

[b] Numbers in parenthesis are estimated standard deviations derived from the least-square fit of the rigid-body model to the U_{ij} of Table III. In this least squares fit all U_{ij} were given unit weights.

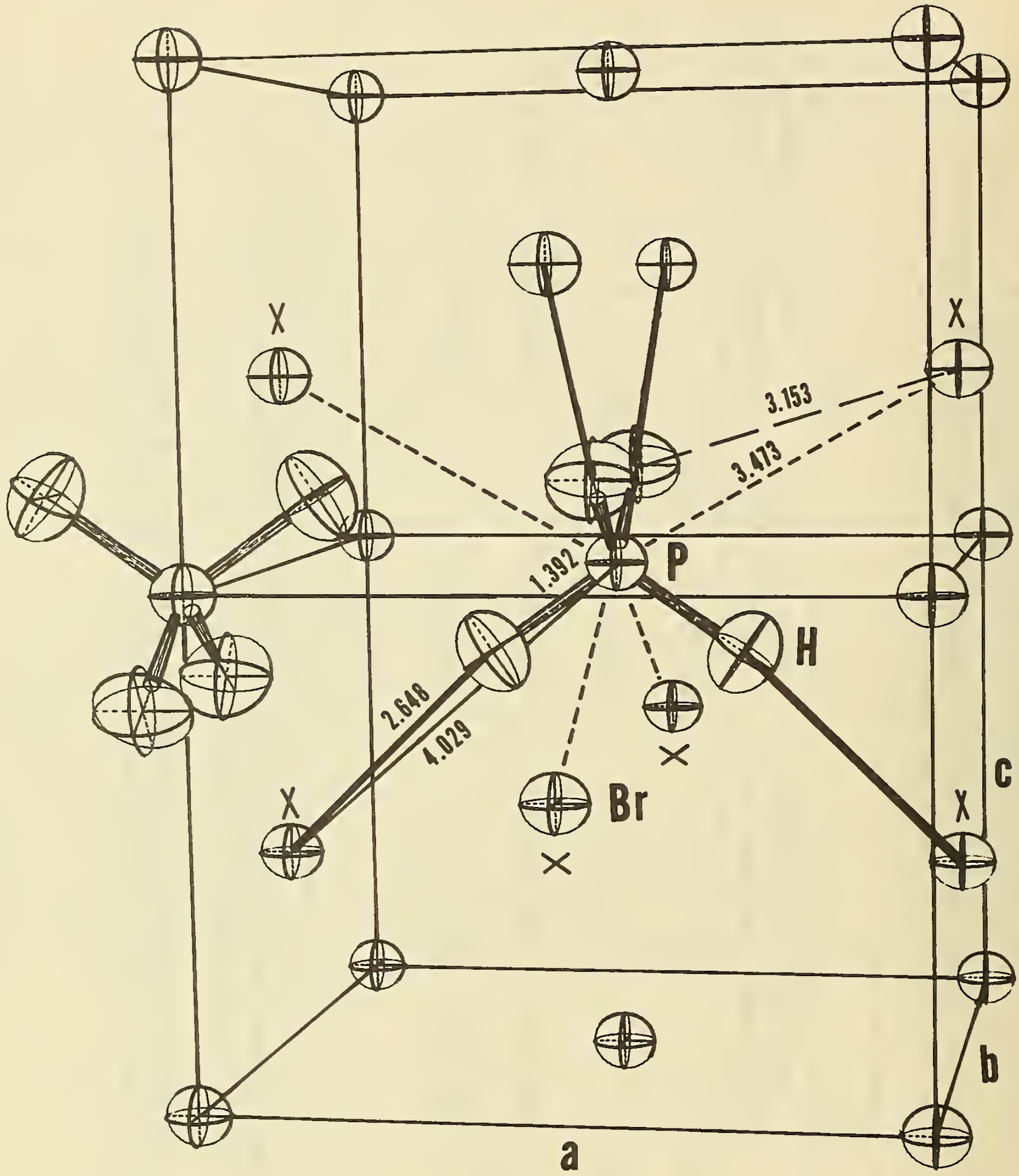

Fig. 1. Structure of PH_4Br showing two unit cells along c. For clarity only the hydrogen atoms for two of the phosphonium ions are shown. Dotted lines show the relation of the phosphonium ion to its four nearest bromide ions which are displaced from the face centers (x). The dark lines from the hydrogen atoms to the four next nearest neighbor bromide ions represent the hydrogen bonds. Numbers are interatomic distances in Angstroms. The axes of the ellipsoids indicate the principal directions of thermal motion for each atom.

Study of Rotational Diffusion in Plastic Crystals

J. J. Rush and L. A. deGraaf

Work has continued on the investigation of the details of molecular behavior in the "plastic" crystalline phases of globular molecular compounds. The analysis of a cold-neutron quasielastic scattering study of rotational diffusion in neopentane in its plastic phase was completed in collaboration with several scientists from Argonne National Laboratory. In addition experiments were planned and begun on the study of the rotational motions in neopentane by analysis of the detailed shape of vibration-rotation bands in the infrared spectrum. It is hoped that a combination of the neutron and optical techniques will provide a more thorough understanding of the plastic phases of this and other molecular crystals and perhaps a better insight into the information provided by both types of measurements.

The Rotator Phase of n-alkanes

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and

J. J. Rush

The NBSR 4-meter time-of-flight (TOF) spectrometer was used to study the energy distribution of neutrons scattered from a thin layer of n-nonadecane supported by a cadmium sheet. Measurements were taken at two different temperatures, 27°C and 20°C. The 27°C temperature is approximately mid-way between the temperature of the "rotator" (plastic crystal) phase transition (ca. 23°C) and the melting point of n-nonadecane. 20°C is somewhat below the transition temperature.

The most striking feature in the data is the presence of a pronounced quasielastic component in the TOF distribution from the rotator phase. This quasielastic scattering presumably reflects molecular motions resulting in rotational jump diffusion of the protons about the molecular chain axis. Earlier workers have observed this quasielastic scattering but have been prevented from making detailed comparisons of experimental data with theoretical models by the vagaries of the beryllium filter technique used in previous studies. Our first attempt at analyzing the nature of the quasielastic scattering consisted of numerically forming the convolution product of a Lorentzian scattering law and the measured instrumental resolution. The resulting widths were then compared with the experimental data in an attempt to estimate the intrinsic width of the Lorentzian line. The widths vs momentum transfer are shown in Fig. 1. These results suggest a jump diffusion process and work is under way to develop a model which will provide a meaningful fit to the observed scattering data.

The inelastic scattering from the same specimen maintained at a temperature of 20°C exhibits a sharp cutoff at an energy transfer corresponding to a frequency of approximately 34 cm^{-1}. This mode has not been previously observed and further study is required to assign its character. For energy transfers greater than 40 cm^{-1} the inelastic portions of the spectra taken at 27°C and at 20°C are, except for quasielastic smearing, nearly identical. The wings of the quasielastic peak tend to obscure the portion of the 27°C data which represents energy transfers less than 40 cm^{-1} due to vibrational modes. A small residual broadening is found in the elastic peak of the 20°C data. The magnitude of this broadening is too small to be assessed with any accuracy by the procedure used for the 27°C data.

The actual data taken so far is too limited in scope to provide clear answers to questions regarding the molecular motions exhibited by n-alkanes in the rotator phase. The data does, however, show that the neutron scattering technique is useful in obtaining information regarding molecular motions associated with phase transition in these compounds, and it also points to the directions which should be taken in future experimentation. Further experiments covering a wider range of momentum transfers and temperatures and utilizing improved resolution are in preparation and will be carried out in the near future. Substantial efforts are under way in developing models to descirbe the scattering to be expected from these systems and in numerically estimating the fit between the models and the data. A particularly acute problem in this connection is to be found in separating the "inelastic" (vibratory) and the quasielastic contributions to the scattering. A numerical procedure for carrying out this separation together with the unfolding of instrumental resolution effects from the experimental data is at an advanced stage of development. This program (named SLAW) should eventually prove useful to others using TOF instrumentaion. It has also been necessary to develop program complexes for handling paper tape data (TAPUTL) and preliminary reduction of TOF data (SCATT).

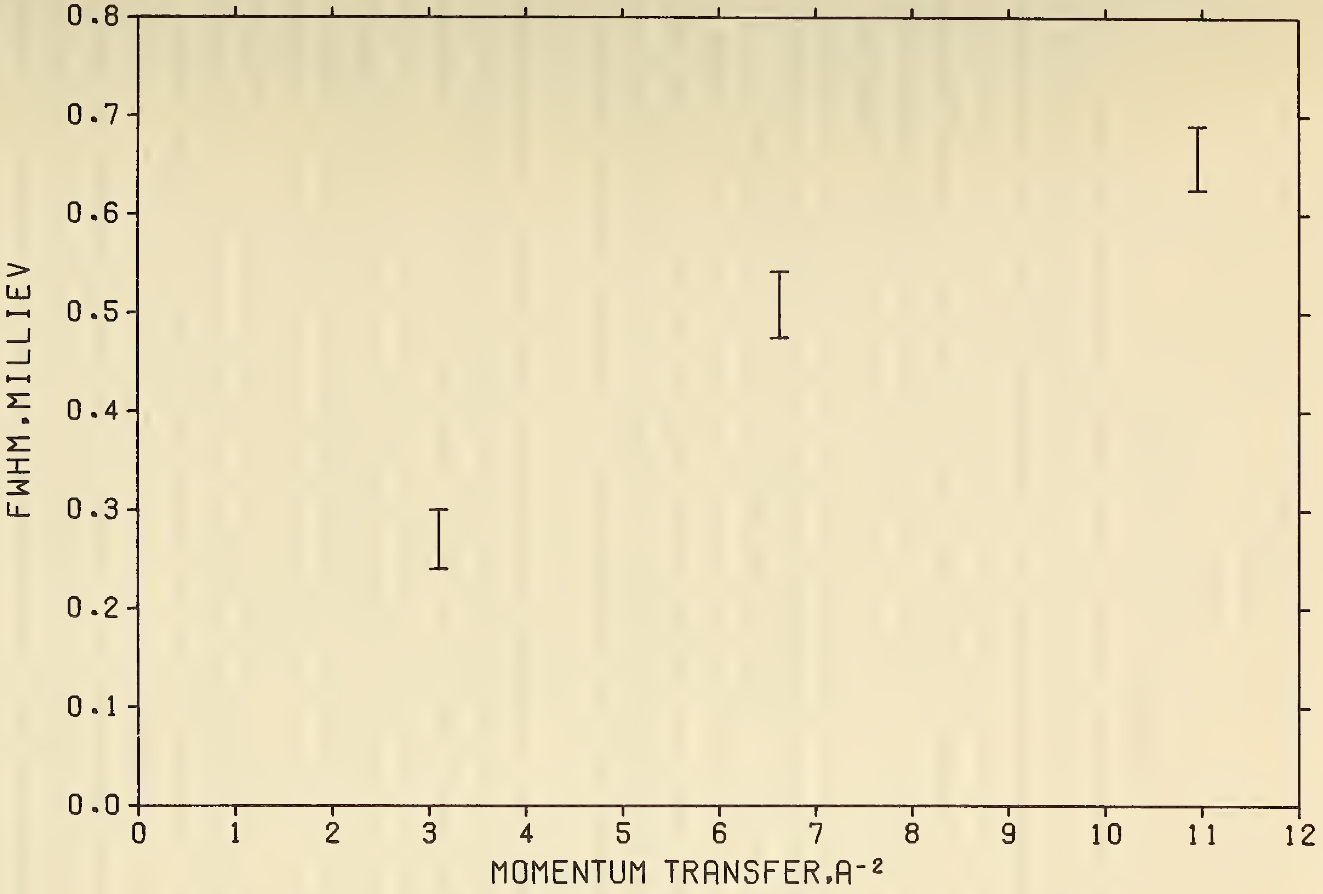

Fig. 1. Full width at half maximum at Quasi-elastic Scattering Law for n-Nonadecane at 27°C as a function of momentum transfer squared.

