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APPLICATIONS OF A PULSED SPALLATION NEUTRON SOURCE

Report of a Workshop held at
Argonne National Laboratory
April 29-May 4, 1973

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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS
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Report of a Workshop
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April 29 - May 4, 1973

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EDITORS' PREFACE

The main body of this document consists of the reports of the six working groups into which the participants were assembled. The final form of these reports is based on draft versions compiled during the course of the workshop and subjected to a limited amount of addition and revision in the subsequent three weeks. The members of each working group with their affiliation are listed at the head of each section. It should be understood that, although all participants are listed as coauthors of the complete report and have been given an opportunity to comment on all sections, they should be considered responsible only for the opinions and conclusions stated in the section prepared by their working group.

We should like to acknowledge the help of G. A. Bartholomew, W. C. Koehler, W. L. Whittemore and D. J. Zaffarano at a meeting held at Argonne on December 13 - 14, 1972, to plan the workshop.

J. M. Carpenter
D. L. Price

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ABSTRACT

This report summarizes the conclusions reached by the participants of a workshop held to discuss the applications of a pulsed spallation neutron source. It contains reports from the six working groups devoted to (1) high energy transfer inelastic scattering, (2) low energy transfer inelastic scattering, (3) diffraction from single crystals, (4) diffraction from isotropic samples, (5) radiation effects and (6) prospects and problems. External reports on pulsed neutron radiography and on a neutron-neutron scattering experiment are included.

WORKSHOP SCHEDULE

<u>Sunday, April 29</u>	8:00 - 10:00 p.m.	Informal get-together
<u>Monday, April 30</u>	9:00 - 11:45 a.m.	Plenary Session
9:00-9:10	Dr. O. C. Simpson, Director Solid State Science Division Argonne National Laboratory	Welcoming address
9:15-9:45	Dr. M. V. Nevitt, Deputy Director for Research Argonne National Laboratory	Role of a pulsed spallation neutron source and motiva- tion for the work- shop
9:50-10:30	Dr. R. L. Martin, Director Accelerator Division Argonne National Laboratory	The ZGS Injector- Booster and com- parable accelera- tors elsewhere*
11:00-11:45	Professor J. M. Carpenter Nuclear Engineering Dept. University of Michigan and Solid State Science Division Argonne National Laboratory	ZING: A pulsed spallation neutron source based on the ZGS Injector- Booster*
	1:30-4:20 p.m.	Plenary Session
1:30-2:15	Professor P. A. Egelstaff Department of Physics University of Guelph Ontario, Canada	Review of applica- tions of pulsed and steady-state neutron sources
2:20-3:00	Dr. D. L. Price Solid State Science Division Argonne National Laboratory	Initial survey of applications of a pulsed spallation source
3:20-4:00	Professor M. Kimura Tohoku University Sendai, Japan	Survey of intense pulsed neutron sources
4:05-4:20	Professor J. M. Carpenter	Working arrange- ments for remain- der of the work- shop
6:00-7:00	Social hour	

WORKSHOP SCHEDULE (Cont.)

<u>Tuesday, May</u>	9:00 a.m. - 12 noon	Working group meetings
	1:00-5:00 p.m.	Plenary session for brief reports from working groups
	7:30-9:30 p.m.	Working group meetings
<u>Wednesday, May 2</u>	9:00 a.m. - 12 noon	
	1:00 p.m. - 5:00 p.m.	Working group meetings
<u>Thursday, May 3</u>	9:00 a.m. - 12 noon	Working groups to draft reports summarizing their ideas, findings, and conclusions
	1:00-5:00 p.m.	Plenary sessions to review draft reports
	7:00-9:00 p.m.	Dinner for workshop participants and invited guests at the Argonne Staff Club
<u>Friday, May 4</u>	9:00 a.m. - 12 noon	Working group meetings (optional)

*Current plans for the ZING facility were discussed in these two talks and will be described in a report to be published separately: J. M. Carpenter, "A Description of ZING--An Intense Spallation Neutron Generator Based on the ZGS Injector-Booster Accelerator".

REPORTS OF THE WORKING GROUPS

1. High Energy Transfer Inelastic ScatteringParticipants

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I. Introduction

For the purposes of this study, we have chosen to concentrate on those problems which require high incident energy ($E_0 \gtrsim 250$ meV), although energy transfers in many experiments of interest will be below 100 meV. This field of research is one that has not received a large amount of attention in past years, principally because of the relatively low fluxes of such "epithermal" neutrons available in present day reactors. The major efforts in this field in the USA have been limited to three neutron facilities--the Rensselaer Polytechnic Institute linac facility, the Battelle Northwest Laboratory reactor and the HFIR reactor at Oak Ridge National Laboratory. The first two of these facilities are no longer operating, and the effort at the HFIR has not been extensive, primarily because the facilities used have been designed for use at lower energies. Thus, in our deliberations, we have had to adopt the philosophy that we would look at those experiments or classes of experiments which we would do tomorrow if a spallation device were available. We have attempted to estimate the future of these fields as work in these areas progresses (the hot source at the new Grenoble reactor can be expected to produce many results in the years ahead) but in any event, we are confident that the field will grow in importance as facilities become available.

An important characteristic of spallation devices is the fact that for all moderator designs, the neutrons are under-moderated leading to a relatively high flux of epithermal neutrons ($E > 0.1$ eV). Normal research reactors are in general well moderated, so that hot sources are necessary to obtain usable neutron fluxes in the epithermal regime. Thus, spallation sources offer unique advantages for this type of work. Although it is difficult to give good numerical estimates of the advantages of pulsed sources in reducing backgrounds, a first approximation would be that the background varies as the time-averaged flux while the signal varies as

the peak flux. For several of the experiments considered in this report, low backgrounds will be essential. In addition, the pulsed feature of the source can lead to significant simplification of equipment design because the first chopper can be eliminated from a phased chopper system. Since the whole source area (possibly 10 cm by 10 cm) is pulsed, large solid angles can be used. It should be noted that crystal reflectivities become quite low at high energies, where reflectivity varies approximately as $1/E$ with peak reflectivities of order 5% at 500 meV. For the design of high resolution spectrometers to work at high energies, much more information on crystal reflectivities is required.

For the purpose of having a concrete example, we have used the figures given by Carpenter and Marmer¹ for the proposed ZING facility to estimate the achievable count rates for various facilities. Although we have attempted to be specific in this regard, considerable uncertainties in moderator performance render the estimates tenuous at best. An example of the intensities obtainable for a typical case is given later.

In the body of this report we have chosen to present four classes of experiment in some detail. We feel that these are representative classes which are our best present estimate of the most significant fields in this area of neutron beam research. We have necessarily left out several promising areas of experiment which cannot now be assessed. We fully expect that such experiments would be attempted if facilities were available and that several of these might turn out to be exciting and fruitful (perhaps more so than the ones we have presented in detail). Included among these are crystal field splittings in actinides and opaque garnets, high energy single particle excitations of various kinds, measurements of complete scattering functions at large energy and momentum transfers for liquids and glasses and many others.

In concluding this introduction it is worth noting that the proposed ZING facility would be more powerful in this range of neutron energies than any existing reactor in the

USA. In fact, the peak flux would be comparable to that at the hot source in Grenoble in the 0.2 - 0.5 eV region, and the pulsed feature of the ZING source offers significant design advantages for some types of experiments. We feel that it is important that the USA develop a capacity for this important field of neutron beam research in the near future and that pulsed accelerator-driven spallation sources are the most promising candidates for obtaining such a capability.