Study of Simple Liquids

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and
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A study of liquid neon at fixed temperature and various pressures was initiated following the suggestion of R. Mountain and H. Raveche.′ Theoretical calculations made by the above indicated that information about the three-body correlation function could be obtained from the density derivative of the radial distribution function of the liquid. The radial distribution of a liquid can be derived from either x-ray or neutron diffraction patterns, the structure factor, of the liquid. We chose to investigate liquid neon because new thermodynamic data characterizing the liquid was available, a theoretical description of the liquid was considered favorable, neither x-ray nor neutron data of the liquid was available over a sufficient range of parameters to allow a density derivative of the liquid structure factor to be obtained. Because neutron data was likely to yield the most information on the liquid structure factor we wanted the system to have favorable nuclear properties. The latter condition was met best using neon since the neutron scattering was almost all coherent and absorption small so that corrections to the data could be kept to a minimum number of reasonably well known quantities and the recourse of going to expensive isotopes was unnecessary.

A sample cell having high strength and an isotropic diffraction pattern was needed to insure the most favorable signal to noise ratio. We constructed a cell from a titanium alloy (13V 11Cr 3 Al) with very high strength at low temperatures and a very small coherent cross-section. Because of the above two properties, the cell would exhibit structure in a diffraction pattern one-tenth that of an equivalent mechanical strength aluminium alloy. The cell was constructed so as to have 20 mil walls in the beam direction and tested at room temperature at pressures above those decided upon for the experimental conditions. The alloy has higher mechanical strength at low temperature so that we felt secure with the mechanical safety aspects of the cell. The cell was attached to the copper temperature controlled block of our cryostat and the temperature of the block maintained to a few milli-degrees around 35.05K which was fixed for the experiment. Neon was introduced into the cell via a capillary leading through the vacuum chamber of the cryostat to a relief valve, bursting disc and close-off valve and thence to the filling system consisting of a high pressure neon bottle, Heise Gauge for pressure measurements, traps, and a vacuum system. Gas volume outside the sample wall was kept to a minimum to reduce any pressure fluctuations arising from possible temperature changes in the room. The pressure was recorded when the cell was filled and thus a known density of neon determined. The cell and safety apparatus was then sealed-off from the remainder of the system to minimize pressure fluctuations.

Liquid neon neutron diffraction patterns at 35.05K and three pressures 21.4 atm., 79.0 atm., and 140.0 atm. were taken on the BT-2 spectrometer at the NBS reactor in $\kappa=4\pi\sin\theta/\lambda$ scans. The diffraction scans were obtained using 3 different wavelength neutrons, 2.468, 1.062 and 0.745Å. The 2.468Å beam was obtained with an oriented graphite monochromator and 2.5 inches of oriented graphite filter. The higher order contamination was 1/1000 of the main beam. The shorter wavelengths were obtained with a copper monochromator using (111) and (002) planes in transmission. The resolution of the system for the three wavelengths was 0.08Å^{-1}, 0.21Å^{-1}, and 0.36Å^{-1} for the three respective kappa scans from 0.2 to 3.5, 3.0 to 9.2, and 9.0 to 13.25Å^{-1}. Data was accumulated at the three different wavelengths so as to give a counting statistical accuracy of 0.2%. Overlap was provided for in the three regions in order to match the patterns. Data for three pressures and a background run were taken at each wavelength.

The data was refined by first subtracting the background including the scattering from the cell from each pressure run. The data was then matched from region to region, multiple scattering corrections were applied and Placzek corrections for inelastic scattering as well as incoherent scattering were taken into account. The final result is then the resolution smeared structure factor approaching unity at large kappa. This latter condition was imposed upon the corrected data instead of relying on an absolute cross-section determination using the wavelength dependence of the monitor efficiency and the imperfectly known properties of the monitor and crude collimation. The final result of all this effort can be seen in the accompanying graph of the structure factor for one pressure scan. The data plotted there was a result of cubic interpolation between experimental points taken at 0.05Å^{-1} intervals from 0.2 to 6.0Å^{-1} to yield data at 0.05Å^{-1} intervals. A slight uncertainty in the wavelength for the two shortest wavelengths necessitated a redetermination of the wavelengths used from copper powder patterns and thus the data from 3.2 to 13.24 is the result of interpolation. Furthermore, the data was smoothed by fitting 5 points to a parabola via least squares. This smoothing procedure would only affect the statistical fluctuations which would hardly be observed in a graph of this scale as noted by our decision to obtain high statistical accuracy. In a graph of the data on an expanded vertical scale one would tend to see the effects of smoothing.

One of the most interesting features in this graph is the observable structure beyond 6Å^{-1}. This has not been observed in any x-ray data nor previous neutron work. Not shown here is the structure factor for the other pressures. A density (pressure) derivative of the data that will show well defined structure leading to significant indications of three-body correlation functions is readily seen in a comparison of the three pressures.

As a comparison study to the diffraction pattern of the liquid neon, we undertook inelastic neutron scattering experiments on liquid neon at 35.05K and 21.4 atmospheres. This very recently completed study should improve on available inelastic neutron scattering data that was limited to small kappa values. We have accumulated data at ten scattering angles equivalent to momentum transfers for elastic scattering ranging from 0.35 to 4Å^{-1}. Analysis of these results has just begun and will be reported in a future publication.

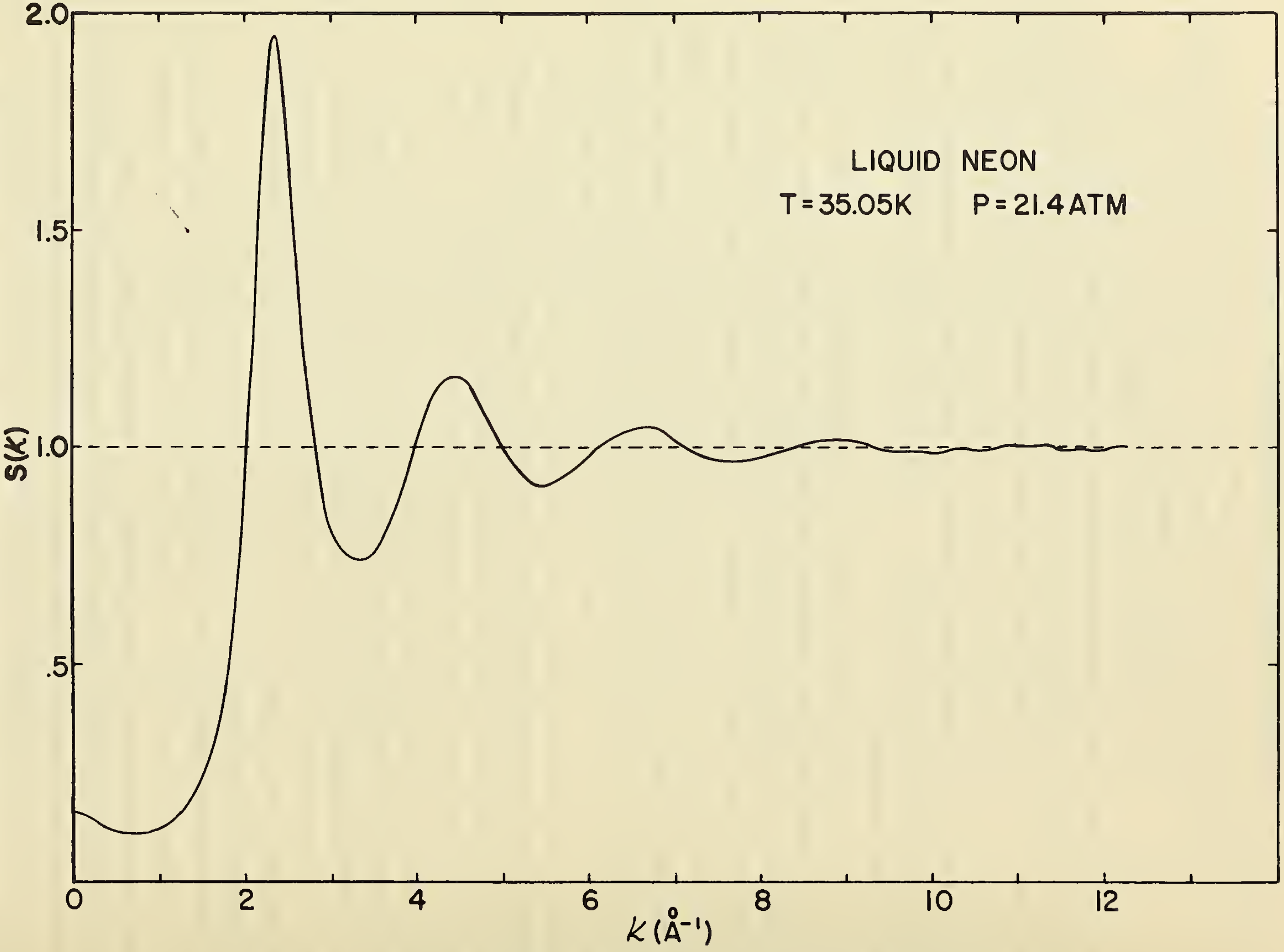

Fig. 1. The liquid structure factor of Neon at 35.05K and a pressure of 21.4 atmospheres. The statistical accuracy of the curve is 0.1% for K less than $3.00 Å^{-1}$ and 0.2% for K greater than $3.00 Å^{-1}$.

Crystal Monochrometer, Time of Flight Spectrometer

B. Mozer
(Inorganic Materials Division)

and

A. Cinquepalma and A. Tudgay

A very versatile spectrometer is being installed at the BT5 beam port. The system consists of a basic two-axis spectrometer plus equipment to do energy analysis by either time of flight or three-axis (crystal) spectrometry. The monochromator is housed in a drum which can be rotated to provide a variable energy monochromatic neutron beam. Both rotation of the drum and the monochromator is accomplished using stepping motors controlled by the computer. In-pile collimation is available with use of a rotating collimator and shutter system of the BNL design. The in-pile collimator is 48 inches long with minimum collimation of 3 minutes of arc and a variety of neutron filters available as inserts in this collimator. Exit beam collimation is accomplished using a Soller slit collimator 36 inches long with vertical slits whose minimum collimation is 10 minutes of arc.