A Specific Example of a Possible Instrument at ZING

From ANL/SSS-72-1,¹ the time averaged beam intensity for 500 meV neutrons would be 10^{13} n/(ster-ev-sec) from a 10 cm x 10 cm source block. According to Carpenter, the pulse width for such neutrons might be reduced to 6 microseconds with no loss in average intensity by using a D₂O reflector. In what follows, we shall assume that this performance can in fact be achieved, and the principal weakness of the estimates following will be this assumption.

- (a) Incident energy $E_0 = 500$ meV
- (b) Full burst width from source will be used
- (c) Chopper burst time will be 3 microseconds
- (d) Source to chopper distance will be 15 m

With these parameters, and noting that 500 meV neutrons travel 1 cm/microsecond, the incident energy resolution can be estimated as follows.

$$\frac{\Delta E_0}{E_0} = 2 \frac{\Delta t}{t} = \frac{2 \sqrt{6^2 + 3^2}}{1500 \times 1} = 8.94 \times 10^{-3}$$

or $\Delta E_0 = 4.5$ meV.

The "monochromatic" beam current at a distance L from the 10 cm x 10 cm source can be estimated from the expression

$$J = \frac{I}{L^2} \Delta E_0 T ,$$

where $I = 10^{13}$ n/(ster-ev-sec). ΔE_0 = the energy spread about E_0 in eV and T = chopper transmission.

From preliminary considerations, it appears possible to construct a chopper for 500 meV neutrons with $T = 0.5$ for this facility. Thus

$$J = 1. \times 10^{13} \times 0.0045 \times 0.5 / (1500)^2$$

$$\underline{J = 10^4 \text{ n/cm}^2/\text{sec.}}$$

This is the time averaged current. The peak current will be (using ANL/SSS-72-1)

$$\underline{J = 3 \times 10^7 \text{ n/cm}^2/\text{sec}}$$

The beam area could be as large as 5 cm x 10 cm.

Reference

¹J. M. Carpenter and G. J. Marmer, "Evaluation of the ZGS Injector-Booster as an Intense Neutron Generator," Argonne National Laboratory Report ANL/SSS-72-1, April 19, 1972.

II. High Momentum Transfer Measurements in Liquids

When neutrons of sufficiently high momentum transfer Q are scattered from materials, the scattering may be thought of as taking place from individual atoms and collective effects play no role in the scattering.

Thus, in liquids, where little may be known about the ground state properties, direct information about the ground state can be obtained by the high Q technique. This can be thought of as scattering from the liquid in a period short compared to interparticle collision times, so that a single atom may be examined at a time. The properties of the liquid are reflected in that the interactions between particles provide this atom with a momentum peculiar to the ground state.

In this case the conservation of energy and momentum in the neutron liquid atom collision gives the relation between the energy loss $\hbar\omega$ and momentum transfer $\hbar\vec{Q}$

$$\hbar\omega = \frac{\hbar^2 Q^2}{2m} + \hbar \frac{\vec{Q} \cdot \vec{p}}{m}$$

where m is the mass of the atom and $\vec{p}$ is the momentum before collision. Since there will be no scattering unless this condition is satisfied the scattering law $S(Q, \omega)$ will be given by a δ function with the argument

$$\hbar\omega - \hbar^2 Q^2 / 2m - \hbar \vec{Q} \cdot \vec{p} / m$$

multiplied by n_p , the number of atoms with momentum $\vec{p}$ and summed over p .

$$S(Q, \omega) = \sum_{\vec{p}} n_p \delta(\omega - \frac{Q^2}{2m} - \vec{Q} \cdot \vec{p}/m)$$

letting $\hbar = 1$.

If we replace this sum by an integral and integrate over

the angle between $\vec{Q}$ and $\vec{p}$ we obtain

$$S(Q, \omega) = \frac{m}{4\pi^2 Q \rho} \int_{p_{\min}}^{\infty} p n_p dp$$

where ρ is the density of the liquid and where $p_{\min}$ is the smallest momentum which will give the energy loss ω

$$p_{\min} = \frac{|\omega - \omega_0|}{Q/m}$$

Thus the slope of $S(Q, \omega)$ as a function of ω measures the product of $p n_p$ evaluated at $p_{\min}$. I.e., n_p can be directly determined by a neutron scattering investigation since the neutron cross section per unit energy per unit solid angle is given by

$$\frac{d\sigma}{d\Omega dE} = \frac{k_i}{k_o} S(Q, \omega)$$

where k_i and k_o are the incident and outgoing wave vectors respectively.

The possibility of determining n_p is particularly exciting in the case of liquid He since central to the theory of superfluid helium is the concept of a condensate of particles having zero momentum. The idea of Bose-Einstein condensation in superfluid helium was first suggested by London¹ in 1939 in order to explain the unusual properties of the superfluid. Despite the fact that this suggestion led to the proposal of the two fluid model for superfluid helium which has been successful in explaining many of the microscopic properties of the superfluid, it has been extremely difficult to provide direct evidence that Bose-Einstein condensation does in fact occur. Recent neutron experiments by O. K. Harling² at Battelle Northwest and H. A. Mook, R. Scherm, and

M. K. Wilkinson³ at Oak Ridge have provided evidence that this condensation does in fact occur, although the condensate fraction n_p appears to be much smaller than has been theoretically suggested and thus is difficult to observe. Harling's analysis of his experiment suggested a condensate fraction of about 8%. The Oak Ridge experiment, which was done with the better resolution of the two, suggested a condensate fraction of only 2% compared to theoretical estimates in the 8 to 20% range. This experiment required about five months counting time at the HFIR reactor on a triple axis spectrometer, and represents the best result possible with conventional reactors with standard equipment.

It is of importance that this experiment be pushed to higher momentum transfers, where it is expected that the condensate can be more clearly distinguished from the momentum distribution of the normal helium atoms, and to higher resolutions. It is also important to determine the temperature dependence of n_p so that a direct connection may be established between the Bose condensation and the superfluid properties of helium. Using incident flux numbers obtained from ANL/SSS-1-72 an experiment was designed that could be performed by the ZING facility. The experiment would be performed at an incident energy of 500 meV and an energy resolution of around 4 millivolts. This is of comparable resolution to the Oak Ridge experiment but at a momentum transfer of $25 \text{ \AA}^{-1}$ compared to $14.3 \text{ \AA}^{-1}$ for the Oak Ridge experiment. It is hoped that the condensate could be seen more clearly at this higher momentum transfer, and that a temperature dependent study could be made since the counting time required is only about ten days per temperature to obtain 1% statistics in the peak of the distribution where the condensate should be visible. An improvement in the chopper design so that it could be adapted to this particular experiment and a special detector bank could bring the measurement time down to approximately two days. An experiment with better resolution would probably be desired and to halve the resolution to 2

millivolts would take on the order of 16 times as long, thus taking around four or five months with standard detectors. A large heavily shielded ring of detectors would probably bring the counting time down to a month or so making the experiment quite feasible.

Thus a series of relatively lower resolution experiments could be performed at various temperatures and pressures giving coarse values of n_p under various conditions and some high quality measurements made at important temperatures such as just above the λ point and at very low temperatures to obtain the zero temperature ground state. In these experiments the number of atoms in the condensate (those with zero momentum) would be determined and the ground state momentum distribution of the normal component of the helium would be obtained. The ground state momentum distribution of the normal component could be compared with theoretical distributions calculated by various techniques.

Measurements of n_p would also be of interest in other liquids and amorphous materials although of course no condensate could exist. Argon and neon are suggested candidates for such measurements in the momentum range of 20 - 80 $\text{\AA}^{-1}$. Sears⁴ and Gibbs and Harling⁵ have indicated how high Q experiments of this type can be used to provide useful information about n_p and the interatomic potential of condensed systems in general. The free particle scattering limit would be more difficult to reach for these materials and quite high energies might be needed. It is expected that ZING will be a useful, perhaps unique device for these measurements since momentum transfers of 80 $\text{\AA}^{-1}$ or so may be desired and it is doubtful that such experiments are possible at existing facilities.

References

- ¹F. London, Nature 141, 643 (1938).
- ²O. K. Harling, Phys. Rev. Letters 24, 1046 (1970).
- ³H. A. Mook, R. Scherm, and M. K. Wilkinson, Phys. Rev. A6, 2268 (1972).
- ⁴V. F. Sears, Phys. Rev. A5, 452 (1972); Phys. Rev. A7, 340 (1973).
- ⁵A. G. Gibbs and O. K. Harling, Phys. Rev. A7, 1748 (1973).

III. Molecular Spectroscopy

Neutron inelastic scattering techniques have been widely applied to the study of vibrations and rotations in hydrogenous molecular systems. A significant amount of effort has been directed toward the study of intramolecular vibrations, most notably large-amplitude modes such as molecular torsional vibrations and hydrogen bond modes. Due to the large incoherent hydrogen scattering cross section, the neutron spectra measured in these studies provide a direct (but not always interpretable) reflection of the proton motions in these systems, including all intramolecular vibrations which involve a nonzero hydrogen displacement. The neutron results have provided important spectroscopic information complementary to that obtained by infrared and Raman techniques, particularly for cases of inactive or weakly active modes in hydrogen bonded systems, organic compounds, polymers, etc. Further, in the last few years efforts have been made (e.g., at Harwell,

Risø and MIT) to calculate detailed neutron spectra on the basis of assumed molecular potential functions for comparison with experimental results on various molecular systems. If successful, these theoretical attempts could make neutron spectroscopy a more reliable method for the assignment of normal modes on the basis of the position and intensity of the observed inelastic scattering peaks and would open the possibility of the further evaluation and refinement of molecular potential functions.

The measurement of relatively high energy vibrational levels (say > 100 meV) by neutron spectroscopy is necessarily carried out by neutron energy loss techniques, in turn requiring high incident neutron energies. Such measurements are limited at conventional steady state reactor sources due to low neutron intensities above about 150 meV. For energy transfers below this value, one is usually forced to accept very low energy scattered neutrons, as exemplified by the Be filter detector spectrometer. We note, however, that this technique may give undesirably high momentum transfers as discussed below. The situation can be improved by using hot neutron sources, giving the possibility of incident energies up to around 0.5 eV as at the high flux reactor at Grenoble. However, decreasing monochromator efficiencies can be a problem at higher energies, implying a preferability for time-of-flight techniques for energy selection.

The proposed ZING facility, being a pulsed source, inherently provides an efficient means of utilizing time-of-flight techniques. In addition there is the considerable advantage for the experiments discussed here of a relatively high epithermal flux. By going to higher incident neutron energies, one extends the available range of energy transfers that can be investigated, and obtains greater freedom in choosing momentum transfers appropriate to the experiment in hand.

There are a number of very interesting molecular experiments which could be done with the copious neutron fluxes in the range 200-500 meV which would be available at a facility such as ZING. Such a source would open up the field of neutron molecular spectroscopy in the energy-transfer range 150 to 400 meV which has been virtually untouched in the past, including the possibility of measuring the dispersion curves of high energy vibrations (100-300 meV) in certain strongly hydrogen-bonded or polymeric crystals. In addition the availability of high energy neutrons with very high resolution would provide the opportunity to pursue a vigorous program of small "momentum transfer" ($Q \sim 1$) neutron scattering investigations of lower energy (< 100 meV) molecular vibrations in molecular fluids. A word of caution here--we are not suggesting that such neutron spectroscopy is now or will become an analytical tool in the same sense as IR or Raman spectroscopy. This is precluded by inherent limitations in available resolution and intensity as well as by the unique ability of neutron incoherent scattering to "see everything" (which sometimes means seeing too much). We can state, however, that neutron scattering can, by the investigation of selected molecular systems with appropriate structures, make a major contribution to the understanding of molecular structure and intramolecular forces and that a facility such as ZING will significantly extend the available spectral range and resolution for such investigations.

With this in mind we would like to give a few examples of appropriate experiments which could be done with the epithermal neutron beams from a ZING-type source. First of all it seems clear that there are a number of possible, highly valuable experiments involving a detailed study of the neutron energy loss spectra for strongly hydrogen-bonded systems in the range of energy transfer from 100-400 meV--under as high resolution as practically possible.

One of the most important available techniques for detecting hydrogen bonds and assessing their strength, symmetry

and other details of the hydrogen-bond potential is the study of hydrogen stretching and bending vibrations by infrared and Raman spectroscopy. In many cases, however, these modes cannot be observed or assigned with certainty due to selection rules or to overlap with other internal vibration bonds or overtones. In addition these peaks are often extremely broad (widths $\sim 5 - 40$ meV) or exhibit splittings. Such effects make the quantitative assignment of vibrational frequencies difficult or impossible (accurate values for these frequencies are critical for studies of hydrogen bond forces and potentials). The observation of these widths and splittings has been the subject of intense experimental and theoretical investigation--both to explain their existence in the optical spectra and to relate them to the detailed nature of the hydrogen bond. Obviously this situation provides an ideal and highly important application of high energy transfer neutron scattering. The neutron spectral peaks for the large amplitude hydrogen modes in such systems (including the bending and stretching modes) should dominate the observed spectra, thus making assignments for these modes quite easy. The simpler and entirely different neutron-scattering interaction with the molecular system should give an independent measure of the frequency and natural widths of the hydrogen vibration peaks--thus providing valuable new insights into the origin of the observed IR bandwidths (e.g., dispersion, anharmonicity, multiphonon events, Fermi-resonance).

A specific example of important chemical systems for which neutron measurements would be of great interest is simple inorganic crystals exhibiting symmetric or nearly symmetric hydrogen bonds. These would include, e.g., NaHF_2 and HCrO_2 , two solids having the same simple trigonal, crystal structure ($Z = 1$) and effectively symmetric hydrogen bonds. One of these solids is highly ionic (NaHF_2), while the other (HCrO_2) has a great deal of covalent character. The observed IR stretching vibration peaks are very broad (~ 40 meV) with "maxima" at ~ 200 meV. Overtone bands are

essentially impossible to interpret in the optical spectra. The deuteration H(D)CrO_2 exhibits the largest isotope effect ever observed and the hydrogen bonds apparently become symmetric. It is clear that neutron scattering investigation of the hydrogen bond modes between 150 - 400 meV would be of fundamental importance in the understanding of these very interesting compounds.