The sample table support mounted is outboard via a cantilever support attached to the drum and both sample table and counter arm are moved by stepping motors controlled by the computer.

Currently the spectrometer is being used in the time of flight mode for energy analysis. The time of flight system consists of a curved slit Fermi chopper, an evacuated flight path roughly four meters long, and a detector bank of eight one inch ^{3}He counters filled to 10 atmospheres. Several rotors are available with various resolution; the best resolution rotor is one with a burst time giving 2.0% resolution (full width at half maximum) at one Angstrom and a peak in its transmission at two Angstroms at a rotor speed of 10,000 rpm. The duty cycle of this rotor is a few tenths of one percent. Other rotors have higher duty cycles and a worse resolution but are used in scattering experiments for energy loss of the neutron where the rotor resolution may not be important compared to the spread in the incoming beam.

The time of flight system is rotated about the sample position and can cover scattering angles from 0 to about 100 degrees. Movement of the whole time of flight system is done by supporting the system on air floats which provide an almost frictionless motion and movement accomplished with a wheel driven by a stepping motor controlled by the computer. Thus one can program the time of flight system to automatically scan a given angular range while storing data taken from the bank of eight detectors either summed, in each detector, or another combination. Each detector counter subtends a scattering angle of approximately 20 minutes of arc. Thus it is possible to program the time of flight analyzer to take data from single crystals in a mode of operation that allows a scan of phonons or magnons along their dispersion curve using constant Q or constant E. The precision of the scattering angle position of the time of flight system about the sample is determined to within a few hundreths of a degree at present, but this can be improved.

The crystal analyzing mechanism needed for converting the system to standard 3 axis work is being designed and hopefully to be tested before the next fiscal year. A simple design is contemplated for the analyzing crystal and counter using some of the ideas in the Canadian and Swedish designs.

Time of Flight Data

B. Mozer
(Inorganic Materials Division)

Data taken with the time of flight system to be fully reported upon in future publications include the behavior of oriented pyrolytic graphite as a neutron filter. This recently manufactured graphite made by Dr. A. W. Moore of the Union Carbide Corporation was loaned to us to look at material having 0.6 degree misorientation and 3.5 degrees misorientation. Filters made of either 0.6 degree or 3.5 degree misorientation were highly satisfactory. The 3.5 degree graphite is now used for one of our filters since cost is important. Comparison of our transmission data on the 3.5 material can be made with data taken by other workers on 7-10 degree material. We find much sharper behavior of the structure in the data as expected. We have made a careful comparison of the ratio of the higher order Bragg contamination in a beam of 2.5 Angstrom neutrons with and without the pyrolytic graphite filter. Without the filter the higher order contamination is of greater intensity than the 2.5 Angstrom beam. Using 2.5 inches of the 3.5 degree oriented graphite as a filter we find the ratio of the second and third order contamination less than 1/1000 of the primary 2.5 Angstrom neutron beam. The use of a oriented graphite monochrometer with a filter for removing order contamination results in a very clean beam of high intensity for inelastic neutron scattering work.

Inelastic neutron scattering data has been taken on vanadium and a specially prepared copper-nickel alloy both of which scatter neutrons incoherently. The vanadium data allows us to compare measurements from our time of flight system with measurements on other equipment. We find the data obtained with the low resolution rotor to be very similar to results obtained by others and by ourselves on other equipment. The results for the copper-nickel alloy are very interesting in that the spectrum shows more structure than that of vanadium, and is significently different than either pure copper or pure nickel.

Cold-Neutron Time of Flight Facility

R. S. Carter, A. A. Cinquepalma, L. A. deGraaf, F. J. Shorten, J. B. Sturrock

The cold neutron time of flight facility is designed for inelastic neutron scattering studies of solids and liquids. A neutron beam from the reactor is filtered through polycrystalline beryllium to eliminate neutrons with wavelengths smaller than 3.96Å. The beam then passes through a four rotor phased chopper system which chops the beam into short bursts of neutrons of selected energy. The sample to be studied is placed immediately after the choppers and the scattered neutrons are detected in a large bank of detectors arranged along the arc of a circle with a 2.4 meter radius. The detector information is fed into a multichannel analyzer and the neutron energy determined from the flight time of the neutrons from the sample to the detectors.

The eight inch thick beryllium filter is kept at 80°K by a liquid nitrogen system. The filter and its associated liquid nitrogen cooling system has been installed in beam hole CT-W of the reactor along with its associated collimator and water shutter. A mechanical rotating shutter using heavy met (density ~18 g/cm^3) to stop the beam has also been installed.

The four rotor phased chopper system has been completed and is in the final stages of testing. The system consists of two concentric counter-rotating rotors in a single vacuum housing and two rotors in a second housing with their axis displaced so the rotors overlap each other by about four inches. The concentric rotors chop the incident beam into bursts of neutrons which spread out in time as they travel the one meter flight path to the second pair of rotors. The second pair of rotors then selects a particular neutron energy depending on the relative phase between the first and second pair of rotors. The rotors are solid aluminum discs, 24" in diameter, 2" thick at the hub and tapering to 1/8" at the periphery. In order to get the shortest burst of neutrons possible for a given slit width, it is necessary to have the chopping faces of the final rotor pair as close as possible. Since they are tapered, the off-set axis system is used. The rotors are driven by two phase synchronous motors at 12,000 rpm by a single high stability oscillator. The oscillator signal is sent through four channels which contain power amplifiers. Vector resolvers in each channel allow the phase of each rotor to be independently controlled. The slits on the rotors are defined by a thin layer of gadolinium and epoxy. Each rotor has several slits of varying width. The slits are arranged in such a way that burst time, choice of one or two bursts per revolution, and neutron energy can be selected simply by changing rotor phase. The choices of final burst width are 6.5 μs, 13 μs and 26 μs FWHM. Spin testing of all rotors to 13,000 rpm has been completed and two rotors have been successfully phased. The measured phase jitter over periods of hours is between one and two microseconds full width at half maximum. The chopper system will be installed at the reactor within the next few months.

The detector bank consists of 24 groups of five BF_3 detectors each. The detectors are 1" diameter by 18" active length and filled to 170 cm Hg. Normally there will be three groups of detectors at each angle yielding eight angles between 10° and 100° from the beam direction. The detectors will be shielded by a completely closed room with one foot walls of hydrogenous material lined with boron carbide. The room will be mounted on air pads so it can be easily moved to provide access to the chopper system. The detectors have all been checked out and the frame to support the detectors designed. The design of the shielding room has been completed and construction will begin shortly.

During this reporting period, programs for use on the Univac 1108 computer have been developed to provide preliminary reduction of data. These programs have been extensively tested from data obtained on the TMC TOF analyzer and found to be generally satisfactory. Additional development to improve their efficiency is planned.

Cold-Neutron Source

R. S. Carter, E. M. Guglielmo, and L. A. deGraaf

The NBS reactor includes a special port, 22" in diameter, which penetrates the reflector up to the edge of the fuel elements. This large opening was designed to provide space for the insertion of a cold neutron source close to the core. By inserting a large, low temperature moderator it is possible to significantly increase the available intensity of very low energy neutrons ($\lambda > 4\text{Å}$) which are very useful for many types of molecular dynamic studies.

The moderator will be a cylinder of D_2O ice 14" in diameter by 12" long. A small amount of H_2O may be added to optimize the moderating property of the source. The source will have a reentrant hole 4" deep by 8" in diameter for beam extraction. The hole will be filled with beryllium which has the property of reflecting the higher energy neutrons back into the moderator while passing the lower energy neutrons. The moderator will be maintained at 25°K by cold helium gas passing through tubing in the moderator. The vacuum cryostat containing the moderator will be constructed of thin-walled aluminum to minimize heating by capture gamma rays. The cryostat will be mounted on a shielding plug and the whole assembly will role inside a second plug and shield assembly which itself will role into the special port of the NBSR. The shield mounted on the second plug surrounds the cryostat and consists of about 4" of lead and bismuth which greatly reduces the gamma ray heating of the moderator. Two 6" and two 2" diameter beams can be extracted from the cold source through the reactor shield. One of the 6" beams will be used for the cold neutron time-of-flight facility while the others await future development.

The helium for cooling is provided by an all helium refrigerator using a turbine expansion engine developed by the NBS cryogenic laboratory in Boulder. It has a one kilowatt capacity at 25°K which is sufficient to handle the heat generated in the moderator at twice the current reactor power.

The refrigerator is complete and operational. The lead and bismuth shield is almost complete and should be ready for final inspection within a month. The cryostat is also under construction and should be ready for shipment within two months. Extensive testing of the system will be required as well as a modification of the reactor Technical Specifications (a document defining the limits of operation of the reactor which is part of the AEC license). Once in place additional testing will have to be undertaken to study the radiolysis of the ice to insure the safety of the facility.

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Crystal Structure of the TiNi Martensite

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TiNi undergoes a crystallographic transformation just above room temperature associated with a novel "shape memory" effect, in which the material, deformed in the low temperature phase, will revert to its original shape if heated above the transition. Understanding this phenomena requires a knowledge of the low temperature phase, which is extremely complicated. A model has been proposed, based on x-ray diffraction measurements, in which certain shearing motions of the atoms cause strains whose relief provides the mechanism of the shape memory. Since Ti and Ni atoms have a very different scattering amplitude for neutrons, neutron diffraction measurements were taken throughout the transition region, and were found to confirm the x-ray model.

Spin Canting in Rare Earth Iron Garnets ($R_3Fe_5O_{12}$)

S. Pickart and H. Alperin
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The accepted picture for the microscopic spin alignment in these technologically important materials has been a collinear arrangement, with a net moment resulting from unequal antiparallel rare earth and iron sublattices. However, due to the large component of the rare earth moment arising from the orbital electron motion, it was predicted theoretically that the rare earth moment would tilt away from the direction of the net moment under the influence of its non-cubic local environment. These arrangements have now been clearly detected by low temperature neutron diffraction measurements in Dy, Tb and Er iron garnets. Measurements of the degree and direction tilt obtained from these results are shown in Table I. (ϕ_s and ϕ_s' are the angles from the [111] direction measured toward the [001]). These results are in accord with theoretical predictions except for the case of Dy^{3+}, for which ϕ_s', should have been negative. These results, particularly if the latter discrepancy can be resolved, will give further insight into the fundamental magnetic forces acting in these materials.

TABLE 1. Values of the canting angles and rare-earth moments in iron garnets.