We can briefly mention other fields open to high incident energy inelastic neutron scattering. For example the usefulness of neutron measurements for studying lower energy vibrational transitions ($\sim 20 - 60$ meV) of molecules in fluid phases has been established using the so-called "small Q method." Typical examples include the characterization of torsional modes in molecules containing methyl groups, where optical techniques are sometimes limited by constraints imposed by selection rules. It is essential that neutron measurements be carried out at low momentum transfer in order to minimize diffusional broadening of the observed levels. This capability would be enhanced in many cases by the availability of intense neutron beams in the 100 - 200 meV range and very small scattering angles. High resolution capability, say $\Delta E \sim 1$ meV, is required to ensure the resolution of low-lying modes.

A third area of application of an intense epithermal neutron source would be the measurement by coherent scattering of dispersion curves for high energy vibrational modes in selected molecular or inorganic solids. These would include, e.g., the stretching and bending vibrations in the strong H-bonded systems discussed above, and the intrachain bending modes in polymers. Theoretical dispersion curves have been calculated for polyethylene which predict a large dispersion in the chain direction for several high energy "intramolecular modes" ($E \sim 100 - 150$ meV). Thus a careful measurement of this dispersion could provide valuable information (in addition to dispersion curves measured for lower frequency modes) toward a detailed understanding of intramolecular forces in

polymers. Similarly, measurements of dispersion curves and phonon line shapes for high frequency modes in simple hydrogen-bonded crystals could provide insight into intramolecular effects on hydrogen bond forces and on the extreme anharmonicity apparent in some hydrogen bonded crystals. It should be stressed here that such measurements would obviously be limited by the availability of deuterated single crystals of appropriate size of these materials.

Finally it would seem useful to give at least an indication of the time which might be required to perform the types of scattering experiments outlined above. We have made some gross estimates based on the preliminary figures for epithermal fluxes available at ZING and with a knowledge of the approximate times required for such experiments in the past at lower intensity sources (e.g., Harling, et al., at Battelle). It is our conclusion that reasonable molecular spectra for hydrogenous systems with no limitation in sample size could be measured in the order of a day under very high resolution ($E_0 = 500$ meV, $\Delta E = 2$ meV) and fairly low Q ($\sim 2-4$ Å). More coarse resolution spectra ($\Delta E \approx 4$ meV) without stringent Q conditions could certainly be done in a few hours.

IV. Hydrogen Modes in Metal Hydrides

The study of the physical properties of impure solids is an important and expanding part of solid state physics. Due to the small size of the hydrogen atom the presence of an interstitial hydrogen in a metal involves a relatively small distortion of the metal lattice, and the metal-hydrogen system is therefore attractive from a theoretical point of view. It is also easy to obtain a wide range of hydrogen concentrations at temperatures and pressures which are convenient from an experimental point of view. For these reasons, the interstitial solution of hydrogen in metals offers a suitable class of prototype systems for the study of the behavior of defects in the solid state. Due to the metallic character of the

metal-hydrogen system the methods of optical spectroscopy are not applicable in this case. The neutron scattering properties of the hydrogen atom, on the other hand, are such that the hydrogen motion is most conveniently studied by the inelastic neutron scattering method. Information about the time-dependent behavior of one interstitial atom is obtained in a very direct way from the incoherent scattering results while the collective aspects of the motion may be derived from the coherent scattering, which is obtained when deuterium is substituted for hydrogen. To use efficiently the information contained in the coherent scattering results, single crystals should be used and, as single crystals are not readily available in this case, the discussion below will be concerned mostly with the study of hydrogenous samples in the polycrystalline form. A significant advantage of the neutron scattering method is that, due to the large value of the incoherent scattering cross-section, it is possible to study dilute systems.

The local vibration of the hydrogen atom in the metal lattice gives rise to discrete peaks in the neutron spectrum well outside the range in which the acoustic phonons of the host lattice are observed. The separation between vibrational levels is of the order of 0.1 eV and therefore only the lowest level is significantly populated at reasonable temperatures. For reasons of intensity it is therefore necessary that the neutron measurements be made in the energy loss mode and, as it is in many cases important that multiple excitations be observed, these measurements therefore require that neutrons in the epithermal range be used. It is also important that the spectrum of scattered neutrons be analyzed over a wide range of energies and, for reasons of neutron economy, the time-of-flight technique is in this case the most suitable experimental method. In fact, neutron scattering results which have been obtained so far and which have measured multiple excitation of levels are without exception obtained by time-of-flight methods. It is thus clear that these

studies are well suited to a pulsed source of the ZING type with an intense epithermal flux.

A number of metal-hydride systems have been studied to date. These include systems in which the hydrogen is rather weakly bound and for which only one discrete level is observed (e.g., UH_x and NbH_x) as well as systems with strong binding of the hydrogen and in which a number of bound levels are excited (e.g., ZrH_x and TiH_x). Although these systems a priori are of comparable physical interest we will discuss in detail only the study of the latter type as these pose experimental requirements which in comparison with existing neutron sources are particularly well met by a pulsed spallation source. In an attempt to explain the shape of the frequency distribution of the first excited level in ZrH_x Slaggie¹ has proposed a model in which the shape of the peak is obtained from a lattice dynamical computation assuming central force interactions between Zr-Zr, Zr-H, H-H (second nearest neighbors) and H-H (third nearest neighbors). In order to estimate the accuracy which is needed in the neutron experiment we may assume that the model results of Slaggie are typical of this kind of computation and we should then require that the neutron experiment resolves the fine structure obtained by Slaggie. In order to specify the problem we may characterize the first excitation level by:

1. Mean energy = 0.14 eV
2. Total width of level = 0.025 eV
3. Width of significant structure = 0.002 eV

We will further require that 10^4 counts are recorded per resolution element in the maximum of the peak and that the transmission of the sample is 90%. The latter requirement is important in order to keep the contribution from multiple scattering events within reasonable limits. It is also important that the momentum transfer be kept small in order to limit the multiphonon scattering. We assume that the incident

energy E_1 is 0.350 eV and that the scattering angle is 15° . This corresponds to $Q = 3.94 \text{ \AA}^{-1}$ which is somewhat smaller than the smallest Q -value used in the study of ZrH_x by Couch et al.² The required energy resolution is 0.002 eV. If we assume that 50 counters of dimension 1" by 20" are used we can calculate the intensity of the scattering into one time channel whose width is chosen to match the energy resolution:

$$\text{Int} = 900 \text{ counts/channel/hour.}$$

To measure 10^4 counts/channel in the peak we would therefore need to run for approximately 11 hours. We thus find that the experiment which we have chosen to study as a reference case will allow us to obtain data at a rate which is much higher than has been possible in previous experiments of this kind, with an energy resolution which is better than in previous experiments and which in fact is good enough to resolve details of the lattice dynamics. Considering the importance of these kinds of studies and also the difficulty in obtaining experimental results of sufficient accuracy at existing neutron sources, the results which we have obtained above are most encouraging.

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V. High Energy Magnetic Inelastic Scattering

The direct observation of the continuum band of Stoner excitations in the ferromagnetic phase of transition metals, such as nickel and iron, by high energy inelastic neutron scattering, would represent a major breakthrough in our scientific and technical understanding of the origin of ferromagnetism in these technologically important elements. As detailed below, such observations require a flux of 0.5 to 1 eV neutrons which is unavailable anywhere in the United States or in the world. Performance estimates indicate the feasibility of such experiments with a ZING-type facility. The positive observation of such excitations is critical to the initiation of programs of research which are the analogs to those of electromagnetic interband transitions.