	Angle from [111]		Moment
Compound	ϕ_s	ϕ_s'	(μB)
DyIG	38	3	8.8
ErIG	43	5	6.0
YbIG	40	25	1.9

Magnetic Structures of Transparent Fluorides

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Powder neutron diffraction data have been taken on two transparent fluorides, $RbNiF_3$ and $CsFeF_3$, that are of interest because they possess a macroscopic magnetic moment and thus offer the possibility of light modulation by means of the Faraday effect. These compounds are hexagonal ($BaTiO_3$ type), with the transition metal cations, Ni^{2+} and Fe^{2+} located on crystallographically inequivalent sites, the 2a and 4f sites of space group $P6_3/mmc$. The magnetic structures determined by the neutron data are ferrimagnetic, i.e. the basic alignment of the magnetic spins is antiparallel, but since the oppositely directed spins are located on unequal numbers of the inequivalent sites, a net resultant moment is observed. The structures consist of moments of $2.2\mu_B$ for Ni^{2+} and $4.4\mu_B$ for Fe^{2+}, lying in the basal plane in $RbNiF_3$ and tilted about 15° out of the plane for $CsFeF_3$. This latter result is especially interesting because it differs greatly from the easy axis observed by magnetization measurements (about 25° from the c-axis) performed in fields of 10 Wb/m^2 single crystals. One thus has the unusual possibility of an anisotropic field that is field or strain dependent.

Crystal Field and Exchange Energy in Cubic Praseodymium Compounds

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Susceptibility measurements indicate that $PrSn_3$ becomes antiferromagnetic at low temperature while $PrIn_3$ does not. Since both these compounds have the cubic order $CuAu_3$ structure with lattice parameters differing by <1%, the speculation was made that exchange energy (which favors ordering) and crystal field energy (favoring a nonmagnetic singlet ground state) are delicately balanced in these compounds. To confirm the susceptibility measurements, neutron diffraction patterns were taken, indicating a simple antiferromagnetic structure with alternating spin layers in $PrSn_3$ and no ordering in $PrIn_3$ down to 2°K. Since the Pr^{3+} spin lattice structure is simple cubic, the crystal field calculations can easily be made, so that further planned neutron measurements of the spin density distribution and inelastic energy spectrum offer a rare opportunity for detailed comparison with theory.

Neutron Diffraction in Liquid Aluminum

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and

C. Davis
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and

S. J. Pickart
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The microscopic state of a liquid is described in terms of the pair correlation function. This function may be obtained by means of neutron diffraction. In the past the statistical theory of the liquid state had not advanced far enough to permit the calculation of the pair correlation. Recently, however, new theoretical approaches to this problem have been developed which are successful in the case of the alkali metals. Hopefully these techniques may also be applied to polyvalent metals for which pseudopotential theory applies. In the past year measurements in aluminum have been carried out near the melting point (660°C) and at 1000°C. The data collection has been completed, and the data are now being analyzed for comparison with theory.

Neutron Diffraction Investigation of Amorphous Solids

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Amorphous solids give diffraction patterns consisting of a small number of relatively weak maxima and these patterns may be analyzed to reveal information about the structural arrangement of the atoms which comprise the solid. Early diffraction studies indicated that in amorphous materials the immediate neighborhood of the atoms is similar to the geometric arrangement found in crystals but order over large distances is absent. Glasses are examples of amorphous solids and are of great importance in modern technology. Although the vitreous state is a rare one, glasses have been formed of a variety of materials such as mixtures of borates, phosphates and silicates.

Silica glass has been investigated extensively due to the fact that it has been considered to be a basic representative of the glassy state. In published reports of structural investigations of vitreous silica using the neutron and x-ray techniques, discrepancies have appeared which now require clarification before the structure of this basic glass is fully understood. A program designed to obtain a precise measurement of the diffraction spectra (neutron and x-ray) over an extended range of momentum change of incident particle (neutron and photon) has been undertaken. A portion of the program (i.e. the neutron diffraction spectrum) is being conducted through interagency collaboratory between The Laboratory for the Structure of Matter, U. S. Naval Research Laboratory and the Reactor Radiation Division, National Bureau of Standards.

The precisely measured diffraction spectra obtained from this program, together with newly devised data reduction procedures, have yielded new structural information and insights into the discrepancies alluded to previously. From these studies there is a clear indication that a greater degree of ordering exists in the structure of silica glass than has heretofore been supposed. Corresponding maxima have been identified beyond 10 A in the radial distribution curves for both the neutron and x-ray diffraction experiments. This result is to be compared with previous beliefs that such significant maxima do not exist beyond approximately 5 A.

An extensive study is being carried out to determine the nature of the ordering. This study includes an effort to construct a three-dimensional structure of partially ordered silicon-oxygen tetrahedra which satisfy pertinent physical criteria of the glass as well as the radial distribution curves obtained from the diffraction measurements. The techniques developed in this program will be employed in the investigation of other simple amorphous solids.

Precision Measurement of Thermal Neutron Coherent Scattering Amplitudes

C. S. Schneider
(U.S. Naval Academy, Annapolis, Md.)
and
E. Prince

This project proposes to measure the forward coherent scattering amplitudes of several elements as manifested by refractive bending of a thermal neutron beam incident at a few degrees arc from a rectangular prism. The resulting bending of almost a minute of arc is detected within a few milliseconds arc using a torsion goniometer[1] control on the second, analyser crystal, of a double perfect crystal spectrometer. The measured refractive bending angle is

$$\Delta\theta = \frac{4N\lambda^2 b}{9\pi} (\tan\phi + \cot\phi)$$

where N is the atomic density, λ the neutron wavelength, b the coherent scattering amplitude and ϕ the angle of incidence.

The reactor port, including collimation and shielding was first built as sketched in Fig. (1). Special care was taken to design and fabricate the internal beam collimator to optimize beam intensity and minimize stray, or background, radiation. The tunnel was made quite large so that the neutron beam could reach the experimental area with essentially no scattering and the beam catcher sufficiently massive to receive the entire direct neutron beam with negligible leakage. Finally, the detector shielding exploited the two stage effect of thermalizing the neutrons in the outer hydrogen-rich shielding and then capturing them in the highly absorbing boron carbide powder. The background count rate of ten neutrons per minute achieved thus far is quite satisfactory.

The spectromter is mounted on a seven-ton base which floats upon a pneumatic suspension yielding a vibration resonance of 1.5 Hz thus effectively eliminating all noise coming from the floor. Other noises, coming from the surrounding air, are effectively reduced by the spectrometer cover, a three layer box of 1/4" Aluminum, 2" styrofoam and 1/2" acoustic tile,

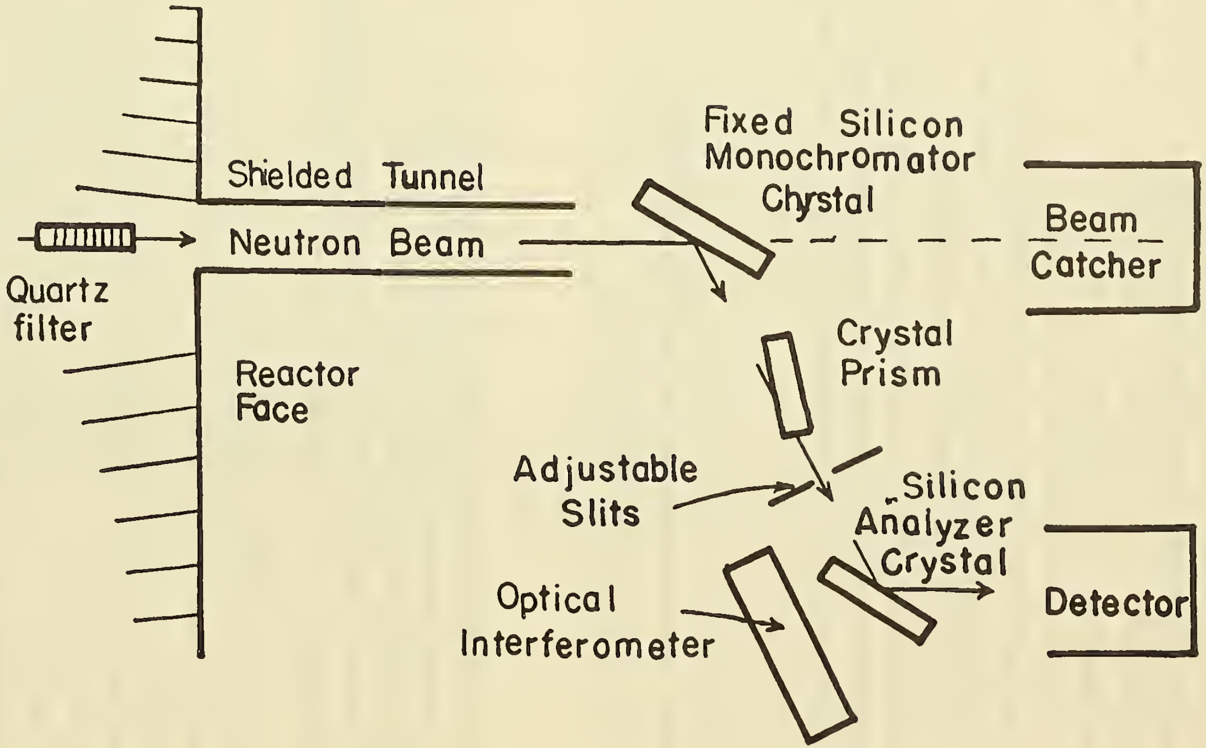

Fig. 1. Double Perfect Crystat Spectrometer.

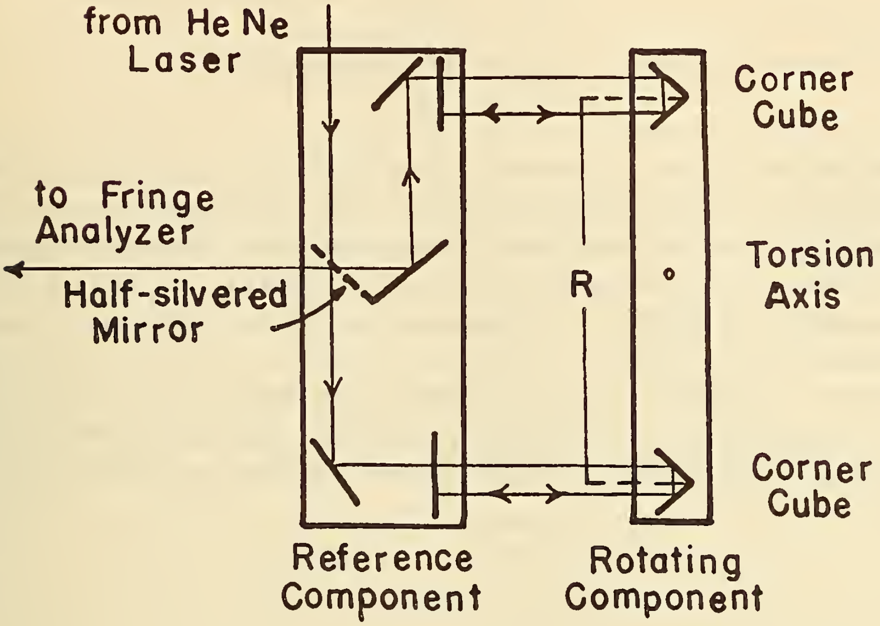

Fig. 2. Angle Measuring Interferometer.

all bolted and glued together to form insulation from both mechanical vibrations and thermal changes, which affect the crystal and angle measuring interferometer operation.

This interferometer was used to calibrate the angle control system[2]. The paths of the HeNe laser light are shown in Fig. (2) and the path difference is

$$n_f = (4R \sin\theta)/\lambda$$

which for small angles θ yields a calibration of 0.10717 seconds arc per fringe. The angular control of the analyser crystal was achieved using a torsion goniometer[1] in which a precision micrometer deflects the end of a glass strip eleven inches long. The resultant torque is applied to a stiff column of aluminum alloy and results in angular control with 10^{-3} seconds arc over a range of 10^2 seconds arc. The calibration of this micrometer-angle instrument is now being determined.

The spectrometer completed to date allows manual measurements of the neutron refracted angles to the desired accuracy. Work continues, however, to complete the computer automation and obtain a crystal of proper mosaic width to complete the wavelength calibration.

[1] The original version of this instrument is described in C. G. Shull, K. W. Billman and F. A. Wedgewood, Phys. Ref., 153, 1415 (1967).

[2] The original version of this instrument is described in J. G. Marzolf, Rev. Sci. Instr., 35, 1212 (1964).

Picatinny Arsenal-NBS Interagency Collaboration

J. J. Rush and R. S. Carter

and

C. S. Choi, H. Prask and S. F. Trevino
(Explosives Laboratory, Picatinny Arsenal, Dover, N.J.)

This group of scientists joined the staff of the Reactor Radiation Division in early September 1970, in a cooperative research program similar to existing effort with NOL. The establishment of the relationship was occasioned by the shutdown of the Army reactor in Watertown, Massachusetts, where the work had been previously carried out. The principle al activity at the Bureau to date has involved the design of the modification required to install the instruments of the reactor. These instruments include a triple axis spectrometer and a two axis diffractometer, both of which will be serviced by a single beam tube. In addition, a second beam tube will be developed which will also serve two spectrometers.

The interests of the group have been in using the neutron scattering techniques in the study of the structure and lattice dynamics of materials. In particular, those materials which exhibit metastable properties are under investigation in an attempt to characterize their properties and hopefully discover the cause of the instability. Neutron diffraction is an essential complement to x-ray diffraction in the structure studies, for many of the materials are composed of both heavy and light atoms, in which case only the heavy atoms can be located with x-rays whereas neutrons can locate both. In addition, x-rays decompose the sample crystal in some cases so rapidly that the measurement cannot be completed. Because of their relatively low energy (~.025 ev) neutrons with wavelengths comparable to interatomic spacing (~1.8Å) can also be used to study the dynamics of materials while x-rays are limited to structure studies only.

Materials other than unstable ones have also been studies and these include, for example, polymers, hydrogen bonded systems, ionic crystals, molecular crystals, molecular liquids, surface adsorption, crystal hydrates and metal hydrides.

Diffractometer Control by Computer

E. Prince

and

H. A. Alperin
(Naval Ordnance Laboratory, White Oak, Md.)

The NBS system for control of diffractometers has been in routine operation for more than a year. The system allows the control of diffractometer experiments by sharing time on an XDS 925 computer. The computer handles the collection and reduction of data, in real time, by means of programs written by the users in FORTRAN language. This represents a large advance over previous, computerized control systems, which have required laborious, assembly-language programming.

The program for the collection of single-crystal diffraction data approaches the problem of time utilization by making a preliminary measurement of the peak intensity in each reflection. If this intensity is not greater than background by a statistically significant amount, an upper limit to the possible intensity is computed, and the program proceeds to the next reflection. On the other hand if the intensity is so large that only a short counting time is necessary to achieve adequate statistical precision, the time spent scanning a reflection is reduced by a factor of up to 6 from the standard counting interval.

The system originally controlled four neutron diffractometers and one x-ray diffractometer. During the past year additional hardware has been installed to handle a fifth neutron diffractometer, and hardware for two more is on order.

A 14 minute, 16 mm sound film entitled, "Diffractometer Control System," describing the control system may be obtained on free loan by addressing a request to Office of Technical Information and Publications, National Bureau of Standards, Washington, D.C. 20234. A detailed description of the system is contained in a forth coming paper.

Equipment Development

S. J. Pickart and R. H. Williams
(Naval Ordnance Laboratory, White Oak, Md.)

During the past year two major items of equipment have been brought close to completion, namely the triple-axis spectrometer and the superconducting magnet.

1. Triple Axis Spectrometer

Elementary excitations of many types in solids and liquids (phonons, molecular spectra, and magnetic spin waves) can be conveniently studied at a reactor because their energies and wavevectors match those of thermal neutrons. An instrument to perform energy analysis of neutrons scattered from a specimen crystal has been constructed by mounting an analyzing crystal axis on the arm of a conventional double axis (monochromator and crystal) spectrometer--hence the name triple axis spectrometer. All of the items necessary for operation of the spectrometer--motors, counters and shields--have been installed, including highly efficient oriented graphite monochromators and filters. An arbitrary tracking in energy-momentum transfer space (e.g. constant energy transfer, constant momentum transfer, or some intermediate mode) is desirable to optimize resolution during the data-taking process, requiring non-linear scanning of up to five stepping motors. This procedure is performed by a computer program, already written and debugged, executed by the on-line time-sharing spectrometer control system. Inelastic scattering measurements will begin as soon as additional shielding for the top of the drum (to lower the fast neutron background) and an extended beam stop (to allow for variable incident energies) are installed.

2. Superconducting Magnet

Many as yet unexplained magnetic transitions, particularly in the rare earth metals, occur in high magnetic fields (~10.0 Wb/m^2) that are comparable to the exchange and anisotropy forces acting in these materials. To study the detailed nature of the magnetic spin structures in these high-field phases, a superconducting magnet has been designed for neutron diffraction work. It must be of the helmholtz split-pair configuration to provide 360° radial access for the incident and scattered neutron beam. The magnet coils are suspended in an outer dewar containing liquid He, while the sample is placed in an insert dewar extending through the bore of the coils that is temperature controlled from 4.2°K to room temperature. The individual coils for the helmholtz pair have been wound and tested, the magnet and insert dewars are on hand, and the magnet support assembly and coil form are in the final stages of construction. When completed, the magnet will operate at 100 amps to provide a field of 10.7 Wb/m^2 in a one-inch gap with 1-3/8" bore. An additional feature is the winding of the coils in concentric pairs, allowing for removal of the inner set to provide a 6.0 Wb/m^2 field in a 3" bore, if desired for larger samples (e.g. magnetic polycrystalline, amorphous or liquid materials).

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C. Non-Division NBS Programs

Activation Analysis Section: Summary of Activities

P. D. LaFleur and D. A. Becker
(Analytical Chemistry Division)

The Activation Analysis Section of the Analytical Chemistry Division is located in the NBS Reactor Building and is one of the major users of the NBSR irradiation facilities. Complete coverage of the section's activities may be found in it's annual progress report therefore only a brief summary will be included here.

In the past year, a study of the operating characteristics of the NBSR irradiation facilities was made, and experimental results reported for neutron flux measurements, and sample temperature and pressure increases under irradiation. These measurements were made in the four pneumatic tube facilities and the vertical thimble long term irradiation facility.

A large number of analyses of various kinds were made by the members of the section. Two sample materials of special interest were the Trace Elements in Glass Standard Reference Material (SRM) and the Trace Elements in Orchard Leaves SRM. Techniques used included both nondestructive analysis using large volume Ge(Li) detectors and analyses employing chemical separations. In addition, the nuclear track technique, in which radiation damage "tracks" from charged particles either from (n,p) or (n,α) reactions in a plastic detector are counted, was used for boron and uranium analysis.

Other work by the section included use of a Cockcroft-Walton neutron generator for 3 MeV and 14 MeV neutron activation analysis, and use of the LINAC for photon activation analysis. Also, eight recently acquired californium-252 neutron sources are presently being evaluated for potential activation analysis use.

Data handling and reduction systems include a computer program, ALSPIS, for peak finding and integration using the UNIVAC 1108, and development of applications of a minicomputer for activation analysis.

Atomic Constants from X-ray and Positronium Decay γ-ray Measurements

R. D. Deslattes, W. C. Sauder, J. A. Hammond and A. Henins
(Optical Physics Division)

From the general expectation that coupled scanning interferometers would work and on the other hand from some fairly sophisticated expectations on the behavior of positronium under strong Zeeman perturbation in low temperature helium, we have developed a large and potentially interesting program of measurement. We expect to measure h/mc to between 1 ppm and 0.1 ppm with relative ease. This will allow inference of the fine structure constant to between 5 in 10^7 and 5 in 10^8.

Within the past year, precision indices have been established for all major components of this exercise. In addition, a controlled environment chamber has been established in an area at the NBS reactor where we can expect to handle the multi-kilo-Curie source required in the h/mc measurement. Scanning interferometry has been shown to go with noise levels of the order 0.01Å. Angle measurement noise has been reduced to about 10 micro arc sec. Density determinations are going at 0.1 ppm.

There are both scientific and technological spin-offs from this work. For technology there are obvious gains from meaningful experience in small distances and small angles. For science, the absolute γ-ray wavelength facility offers the chance for measurement of such reference wavelengths as will be of special use is mesic x-ray wavelength determinations. These, in turn, will lead via a presumed validation of electrodynamics in the case of μ-mesic x-ray from whose energies have been subtracted such contributions as follow from the Coulomb interaction.

Intense Fission Neutron Source

James A. Grundl
(Nuclear Radiation Division)

An intense ^{235}U fission neutron source is under joint development by the National Bureau of Standards and the Los Alamos Scientific Laboratory. A preliminary arrangement for reproducible irradiations inside of the NBS Research Reactor thermal column is complete and a measure of the source strength has been obtained. Such a spectrum is a natural and relevant source of fast neutrons and, with a calibration based on well-studied thermal neutron parameters, it provides a base for activation detector calibration and fast-neutron cross section validation. An initial application of the fission source was carried out under the auspices of the ASTM Subcommittee on Neutron Dosimetry. In April 1970, some dozen laboratories determined the responses of sulfur, aluminum, and fission track detectors exposed to the fission source operating at 5×10^{10} neutrons/second, a limit necessitated by rudimentary source handling techniques. Results from the participating laboratories indicate disagreements which justify additional efforts in this area. The matter was discussed at the Washington meeting of the American Nuclear Society in November 1970.

Parity Violation in Strong Interactions

J. L. Alberi and R. Wilson
(Harvard University, Cambridge, Mass.)

and

G. Scharff Goldhaber
(Brookhaven National Laboratory)

and

I. G. Schröder
(Nculear Radiation Division)

A program to study parity violation due to the weak interaction in the overall nucleon-nucleon interaction (weak forces between nucleons) is underway in collaboration with Harvard University and Brookhaven National Laboratory.