Current theories of ferromagnetism in the transition metals divide the magnetic-type excitations into two classes: the first class is the collective excitations, in which almost all magnetic carriers contribute to the excitations; the second class is the Stoner excitations naively viewed as those arising from the magnetic excitation of a single electron. In the first class, the collective excitations consist of those which begin at zero energy for zero wave vector and are called magnons and of those which begun at finite nonzero energy and are called optical magnons or interband spin waves. The single particle continuum excitations cover a broad range of energies, starting at zero or almost zero (100 meV) energy and ending at 1 eV or higher energy. The excitations are generally grouped together according to their wave-vector, and the general energy, wave-vector position for such states are indicated schematically in Fig. 1.1 for nickel.¹ A clear distinction between single particle continuum and collective states is evident in this figure. For a given wave vector, the single particle excitations span a broad range of energies whereas the collective excitations have a well-defined energy.

Of all the excitations shown in this figure, only the

solid heavy line of the magnon has been experimentally confirmed.² Verification of the remainder of this figure requires high energy neutrons. Theoretical considerations show that the single particle continuum scatters neutrons appreciably but that the inelastic scattering is spread over a broad energy range.^{3,4,5,6} This scattering does show very broad peaks in the lower part of the single particle continuum for wave-vectors in the neighborhood of $0.5 \text{ \AA}^{-1}$. Observation of this very broad peak appears as the most favorable experiment to verify the existence of such excitations.

As a specific example, feasibility estimates are presented for the direct observation of Stoner excitations in nickel. This experiment is chosen as much experimental and theoretical work^{1,2,4,5,6} has been conducted on nickel and Ni^{60} is a convenient isotope which yields small nuclear scattering background. Our estimates therefore have a high degree of reliability.

The neutron energy loss range of 0.08 to say 0.3 eV is the estimated range in which single particle modes in nickel show a broad peak in the neutron scattering. The Q range over

Fig. 1.1 Schematic illustration of magnetic excitations in nickel.

which this type of experiment is possible is determined by the form factor. For example, in nickel near the 200 reciprocal lattice point ($Q \sim 5 \text{ \AA}^{-1}$) the square of the form factor has already fallen to one half. At the same time the multi-phonon scattering against which the magnetic scattering must compete will be going up in intensity at least as fast as Q^2 . Thus the range of Q available in practice is limited to a rather narrow range near $4 \text{ \AA}^{-1}$. (As we shall show later the difficulties of the experiment increase rapidly if Q is smaller than is strictly necessary.)

Incident Energy Required

Energy conservation determines the minimum incident energy at which the experiment could be done for a given Q value of interest ($4 \text{ \AA}^{-1}$) and a range of energy loss $0.08 - 0.3 \text{ eV}$.

It is easily seen that the minimum incident energy will occur for scattering at very small angles where approximately

$$Q = k_0 - k_1 \quad .$$

At the same time the energy loss $\hbar\omega$ is given by

$$\hbar\omega = \frac{\hbar^2}{2m} (k_0^2 - k_1^2) \quad .$$

With a little manipulation of these two equations it is easily shown that the minimum incident energy E_0 is given in terms of our fixed wave vector Q and variable energy transfer $\hbar\omega$ by

$$E_0 = \frac{(\hbar\omega + \frac{\hbar^2 Q^2}{2m})^2}{4(\frac{\hbar^2 Q^2}{2m})}$$

For $Q \sim 4 \text{ \AA}^{-1}$, the following incident energies are required:

Energy Loss (eV)	Minimum Incident Energy (eV)
0.060	0.069
0.080	0.098
0.100	0.132
0.120	0.180
0.2	0.42
0.3	0.86

Thus in order to measure energy transfers in the desired range of 0.08 - 0.3 eV, extremely high incident energies approaching the 1 eV region are required.

Resolution Requirements in $\hbar\omega$ and Q

The intensity of an inelastic scattering experiment depends strongly on the resolution requirements since the resolution in q_x q_y q_z and $\hbar\omega$ must all be considered. In the important region where the spin wave mode first enters the continuum it would be strongly desirable to have adequate resolution to investigate any broadening of the spin wave mode

during the entry, and any anisotropy of this effect with Q direction. For such purposes the resolution in energy should be of order

$$\Delta\omega \sim 0.01 \text{ eV}$$

and in particular the Q resolution must match this, taking into account the slope of the spin wave mode at this point. This slope is of order $200 \text{ meV}/\text{\AA}^{-1}$ so the Q resolution necessary is of order

$$\Delta Q \sim \frac{10}{200} \sim 0.05 \text{ \AA}^{-1}$$

On the other hand at rather higher energies when the continuum band modes are highly damped, then the intensity distribution will be only slowly varying in Q and ω and much worse resolution will be tolerable, particularly in the case of early experiments to identify the rough form of these excitations. For this region we may specify

$$\Delta\omega \sim 0.02 \text{ eV}$$

$$\Delta Q \sim 0.2 \text{ \AA}^{-1}$$

which would enable measurements to be taken in 10 steps across the zone in Q and 20 energy steps up to 0.4 eV.

Clearly these two resolution requirements are very different and require source intensities differing by two orders of magnitude ($4 \times 4 \times 4 \times 2$). We shall therefore adopt a third approach of seeing the resolution figures obtainable from the machine discussed in the introduction for the case of a typical energy transfer required for this type of experiment. We take the following scattering diagram

$$E_0 = 0.5 \text{ eV}$$

$$E_1 = 0.24 \text{ eV}$$

$$\hbar\omega = 0.25 \text{ eV}$$

$$k_0 = 15 \text{ \AA}^{-1}$$

$$k_1 = 11 \text{ \AA}^{-1}$$

$$Q = 4 \text{ \AA}^{-1}$$

The machine parameters proposed were as follows:

source pulse length	6 μs	source-chopper distance	15 m
chopper pulse length	3 μs	source size	10 cm x 10 cm

These parameters give an energy resolution on the incident beam $\Delta E_0 \sim 5 \text{ meV}$. This is therefore in the upper range of our desired resolution requirements in energy. To obtain adequate Q-resolution we choose a sample size of 2 x 5 cm. This gives a machine with the following overall resolution requirements

$$\Delta E = 7 \text{ meV}$$

$$\Delta Q = 0.1 \text{ \AA}^{-1}$$

and thus fits in fairly well with the range of resolution requirements suggested except that the Q resolution is somewhat worse than ideal for a given energy resolution.

Expected Intensity

The incident flux calculated for this experimental configuration is

$$J = 10^4 \text{ n cm}^{-2} \text{ sec}^{-1}$$

if we assume the scattering intensity corresponding to the larger contour in the calculations of Ref. 5, Fig. 22. The count rate expected from a 100 gm Ni^{60} specimen of 10 cm^2

area is then

$$N = \frac{m}{M_{Ni}} \times N_O \times J \times \frac{d^2 \sigma}{d\Omega dE} \times 4\Omega \times \Delta E \times t$$

$$= 160 \text{ cts/day}$$

m = weight of crystal

M = molecular weight

N_O = Avogadro's number

t = run time

This count rate is near the minimum which could be used successfully but could be used to great advantage if the background counts were also of this order. For example with equal signal to background we could measure the signal to within 15% in one day if the background could be assumed sufficiently flat that it could be deduced from the energy gain side of the spectrum where there will be no possible true scattering from the sample at these energies.