According to the current-current theory of weak interactions proposed by Feynman and Gell-Mann the weak interactions may be expressed as a current-current interaction analogous to the electromagnetic interaction. Unlike the electromagnetic current, which is purely vector, the weak current is the sum of a vector and an axial vector part giving rise to parity violating interactions. The weak current, according to Feynman and Gell-Mann, is composed of two leptonic terms (j_e and j_μ, electronic and muonic, respectively) and two hadronic terms (j_n and j_s, non-strange and strange) which lead to a series of terms in the current-current interaction some of which have been observed. Thus, for instance, j_e, $j_\mu{}^+$ is the term that gives rise to the leptonic decay of the muon, $j_n j_e{}^+$ is the ordinary beta-decay term, and $j_n j_s{}^+$ gives rise to nonleptonic decays of the strange particles. The theory also predicts the existence of the following three terms $j_e j_e{}^+$, $j_n j_n{}^+$; the first gives rise to neutrino-electron scattering while the last two give rise to the parity non-conserving effects which are of interest to the present program, i.e. nuclear states are no longer eigenstates of parity and small parity impurities can be expected.

The reaction we are studying is ^{113}Cd (n, γ) ^{114}Cd and in particular the transitions in ^{114}Cd from the 9.04,1^+ state to the 0^+ ground state and the .56 MeV 2^+ state. Both these are regular parity conserving M1 transitions. The partity non-conserving part is E1. The admixture of the parity non-conserving part is measured by detecting the circular polarization of the emitted gamma rays from these two transitions. The circular polarization is proportional to E1/M1. In Cd due to a favorable nuclear structure this ratio is $\simeq$500 F where F is the ratio of the strength of parity non-conserving and conserving potentials. Taking F $\simeq 10^{-6}$ to 10^{-7} gives an effect anywhere between 5×10^{-4} to 5×10^{-5}.

The apparatus consists of a Cd target at the center of GT-2 in the NBSR. The gamma beam is collimated and passed through a transmission compton polarimeter with an analyzing efficiency of $\simeq$10%. The beam is recollimated and detected by a 4" x 5" sodium iodine crystal appropriately shielded from magnet influences. Hence our count rate change at the detector is anywhere between 5×10^{-5} to 5×10^{-6}. We expect to detect 10^4 gammas/sec in the two photopeaks of the interesting gamma rays. Hence we are just sensitive to the minimum expected effect in 50 days.

Fast Neutron ^{3}He Spectrometer

J. C. Glynn
(University of Maryland, College Park, Md.)

and

I. G. Schröder
(Nuclear Radiation Division)

In collaboration with the University of Maryland the resolution, efficiency and use of ^{3}He semiconductor spectrometers as fast neutron spectrometers in the energy range between 500 keV and 1500 keV have been investigated. Such a spectrometer has been used in conjunction with a specially designed collimator in an attempt to measure the fast neutron distribution in the core region of the NBS reactor. This has been accomplished by placing a scattering sample in the core, and collimating a beam of scattered neutrons directly into the spectrometer. The use of different scatterers enabled a determination of the spectrum in the anisotropic neutron field. The perturbation of this spectrum by the collimator and scatterer could not be accurately determined; however, the present program has succeeded in measuring the perturbed scalar flux in the core region and has provided a known fast neutron spectrum which can be used to test and calibrate other neutron spectrometer systems.

Two-Photon Emission in (n,p) Capture

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(American University, Washington, D.C.)

and

J. L. Alberi
(Harvard University, Cambridge, Mass.)

and

I. G. Schröder
(Nuclear Radiation Division)

A search for two-photo emission in thermal (n,p) capture is being conducted with the aim of establishing the probability of occurrence of this process in an effort to help explain part of the apparent discrepancies between the measured and calculated values of the thermal (n,p) capture cross section. To detect the possible emission of two photons in radiative (n,p) capture two NaI(Tl) detectors, mounted on fast photomultiplier tubes, were placed 180° with respect to each other with an H_2O target between them, and 90° to a neutron beam. (The neutron beam has a better than 4000 to 1 cadmium ratio for a detector with a $1/\nu$ response and is obtained by total neutron reflection down a 4.5 meter long 1 cm diameter Ni coated glass tube.) The simultaneous emission of two photons is ascertained by a fast coincidence circuit, with a time resolution FWHM of 2 nsec, composed essentially of two constant fraction timing discriminators and a time-to-pulse-height (TAC) converter and a slow triple coincidence circuit (30 nsec resolving time) which demands coincidence between the pulse heights from the two detectors and the TAC. The gate from this "fast-slow" coincidence system triggers a linear gate which allows the pulse-height distribution from one of the detectors to be recorded in a 100-channel pulse-height analyzer. In order to measure the two-photon effect the gated spectra are compared with normalized ungated spectra to eliminate the effect of chance coincidences, and with gated spectra obtained with an identical D_2O target to account for the oxygen in the hydrogen target.

Decay Characteristics of Krypton Isotopes

F. J. Schima
(Nuclear Radiation Division)

A program has been initiated into the investigation of the decay characteristics of the isotopes of krypton.

Aluminum foils containing ^{84}Kr ions implanted by the isotope separator were irradiated in RT-4 of the NBS reactor for periods up to 4 hours. The decay characteristics of the ^{85m}Kr produced have been studied. The half-life was found to be

$$4.557 \pm .029 \text{ hr}$$

and the γ-ray energies were found to be

$$151.0 \pm 0.2 \text{ keV and } 304.8 \pm 0.1 \text{ keV.}$$

The half-life is significantly longer (3.4%) than the accepted value in the literature.

An ultra pure iron foil containing implanted ^{80}Kr was irradiated to produce ^{81m}Kr. The decay characteristics determined were

$$t_{1/2} = 13.29 \pm .06 \text{ sec}$$

$$E_\gamma = 190.663 \pm .046 \text{ keV.}$$

Further work in progress on the investigation of krypton decay parameters include the production in the NBS reactor of ^{79}Kr as well as an attempt to produce detectable quantities of 200,000 year ^{81g}Kr. This is being accomplished by a long term irradiation of isotopically enriched ^{80}Kr gas in the reactor gap where the thermal neutron flux is approximately $1.7 \times 10^{14}/\text{cm}^2$ sec.

Nuclear Orientation Studies of ^{82}Br

A. T. Hirshfield, D. D. Hoppes, W. B. Mann and F. J. Schima
(Nuclear Radiation Division)

Samples of ^{82}Br produced in the NBS reactor were implanted in iron foils with the isotope separator of the Radioactivity Section. These samples were used to cryogenically orient the ^{82}Br nuclei and measure the beta- and gamma-ray angular distributions with respect to the nuclear spin direction. A preliminary report of the approximate effective magnetic field for bromine in iron, the fraction of nuclei experiencing the full field, and sign of the magnetic moment of ^{82}Br was given at the International Conference on Hyperfine Interactions Detected by Nuclear Radiation, Sept. 6-11, 1970, at Rehovot and Jerusalem, Israel and is being published in the Conference Proceedings.

Further bromine bombardments are anticipated to determine the effect of lower implantation concentrations and to measure the E2/M1 mixing ratios for 5 gamma rays occurring in the decay. Other activities of immediate interest for orientation studies are ^{86}Rb and ^{186,188}Re.

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REACTOR OPERATIONS AND SERVICE PROGRAMS

In the past year, the NBS reactor has become fully operational. All required reactor testing was satisfactorily completed. Application for a full term license was made to the AEC. Test results and reactor performance analysis were presented before the Advisory Committee on Reactor Safeguards which considered the application in March 1970 with favorable recommendations. A full term 15 year license was issued by the AEC in June 1970.

A. Operating Statistics

During this period more than 6,000 hours of reactor time was provided to users. This is equivalent to about 60,000 megawatt-hours of energy produced. Currently the reactor is operated at the maximum rated power of 10 Mw on continuous a-round-the-clock basis about two thirds of the time. This schedule permits operation with minimum staffing. Reactor performance to date has been excellent. Efficiency and time utilization was very high. Very little time was lost due to malfunctions or unscheduled shutdowns. A summary of operating statistics is presented in the following table.

NBSR Operation Summary
November, 1969 - November, 1970

Reactor Operations to date	78213.3 Mwh
Reactor Operations for year	59186.0 Mwh
Hours of Operation	6000
On-line time at 10 Mw	67%
Number of fuel elements used	44
Average U-235 burnup	34%
Number of Refueling Operations	11
Number of Unscheduled Shutdowns	3
Number of Visitors	4000

B. Operating Summary

The NBSR core employs 30 fuel elements arranged in three hexagonal rings. An inner ring composed of six elements, a middle ring which accomodates another six fuel elements and six experimental thimbles and an outer ring of 18 fuel elements. The initial NBSR core was composed of 23 elements of 170 gm U-235 each and seven elements of 205 gm U-235 each. As operations progressed and new fuel was needed the lighter 170 gm elements were substituted by the heavier 205 gm elements to gain more operating lifetime. The 170 gm elements provided an average operating lifetime of about four days (length of cycle divided by the number of elements replaced by fresh elements to achieve the cycle length). The heavier 205 gm element increased the lifetime time to more than six days. More recently even more heavily loaded elements containing 250 gm of U-235 were fabricated. These elements have just been introduced into the reactor and it is anticipated that they will improve the operating lifetime to as much as nine or ten days per element. The use of more heavily loaded elements was made possible by improved fuel fabrication technology and by special safety analysis leading to the development of a fuel rotation plan that permits their utilization in the NBSR.

The distinct advantage of using heavier elements which more than double the average element lifetime is reflected by a substantial fuel economy which is especially significant at a time when fabrication, shipping and reprocessing charges are escalating. In addition reactor downtime for refueling is considerably reduced.

The NBSR fuel rotation plan utilizes a system whereby the most depleted elements located in positions of highest flux in the center of the core are removed from the core for replacement. Then the remaining elements are moved in turn from positions of lower power burnup to positions of higher power and burnup. Fresh elements are then inserted on the outside of the core into positions of low power. Normally four elements are exchanged each operating cycle. This fuel rotation plan has the advantage of maintaining essentially the same fuel element history for all operating cycles thereby preserving as nearly as possible the same flux shape and distribution for reactor users. Also it is a good method for limiting core reactivities, and for reducing power peaks and spikes as well as for achieving near optimum fuel burnup rates. Initially fuel burnup was less than 19% for light elements but continued to improve and reach about 40% by use of heavier elements. Last year approximately 3.2 Kg of U-235 were consumed representing an average burnup of almost 34%.

During the reactor checkout and testing program, most results were almost as predicted by calculations and analysis, and the entire program as well as reactor operations proceeded smoothly. Throughout, the reactor and its systems functioned almost flawlessly. Results of two tests merit special mention because of future possibilities of the methods used. One test was measurement of the power distribution in the core, and the other was fuel plate temperature measurements of an uncooled fuel element a short time after shutdown from power.