Background Considerations

It is impossible to be quantitative in this connection, but the expectations are that the ZING spectrometers might have a very favorable background level compared to a steady state reactor giving an equal signal. At 60 Hz repetition frequency the total time before the next pulse is created at the source is some 14,000 μ s. In the above example the signal of interest arrives at the counter after a total time of 1750 μ s. Thus there should be ample time for the fast neutron and γ flash to decay before the time interval during which the counter would be measuring the energy transfers of interest.

The length of the time interval(s) during which the

counter measures the energy range $\Delta E = 0.007$ eV which we assumed may be estimated from

$$\delta^2 = (\Delta t)_{\text{burst}}^2 + \left(\frac{1}{2} t_1 \frac{\Delta E}{E_0}\right)^2$$

$$\delta = 4 \mu\text{s}$$

A well shielded counter in a reactor environment will usually have a background say of order 1 ct/min. This figure is 1,400 cts/day but will be divided by the ratio of the "gate width" δ to the total time between bursts. Thus the effective background count rate may be

$$n = \frac{1,400 \times 4}{14,000}$$

$$= 1/2 \text{ ct /day}$$

and is thus quite negligible, particularly since the low average thermal flux from ZING should enable the total counter background to be reduced below 1 ct/min.

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2. Low Energy Transfer Inelastic Scattering

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I. Introduction

The applications we have identified for low energy transfer inelastic scattering lie primarily in the field of materials research. Of wide practicality are applications in the fields of liquids, superconductors and molecular biology. In all these areas there should be a continuing need for inelastic scattering work over the foreseeable future (one or two decades) and ZING is competitive with the best available neutron sources at Grenoble, Oak Ridge, and Brookhaven.

It seems likely that conventional reactors are approaching a limit of thermal neutron flux due to heat dissipation problems and will not be capable of much further development. Pulsed instruments such as ZING have not reached any such limit and future developments for the 1980's would seem likely to be in this area. ZING may thus serve as an important step in the long-term development of neutron beam research. Over the past two and half decades this technique has been highly successful in providing major breakthroughs in describing materials, particularly in the areas of lattice vibrations and magnetic structure and dynamics and in producing scattering law data essential for the design of reactors. In the area of scattering law applications data of sufficient accuracy now exists to satisfy the needs of reactor engineers, but in the areas of lattice vibrations and magnetism it is probable that work will continue for more than two decades.

In Tables 2.1 - 2.4, the areas of application of ZING to low-energy transfer inelastic scattering are shown in some detail. Under the general heading of materials research, the applications have been subdivided into five categories: solids, liquids, superconductors, molecules and molecular biology. These fields are discussed in detail in Sections II through VI.

We have selected three of these applications as being especially noteworthy. Firstly, liquids is a field of vast

extent which is imperfectly understood despite recent advances triggered by neutron scattering measurements. We would like to see programs mounted to study water (pure and impure), petroleum products and liquid metals.

Secondly, the need for higher temperature superconductors is clear and we believe a program should be mounted to give basic data in this area. Superconductivity involves a coupling of electrons and phonons in a metal. Current work has largely proceeded with data available only about the electrons and a program should and could be started to give basic phonon data over a wide variety of superconducting alloys.

Thirdly, there is an embryonic interest in molecular biology. Neutron inelastic scattering can tell of the dynamics of biological molecules. In the past such dynamical studies have tended to follow structural studies by about two decades and there is no reason to expect this trend not to apply here too.

The performance of ZING for each area within a category has been characterized by the letter A, B, or C. Class A areas are those where this neutron source is expected to be fully competitive with the best steady-state reactors in the world. There are no areas in the field of low energy inelastic scattering where ZING can be greatly superior since its peak thermal flux is of the same order as in the best steady-state reactors. Class B experiments are those where ZING will be around an order of magnitude weaker than the best steady-state reactors. Even here ZING gives an improvement by an order of magnitude over the existing CP-5 reactor at Argonne and would represent a useful regional facility. Class C experiments represent those cases where ZING is comparable with the Argonne CP-5 reactor.

The final section of the report describes some possibilities for instruments that might be built to carry out the types of study envisaged.

Table 2.1: Areas of application of ZING for low energy inelastic scattering. Tables 2.2-2.4 detail each of the indicated subareas.

(a) SOLIDS

(A occasionally)

Table 2.2: Application of ZING to solids.

1. Metals 2. Salts 3. Petroleum 4. Water 5. Helium 6. Hydrogen
products

e.g., lithium

A

A

A

A

B

A

Table 2.3: Application of ZING to liquids.

(c) SUPERCONDUCTORS

Measurements of phonon
frequency spectra

A

(d) MOLECULES

1. Lattice dynamics in molecu- lar crystals	2. Molecular vibration spectros- copy	3. Dynamics of Polymers	4. High conduc- tivity organic salts (field just becoming established)
C	A	B	A

(e) MOLECULAR BIOLOGY

This field will open soon

A

Table 2.4: Application of ZING to superconductors, molecules
and molecular biology.

II. Solids

(a) Single Crystals. Experiments in this category have up to now been done very successfully on steady-state reactors using a triple axis spectrometer. In this way the lattice dynamics (and magnon dispersion curves) have been measured along high symmetry directions for a large number of simple systems, and it is quite clear that a steady-state neutron source is superior to a pulsed source for this particular application. However, the effectiveness of the pulsed source is increased for systems with low symmetry as well as for experiments where phonons (or magnons) in off-symmetry directions are of value. Present trends indicate that interest is heightening in these areas.

(b) Polycrystalline Materials. Experiments done using polycrystalline samples can be done equally well on a pulsed or on a steady-state source. In this category measurements of the phonon density of states are probably the most important. Even though the phonon density of states provides much less detailed information about the lattice dynamics of a system than a set of single crystal phonon-dispersion curves, the ready availability of polycrystalline materials makes this class of experiments important. The phonon density of states, intimately connected with the thermal properties of a material, could be measured and catalogued for a large number of materials. (It should be noted that neutron inelastic scattering from materials with more than one type of atom gives a frequency distribution weighted in a special way according to the eigenvectors. This information is still useful, however, for identification purposes and qualitative comparisons of materials.) Effects of impurities and defects are a newly developing subfield which should become widely studied in the next five years.

(c) Diffusion. Quasielastic neutron scattering has proved a valuable tool in the study of diffusional motions in solids, both translational diffusion (e.g., of interstitial

hydrogen in transition metals) and rotational diffusion (e.g., of SH^- and similar molecular groups in crystals). By studying the width of the peak as a function of momentum transfer, one obtains information about jump rates and mechanisms and site occupations. Present neutron techniques are mostly limited to diffusion constants of the order of $10^{-5} \text{ cm}^2/\text{sec}$. It appears feasible to use this type of source to extend these measurements down to $5 \times 10^{-7} \text{ cm}^2/\text{sec}$, where a whole new range of materials problems can be explored; for example, diffusion in substitutional alloys in which thermal vacancies are preferentially associated with the solute atoms. This has a promising potential for the study of defect and radiation damage problems incurred in structural materials for fission reactors, since stable voids are invariably nucleated at special points. The present studies of hydrogen diffusion through transition metals, a problem of great practical interest in both fission and fusion reactor design, could be extended to lower temperatures and concentrations.

(d) Magnetism. Over the past 20 years the neutron scattering technique has been shown to be a very important tool for studying the properties of magnetic materials. For many experiments the pulsed neutron source is as good as the steady-state source; e.g., paramagnetic scattering experiments from which important information about crystal-field and exchange interactions can be obtained.