Initially, power distribution measurements were made by irradiation of cobalt and tungsten wires strung along fuel plates of a number of fresh fuel elements in representative positions in the core. The wires were activated at low power (about 400 watts), then counted on a wire scan. Normalized activities obtained from counting yielded the relative flux and hence power distribution in the clean core. Measured flux values when compared to calculations showed very good agreement even though the calculations tended to be a little more conservative. For the six elements in the innermost ring the area of highest flux calculated values were about 5% higher than measured. For 18 elements in the middle and outer rings calculated and measured values were almost identical. For the six corner elements (the lowest power elements) calculated values were about 10% lower than measured. Thus use of calculated power in thermal and safety analyses would be on the conservative side.

Power distribution measurements were repeated using a different technique - that of gross gamma counting of fuel elements. The relative power generation as a function of element position is determined by measuring the gross fission product activity for each element shortly after shutdown from full power using a gamma ion chamber in a fixed location. This method is particularly useful after operations when highly irradiated fuel is involved and flux wires cannot be inserted and because of the ease and quickness by which it is performed. The method proved very successful and the results were excellent. Agreement between the flux wire and gamma scanning techniques was very good with less than 2% differences observed for most elements. For this reason the gamma scanning technique is currently being routinely utilized to measure the power distribution in the core and to determine fuel burnup and coolant flow requirements.

One of the license requirements for the NBSR is that fuel plate temperatures may not exceed 800° F at any time during fuel element transfer including the normally used completely dry and uncooled transfer method. For this reason it was necessary to measure the maximum temperature reached by the fuel elements during the process of remotely transferring spent fuel elements from the reactor to storage facilities in the pool. Since the NBSR fuel transfer system design, which is confined and completely enclosed, did not permit the attachment of thermocouples to the fuel plate itself, infrared pyrometry techniques were devised to remotely measure the fuel plate temperatures. In addition to confirming that the NBSR fuel elements always remained at safe temperatures (maximum was 535° F), the development of this technique has provided a generally useful method for studying fuel element temperatures in a highly radioactive environment.

Over the year numerous improvements, additions and innovations were incorporated into the NBSR systems. Some of these are itemized below.

1. Design and installation of new irradiation facilities. Because of the increased demand on existing facilities, four new irradiation facilities were designed. One, a long term water cooled irradiation facility is already installed and operational. The remaining three are in the process of being fabricated for installation and testing in the near future.
2. The existing four pneumatic irradiation facilities were extensively modified to provide for a more efficient, more reliable and leaktight system. All of the jointed aluminum flight tubes were replaced by a single continuous newly developed armored polyethylene tubing. The senders-receivers and all control boxes were replaced by advanced types developed in-house that provide for simpler, smoother and trouble free operation.
3. Design and installation of cooling tower bypass valves that permitted remote and automatic secondary system water temperature control.
4. Design of an automatic acid injection system for secondary system water chemistry and quality control.
5. Installed bulk liquid CO_2 supply system that resulted in substantial cost savings as well as eliminated handling of individual CO_2 bottles.

6. The helium sweep system blowers were completely redesigned to improve their reliability and effectiveness and to provide for better and easier maintenance. The old system was susceptible to self-induced corrosion and to contamination by lubricant and was replaced by sealed stainless steel units. In addition specially designed cold traps were added to the system to remove and recover D_2O moisture. This permitted dry helium gas to be supplied to the fission product monitor and eliminated the problem of the monitor continuously saturating due to moisture. Also dry helium facilitated recombination of radiolytic gasses present in the sweep system.

C. Summary of Service and Service Programs

Extensive use was made of the reactor in-core irradiation facilities. More than 1,500 irradiations involving almost 10,000 samples were made during the year. These include a wide variety of research and application programs ranging from biological and medical studies to activation analysis programs, radiation effects studies and the production of numerous radioisotopes. More detailed reports of some of this work are presented below. Other programs not described in detail include radiation effect studies of numerous semi-conductor materials for the Naval Research Laboratory, irradiation of crystals for Catholic University, and air pollution studies for Rensselaer Polytechnic Institute.

Numerous other irradiation programs for outside organizations are presently being planned and evaluated which will contribute significantly to the increase of service programs over the coming years.

Trace Constituents of Geological Materials

J. J. Rowe and L. P. Greenland
(U.S. Geological Survey, Washington, D.C.)

The Radioactivation and Radiochemistry project of the Analytical Laboratories Branch, Geologic Division, U.S. Geological Survey has been using the NBS Reactor facility since July 1970. This project has been developing and applying activation analysis methods for the determination of trace constituents in geologic materials. These analyses are utilized in geochemical studies of magmatic crystallization-differentiation, post-magmatic alteration and the mechanism of ore deposition.

Platinum, palladium and gold are being determined at the low parts per billion concentration for Heavy Metals Program, to study background levels and magmatic fractionation. Platinum and palladium are preconcentrated by fire assay. After irradiation of the silver bead, platinum and palladium are separated radiochemically for beta counting (Rowe and Simon, Talanta, in press). Gold is determined on a one gram sample without preconcentration, using fire assay for the radiochemical separations (Rowe and Simon, USGS Circular 599). Greenland is studying the distribution of iridium in various magmatic provinces, using triple coincidence and radiochemical separations (Greenland, Rowe and Dinnin, USGS Annual Review, in press). Rowe is developing methods for the determination of osmium and ruthenium

Rowe is studying the transport of gold by thermal waters as a possible mechanism for the deposition of gold. He is developing a method wherein gold is collected from the water onto an ion exchange resin. The resin is subsequently irradiated for the determination of gold by activation analysis.

The determination of low concentrations of rare earths are valuable for studies of magmatic crystallization-differentiation. Both radiochemical and instrumental methods are being developed for the determination of rare earths (Rowe and Greenland).

Greenland is preparing for studies of the distribution of tantalum, hafnium, antimony and tungsten in lunar samples.

Simon is studying the distribution of tungsten and chromium in coexisting minerals of the California Batholith.

Other elements which are being determined by activation analysis are: Sc, La, Cr, Th, As, Hg and Se in various geologic materials.

Activation Analysis Program of the Food and Drug Administration at the NBSR

J. T. Tanner and M. H. Friedman
(Food and Drug Administration, Washington, D.C.)

1. Hydrofluoric Acid in Rust Stain Removers for Household Use.

The Food and Drug Administration is responsible for regulation of safety of household products. Among the problems is that of determining the concentration of hydrofluoric acid in rust stain removers. Activation analysis provided a most effective and rapid means for the purpose through measurement of the fluorine content.

2. Bromine Distribution in Test Pigs for Toxicological Studies.

Bromine provided a convenient index of the distribution of a bromine-containing material in the organs and tissues of pigs which were fed the material for evaluation of its toxicity. Because of the sensitivity and the large number of analyses required as well as the time involved in conventional methods, neutron activation analysis was especially well adapted for determination of bromine in the test samples (pig heart, liver, kidney, tissues, etc.).

3. Fluorine in Bone Meal.

Bone meal as a potential source of fluorine to replace or supplement fluoridated water in the diet obviously must contain a significant but not excessive fluorine concentration. Neutron activation analysis was used as a most convenient and rapid method for estimating fluorine in bone meal products.

4. Mercury in Foods.

Mercury has become of primary concern as an environmental contaminant. Because of its great sensitivity and reliability in conjunction with radiochemical separation techniques neutron activation analysis is particularly well suited for determination of trace concentrations of mercury in such a complex matrix as food. Of special significance is the elimination of the major problem of mercury impurities in reagents as interferences in trace level determinations. A variety of food categories has been examined in collaboration with Dr. David Lincoln of the Food and Drug Administration.

Neutron Activation Analysis in Scientific Crime Detection at IRS

M. J. Pro, C. M. Hoffman, K. B. Snow and W. D. Washington
(Internal Revenue Service, Washington, D.C.)

The Internal Revenue Service started to use the National Bureau of Standards reactor facilities July 1, 1970, and since that time the chemists have been involved in the irradiation of elemental standards and fluxmonitors for use in the quantitative analysis of trace elements found in physical evidence specimens. Also, a large number of irradiations have been carried out in connection with actual case work. This involved a large variety of sample matrices and some of the more frequently irradiated types of material are shown below:

1. Gunshot Residue - This determination involves the detection and measurement of barium and antimony deposited on the web of the hand of an individual who fired a gun. The residue is removed from the web of the hand with a cotton-tipped swab or with paraffin and the barium and antimony (from the primer) in the irradiated material are separated by simple radiochemistry and measured. This technique has been primarily used in suicide and murder cases.

2. Soil - Many soil samples have been irradiated to determine elements that are not amenable to analysis by chemical methods and atomic absorption analysis. Through the use of neutron activation analysis and other techniques, approximately 25 elements are quantitatively measured in soil and this information is used to compare soil from an unknown source with soils from a specific location.

3. Bomb Associated Residues - In connection with the Organized Crime Control Act of 1970 - Title 11, material such as tape, metals, paints, and etc. recovered from a bomb scene are compared by neutron activation analysis to similar material in a suspect's possession. The use of neutron activation analysis in this type of work is extremely helpful since many of these samples are very small in size.

4. Illicit Distilled Spirits - Moonshine seizures are routinely compared to products obtained from illicit distilleries by neutron activation analysis and other techniques. Again, neutron activation analysis offers the potential for measuring elements that cannot be measured by techniques such as atomic absorption analysis and spectrography. In addition, physical evidence such as soil, paints, metals, and etc. are also submitted in illicit distillery cases and the comparison of these materials very often is used in preparing evidence in conspiracy type cases. It is also significant to note that the successful prosecution of illicit distillery cases reduces the government's tax loss ($10.50 proof gallon) by helping cut down the number of illicit distilleries or by actual assessing of the violator.

In the area of research, the Internal Revenue Service chemists involved in the neutron activation analysis program are continuing their studies to determine trace elements in lead bullets. This type of examination is particularly useful when the comparison of bullets by microscopy fails due to the lack of sample or the mutilation of the questioned bullet. Other research activities involving the incorporation of high cross section elements into materials such as inks, pencil leads, and paper in connection with the Intelligence and Audit activities of the Internal Revenue Service are continuing.

Federal Bureau of Investigation Activation Analysis Program

G. Mahoney, J. Kilty, J. Riley
(Federal Brueau of Investigation, Washington, D.C.)

Variety characterizes the specimens received for analysis in the FBI Laboratory. The forensic laboratory often has little or no control over the size, shape or condition of the specimens presented for analysis. These factors must be considered along with the physical and chemical properties of the specimen when the methods of analysis are evaluated.

Generally neutron activation analysis does not place restrictions as far as size is concerned. It affords a very reliable quantitative analysis for many of the elements constituting the questioned specimen. The fact that many times the technique is non-consuming makes it the method of choice for an element determination in many cases where the integrity of the specimen must be preserved. Neutron activation analysis is the best method available for detecting and quantifying microgram quantities of antimony and barium on the hands of a person who is suspected of having fired a handgun.

U.S. Post Office Department Activation Analysis Program

A. W. Somerford and J. B. Upton

In recent years one of the most effective tools of forensic science used by the Chief Inspectors Department of the U. S. Postal Service has been neutron activation analysis. The ability of this nuclear method to detect traces of elements in minute samples enables it to solve many problems of identification that have heretofore been considered impracticable.

The Chief Inspectors Department has regularly used the NBS reactor in its crime investigation program since July 1, 1970. The analysis program deals with fine detailed examination of physical evidence, involved in some 2000 post office burglaries yearly, bomb parcel mailings, poison impregnated candies and cakes sent through the mail, and the tremendous increase in the mailing of narcotics and dangerous drugs, among others. Among the items analyzed, so far, are metal particles, glass fragments, refractories, hair, soil, wood, petroleum products, poisons, bomb fragments, and items used in bomb mailings.

Inspectors and scientists of the Chief Inspectors Department, U. S. Postal Service, have regularly testified in numerous cases on evidence discovered by neutron activation analysis. The method has proven to be extremely valuable in the solution and control of illegal activities and crimes and in providing substantial proof in court cases. The jurists of the Sixth U. S. Circuit Court of appeals, Northern District of Ohio, Eastern Division, decided October 29, 1970, the validity of Neutron Activation Analysis by affirming the lower court conviction.

Role of Trace Metals in Bone Healing

G. C. Battistone
(U.S. Army Institute of Dental Research
Walter Reed Hospital, Washington, D.C.)

The U.S. Army Institute of Dental Research is currently engaged in studies of means of accelerating wound healing. One aspect of these studies is the role of trace metals in bone healing. A successful outcome in this work could be of considerable value with respect to improved patient care and morale and significant savings in time and money. Thus far investigations have indicated that appropriately administered zinc can significantly accelerate bone healing in experimentally injured animals. Manganese has also been shown to have a small effect. Work has just begun during the past year on the role of copper in wound healing. The analytical portion of this study is being conducted by neutron activation analysis at the National Bureau of Standard's reactor using a technique developed at USAIDR. Copper chelates administered daily by i.p. injection to experimentally injured animals have demonstrated toxic effects which have necessitated changes in dosage levels and experiments are now in progress which indicate that toxicity has been eliminated. Preliminary analysis of copper values obtained for various tissues before and after the trauma of bone injury and during the healing process indicate a profound change in copper metabolism due apparently to a characteristic stress reaction. No conclusions have been reached as yet regarding the ability of copper chelate treatments to favorably alter copper metabolism during bone healing or to accelerate the healing process.

Study of Fission Product Gamma Ray Spectra

D. Lightbody and T. Dempsey
(Teledyne Isotopes, Westwood, N.J.)

The 10 megawatt research reactor facility at the National Bureau of Standards was used as a high flux source of fission spectrum neutrons. This program, sponsored by ARPA under Contract DA-01-70-C-0040, is mainly concerned with the study of gamma ray spectra resulting from the decay of fission products. These fission products result from the neutron-induced fission of enriched metallic targets of ^{235}U, ^{239}Pu, and ^{238}U. In addition to fission spectrum neutrons 14 MeV neutrons and thermal neutrons have also been used to induce fission.

Of primary concern to this program is detailing the relative differences observed in the gamma ray spectra for the above mentioned targets and neutron distributions. These differences result from variations in mass yield and independent yield distributions with both the excitation energy and mass of the fissioning system. Additional differences result from the variation with energy of the activation cross section relative to the fission cross section for the target system of interest.

Briefly, the experiment at NBS consisted of the simultaneous irradiation of ^{239}Pu, ^{235}U, and ^{238}U (10 mg, 10 mg, and 50 mg respectively) using the RT-2 pneumatic tube. Simultaneous counting of pairs of samples was performed from times as close to fission as possible (~4 minutes). Fission product gamma ray spectra were taken periodically for up to a month. The data acquisition system consisted of two high resolution Ge(Li) detectors which were multiplexed into an 8K multi-channel analyzer. A small on-line computer aided with preliminary analysis. Detailed analysis (gamma ray energies and intensities) was performed using a data reduction program written for the Isotopes IBM 360/30.

The data is still in the process of being analyzed. For the fission of ^{235}U and ^{239}Pu by fission spectrum neutrons, 19 mass chains representing 30 nuclides, have been identified in the fission product gamma ray spectra. From these type of data, one is able to extract information pertaining to changes in both the mass and charge distribution with excitation energy and initial neutron and proton number.

Direct Fission Population Measurements

P. Alexander
(Teledyne Isotopes, Westwood, N.J.)

Due to the short lifetimes possessed by fissioning nuclei it is often necessary to study the dynamics of the fission process as reflected in the properties of the fission products. These properties include the mass, charge, energy, angular momentum and directional distributions. Investigation of these quantities yields basic information concerning the fission mechanism of the highly deformed, compound nucleus.

Although some features of the fission process, such as the fission yield versus mass distribution are accurately detailed, there are areas where little detailed experimental information is available. The fission product charge distribution and isomer ratio values pertaining to the various mass chains are examples of areas where more experimental data is required. The isomer ratio is defined as the population of the isomeric state relative to the ground state for the same nucleus. Knowledge of both isomer ratio values and the shape of the charge distribution curves are important factors in developing a comprehensive fission theory.

The experiments being performed at the NBS Reactor high flux pneumatic tube facility involve measurement of the initial fission population values of the $11/2^-$ (isomeric) states relative to the $3/2^+$ ground states in ^{133}Xe and ^{135}Xe. These measurements were carried out using ^{235}U, ^{238}U, and ^{239}Pu as target nuclei. A large number of irradiations were made using separately encapsulated targets for bombardment with: thermal neutrons, or fast neutrons--using a cadmium jacket around the target. After each irradiation the xenon fission gas was removed as quickly as possible in order that the independently populated xenon states would not be too diluted by the ingrowth of xenon from the fission chain precursors.

After the initial extraction of xenon fission gas, the xenon ingrowth from fission chain precursors was successively milked. The gamma rays emitted during the decay of xenon in these extracted samples were then counted using a Ge(Li) detector. After correcting for relative efficiency and internal conversion effects the Ge detector data was analyzed using the computer programs ISIS-1 and ISIS-2 described in Nuclear Instruments and Methods 86 (1970) 99. Using these programs "best fit" values were obtained for the parameters Zp and ω which characterize the independent charge distributions in the mass chains A = 133 and 135. This work is supported by the Advanced Research Projects Agency under Contract DASA 01-69-C-0057 with the Defense Atomic Support Agency.

A Preliminary Investigation of the Mercury Levels in Chesapeake Bay Oysters

F. B. Birkner
(University of Md., College Park, Md.)

An investigation was undertaken to determine if mercury contamination of Chesapeake Bay oysters is a problem now or could become a problem in future years. The important public health questions which must be answered are: first, can oysters concentrate mercury in their body; and second, can the mercury levels in oysters reach a level which presents a distinct health hazard to man? Until recently, the analysis for trace concentrations of mercury in foods has been impossible, but with the advent of neutron activation analysis, concentrations as low as .02 ppm can be detected very easily.

Laboratory investigations were undertaken to determine if isotopic exchange separation when coupled with neutron activation analysis could be used for the determination of trace amounts of mercury in oyster tissue. The inorganic mercury recovery efficiency was found to be greater than ninety percent and calculations indicate that total mercury concentrations as low as one ppb can be measured in oyster tissue.

Oyster samples were collected from the Baltimore harbor area by the Natural Resources Institute of the University of Maryland. The oyster samples were freeze-dried, encapsulated and irradiated at the National Bureau of Standards. The oyster tissue was acid digested and mercury was recovered by isotopic exchange. An analysis of fifteen oysters revealed mercury concentrations ranging from 1 to 12 ppb (dry weight). Gold and silver can also be recovered in the isotopic exchange extraction, however, the recovery efficiencies for these metals is probably less than the ninety percent recovery efficiency which was found for mercury. Gold concentrations ranging from 1 to 5 ppm (dry weight) were also found in these oyster samples. These calculations are based on the assumption that the gold recovery efficiency is one hundred percent. Since the gold recovery efficiency is probably less than one hundred percent, the gold concentrations mentioned above represent conservative estimates of the gold levels in these oysters.

Although the mercury levels in these oysters are very low, it is not known at this time how mercury which is complexed with organic materials may be lost in the high temperature acid digestion step. Additional work is needed to determine the recovery efficiencies for methyl mercury and other organic mercury containing compounds which may represent a major portion of the total mercury in the oyster. Additional work must also be undertaken to determine the gold recovery efficiency in the mercury isotopic exchange extraction.

Trace Elements in the Environment

G. E. Gordon and W. H. Zoller
(University of Maryland, College Park, Md.)

Our major use of the reactor will be that of irradiating various types of environmental samples in order that we can determine the concentration of almost 20 to 30 (mostly trace) elements via instrumental neutron activation analysis employing Ge(Li) γ-ray detectors. The samples we are working on now consist of atmospheric particulate matter collected on Millipore filters during a systematic sampling program conducted in the Boston, Massachusetts area. In addition we have a number of samples taken at the same time with cascade impactors which separate particulate matter into various size fractions. When Professor Zoller returns from the South Pole he expects to have a number of atmospheric samples from that region which we wish to analyze in order to determine world-wide atmospheric "background" levels for many elements. Related to the studies of urban air pollution, we have many samples of pollution source materials (particularly, crude oils and distillates of them) to analyze. As a side project, we are also looking at tree-ring sections from some very old trees, seeking correlations with geochemical history of the areas in which the trees grew and looking for any build-up of industrially important elements since the start of the Industrial Revolution. An essential feature of all of this work is the necessity for us to develop monitors that will contain accurately known amounts of each element to be measured in the samples. Furthermore, the relative amounts of the various elements should be similar to those of the samples under investigation. This is a very difficult problem facing all activation analysis. In cooperation with Dr. Harry Rook of the Activation Analysis Section of NBS, we are trying several methods of attack on the problem.

We will occasionally be irradiating small amounts of material (often enriched in particular isotopes) to make radioactive species for investigation of their decay schemes in order to obtain fundamental information about the structure of nuclear levels.

Neutron Damage Studies on Lead Salts

R. M. Cullpepper
(Naval Ordnance Laboratory, White Oak, Md.)

The National Bureau of Standards and the Naval Ordnance Laboratory have jointly undertaken the study of neutron induced damage in lead sulfide expitaxial films. The films are grown and measured at NOL and are irradiated in the RT-2 facility of the NBS reactor. The samples are being irradiated in steps of approximately 10^{17} neutrons/cm^2 and the changes in the number of free carriers and sample resistivity are measured as a function of temperature after each irradiation.

The data taken to date indicates that approximately one free carrier is removed from the sample for each neutron in the flux. This reduction in the number of free carriers is true for both n and p type samples. More irradiations are planned, and a total exposure of about 5×10^{17} neutrons/cm^2 is anticipated.

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