In the past few years the crystal field interactions in many metallic rare-earth compounds have been determined from inelastic neutron scattering experiments, and it has been shown that the inelastic neutron scattering technique is a very powerful tool for the study of crystal field effects in metallic systems for which the optical spectroscopy technique is not applicable. For many compounds that one is interested in studying, the exchange interactions are as strong or stronger than the crystal field interactions, and neutron spectra that one would obtain for such systems will be difficult to analyze. A successful interpretation of the results,

however, would provide very valuable information on magnetic interactions in the rare earth and actinide metals and metallic compounds. In particular we could enhance significantly our present understanding of the interaction between the f-electrons and the conduction electrons.

(e) Amorphous Solids. In this category there are many technologically important materials; e.g., amorphous semiconductors, amorphous magnets, glasses and some ceramics. The study of the dynamical behavior of these materials by neutron scattering will provide information that is important in understanding the properties of amorphous solids. For this application the pulsed neutron source is as good as the best steady-state source. The importance of this area and of studies of the type proposed has been emphasized by the National Research Council ad hoc committee report entitled "Fundamentals of Amorphous Semiconductors."

III. Liquids

The liquid state, as one of the three states of matter, lies midway between the long-range order of the solid state and the random order of the gaseous state.

Liquids are an important part of the "real world", the world in which man lives. Ninety percent of the human body, for example, is water. Yet the liquid state is probably the least well defined state of matter. It appears at times to have solid-like behavior, and at other times to have gaseous-like behavior and adaptations of the theoretical descriptions of solids or gases are used at times to explain liquids. There is really not an adequate theory of liquids.

Because of its relevance and its ill definition, research on the liquid state is an important area and neutrons can add significant data and insight. Thermal neutrons carry momentum and energy that are well matched to measuring the spatial relations and motions of the atoms in liquids. One outstanding example of this is the measurement of the detailed behavior of

the excitations in liquid He. These excitations had been postulated by Landau and Feynman but were detected and measured only when a large flux of thermal neutrons became available.

Neutron studies of liquids are divided into several areas:

(a) Liquid metals. Here one is interested in the primary and binary liquid metals. One can measure both the collective motions and the motions of individual atoms. This gives information about interatomic forces. Measurements could be made across "phase changes", extensions of solid phase changes into the liquid state. The persistence of order as one moves away from the liquidus line would also be interesting. Studies of liquid metals under very high pressures as they become viscous and near solidification are probably important. Besides being germane to understanding liquid metals this knowledge may be valuable for better engineering predictions in systems that use liquid metals, such as Na in the LMFBR and Li in the CTR reactors. The knowledge should also be important to metal casting.

(b) Molten Salts. These studies parallel those of liquid metals, but the forces are ionic instead of metallic. Besides basic information, molten salts are important to the chemical processing industry.

(c) Petroleum liquids. Petroleum products are basic to the US energy base. All petroleum products contain hydrogen and neutrons have been demonstrated to be uniquely capable of analyzing the dynamical behavior of hydrogen atoms in water. The evidence of jump type diffusion is one example where neutrons have provided unique data.

(d) Water. Water is basic to life on earth; yet it is one of the more complicated and poorly understood materials. Since water contains hydrogen, neutrons are again well suited to its study. Water possesses a set of low energy states that should be precisely measured as functions of temperature and pressure and along the freezing curve.

(e) Hydrogen. Study of liquid hydrogen both cold and under pressure should be interesting. Studies under pressure might provide evidence of the change in short-range order that would allow extrapolation to the metallic state which is postulated to be a room temperature superconductor. Since for this measurement the sample is hydrogen and must be placed in a thick walled pressure cell, neutrons are the only form of radiation that stand a chance of providing such information.

(f) Helium. As mentioned previously, much valuable information has already been obtained by thermal neutron inelastic scattering on the quantum nature of liquid ^4He . Measurements of liquid ^3He and ^3He - ^4He mixtures are also needed to provide basic comparisons between Fermi and Bose type materials. The area of study should be extended to cover a wider range of temperature and pressure. This field overlaps that of the high-energy transfer group and is discussed in more detail by that group.

IV. Superconductors

There are enormous potential uses for superconductors if transition temperatures can be raised from the present upper limit of 21°K to something nearer to 100°K . We believe a neutron scattering program should be mounted to further the search and to broaden its base.

It is established that superconductivity arises from an interaction between electrons and phonons in a metal. To date searches for superconductors have mainly been based on electron properties because there is almost no information available on phonon properties in alloys. ZING would be ideally suited to the measurement of phonon densities of states in a large variety of alloys. The measuring techniques are well established and will give sufficiently accurate data for the purposes in hand.

V. Molecules

There are at least five areas of molecular chemistry to which inelastic neutron scattering (INS) has made significant contributions. These are: (1) the study of intermolecular forces in molecular crystals, (2) the dynamics of molecular rate processes in molecular crystals, liquids, and solutions, (3) molecular vibration spectroscopy, (4) surface chemistry and catalysis, and (5) dynamics of polymers. References to review articles on these subjects can be found in the Proceedings of the 1972 Conference on Neutron Inelastic Scattering.¹ It is noteworthy that approximately one-fifth of that conference proceedings concerns molecular crystals, chemistry and polymers.

The time-of-flight (TOF) method of INS spectroscopy has made contributions to all of the five areas referred to above, but the area where it promises to make its most important contribution in the future is molecular vibration spectroscopy. INS spectroscopy is not restricted by selection rules such as those governing infrared (IR) and Raman spectroscopies. There is a great interest among chemists in measuring optically inactive modes in molecules. There are also cases where optically allowed modes are very weakly seen by IR and Raman methods but are quite easily seen with INS. The following are some examples of molecular vibration spectroscopy. The libration motion of methyl groups in neopentane and propane have been observed by Grant, et al.,² using the TOF apparatus of Brugger at the MTR. Strong and Brugger also observe methyl motions in ethane. The characteristic spectra of such molecules as KHF_2 , maleic acid ($\text{C}_4\text{H}_4\text{O}_4$) and trichloroacetic acid ($\text{C}_2\text{Cl}_3\text{O}_2\text{H}$) have been measured; even aromatic molecules like paradichlorobenzene ($\text{C}_6\text{H}_4\text{Cl}_2$) and 1,2,4,5-tetrachlorobenzene have been measured. In the later case there is good agreement between the measured and calculated spectra.

These studies are well suited to the TOF method because the systems studied are either liquids or gases under pressure.

Although the cross section for observing these effects is dependent on momentum transfer there is cylindrical symmetry and this allows one to use multiple detector arrays or ring counters and thereby gain some advantage over triple axis spectrometers (TAS). The fact that the energy of these vibrations is from 20 meV to perhaps 120 meV favors neutron energy loss scattering with neutrons initially in the 100 meV to 300 meV range. The spallation source will have more neutrons in this energy range than a reactor. One of the difficulties with the present measurements is the lack of energy resolution. Although INS will never approach the resolution of IR and Raman scattering, the effective higher flux of a machine such as ZING will probably give a gain of intensity of one to two orders of magnitude over reactors such as CP-5 and so allow considerably improved resolution to be obtained.

The recent discovery of high conductivity (superconductivity?) in conducting organic salts³ suggests a new area of inelastic neutron scattering from molecular systems which may well be very important in the near future.

The study of polymers by TOF-INS has to date provided only qualitative information on their dynamics. For an excellent review of this subject, see G. Allen in the Neutron Inelastic Scattering Conference Proceedings.⁴ More quantitative information is coming from studies on crystalline polymers using TAS. Nevertheless, we must remember that polymers are often rubbers and amorphous or glassy substances. With a gain of a factor of 10 in resolution, incoherent INS will stimulate advances in the field. Once again the characteristic spectra obtained for molecular subunits will give information on the effect of the polymer environment on these molecular units. The temperature and pressure variation of these characteristic spectra will also be useful. The importance of studying polymers cannot be overemphasized. Rubbers and plastics play a very important part in our lives. Furthermore polymers have many of the properties of proteins. Proteins are classed as special types of copolymers; i.e.,

polymers with more than one species of molecular subunit in the polymeric chain. This brings us to the discussion of molecular biology.

VI. Molecular Biology

Very little work has been done using INS from biological molecules. This is probably because biophysicists are concentrating on the determination of the structure of biomolecules. It is just a matter of time (perhaps five years) before interest turns to the dynamics of these molecules. It is hard to guess at this time what then will be of importance. We know that muscle fiber and even DNA molecules have properties in common with polymers. This suggests that expertise gained in studying the properties of polymers could be applied to understand the dynamics of proteins. One can conceive of measuring DNA vibrational modes in crystals with one-dimensional order. INS spectroscopy has at least one advantage over more energetic radiations--neutrons are less destructive to the sample.

The TOF method would have an advantage over TAS because, initially at least, experiments would be done on isotropic systems where large volumes of momentum space can be covered at once. Because of the large number of biologists in this country, we feel confident in predicting a large number of future users for a ZING-type neutron source.

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VII. Instrumentation

(a) A General Purpose Instrument for Inelastic Scattering

A simple way to design and commission an inelastic scattering instrument for a pulsed spallation source would be to transfer an instrument already existing at a steady-state reactor. An instructive example is to consider the gain in intensity realized by moving the Thermal Neutron Time-of-Flight System now at the CP-5 reactor at Argonne to a machine like ZING. The specifications and performance of this machine are well documented.¹ It has been used for a wide variety of inelastic and quasi-elastic scattering experiments in solids, liquids and glasses, and the corresponding performance at ZING can be easily calculated on the basis of numbers given in ANL/SSS-72-1.

Schematically, the change can be described by the two sketches shown in Fig. 2.1.

Fig. 2.1. Schematic illustration of an inelastic scattering instrument as set up (a) at CP-5 and (b) at ZING.

At CP-5 the highest incident energy used is $E_0 = 35$ meV with the incident energy resolution (FWHM) ΔE_0 given by

$$\frac{\Delta E_0}{E_0} \sim \frac{1.1 \text{ meV}}{35 \text{ meV}} = 3.1\%$$

The burst time τ_1 of the rotor is $6 \mu\text{s}$, giving somewhat better resolution in ΔE_1 for elastically scattered neutrons which becomes poorer, however, for neutrons scattered with energy

gain. For ZING, which has a spectrum relatively richer at higher energies, it would seem logical to choose $E_0 = 50$ meV and $\tau_1 = 5$ μ s: In order to keep ΔE_0 the same, we require a spread Δn_0 in the incident inverse velocity n_0 given by

$$\frac{\Delta n_0}{n_0} = \frac{1}{2} \frac{\Delta E_0}{E_0} = \frac{1.1 \text{ meV}}{2 \times 50 \text{ meV}} = 1.1\%$$

According to ANL/SSS-72-1 the source pulse width τ_0 for 50 meV neutrons is about 30 μ s, so the required incident path length l_0 is given by

$$\frac{\tau_0}{l_0 n_0} = \frac{30 \text{ } \mu\text{s}}{l_0 \times 320 \text{ } \mu\text{s/m}} = 1.1\%$$

and so $l_0 = 8.5$ m.

The average intensity on the sample is given by (compare Eq. (11) of Ref. 1)

$$I_0 = \hat{I}_{th} \frac{A}{l_1^2} M \frac{\Delta \lambda_0}{\lambda_0} T$$

where I_0 = average no. of neutrons hitting the sample per second

$\hat{I}_{th}$ = peak thermal beam current per unit solid angle
 $= 1.34 \times 10^{16}$ n/str/sec (from ANL/SSS-72-1)

A = usable sample area
 $= 25 \text{ cm}^2$ for a 2.5 cm x 10 cm rotor aperture

M = Maxwellian factor $2e^{-(V_0/V_T)^2} (V_0/V_T)^4$
 where V_T is neutron velocity corresponding to the moderator temperature

≈ 1 at 50 meV for room temperature moderator

$$\begin{aligned} T &= \text{chopper transmission} = 5 \text{ } \mu\text{s} \times 60 \text{ cps} \times \text{attenuation} \\ &= 3 \times 10^{-4} \times \text{attenuation} \\ &\approx 2.4 \times 10^{-4} \text{ for present design} \end{aligned}$$

Thus

$$\begin{aligned} I_o &= \frac{(1.34 \times 10^{16}) \times 25 \times (1.1 \times 10^{-2}) \times (2.4 \times 10^{-4})}{(8.5 \times 10^2)^2} \\ &= 1.22 \times 10^6 \text{ n/sec} \end{aligned}$$

Other attenuation factors (duty cycle of ZING, windows in the incident flight path, etc.) might bring this down to a practically reliable figure of 1.0×10^6 n/sec:

$$I_o = 1.0 \times 10^6 \text{ n/sec}$$

compared with the value measured¹ at CP-5 (which agreed quite well with that calculated) of 1.7×10^4 . It is seen that an intensity gain of about 60 can be immediately realized at ZING. This is in spite of the fact that the energy resolution expressed as a percentage of the incident energy has actually improved. The gain is even higher than that which would be obtained by moving the facility to a reactor like HFBR or HFIR, since the facility makes maximum use of one of the two 12" diameter beam tubes at CP-5 which do not exist in the higher flux reactors.

A factor of this sort would make some presently formidable experiments feasible. For example, measurements of the elementary excitations of a 5% solution of ^3He in ^4He have been recently made at CP-5.² In the corresponding facility at ZING measurements on pure ^3He (which has a 5000-barn

absorption cross-section for thermal neutrons) are clearly feasible. For most other experiments it would be preferable to absorb the gain in signal into improvement in the energy resolution. Probably a factor of three in resolution could be achieved for counting rates comparable to those now found at CP-5, bringing the overall energy resolution ΔE down to ~ 0.5 meV, which is 1% of the incident energy E_0 .

It should be noted that this instrument can be very simply adapted to make use of correlation techniques appropriate for pulsed sources.³ This can be achieved by replacing the simple chopper shown in Fig. 2.1 (b) by a chopper with a transmission function corresponding to a pseudo-random binary sequence. In this way about 50% of the energy spectrum incident on the chopper can be utilized. This technique would be advantageous for certain classes of experiment.

(b) High-Resolution Quasi-Elastic Scattering Instrument

For a high-resolution quasi-elastic scattering instrument it is more economic to work in "inverse" geometry; i.e., to use a monochromator in the scattered beam and measure the energy spectrum of the incident beam by time-of-flight. Schematically, this can be represented as in Fig. 2.2.

Fig. 2.2. Schematic illustration of high-resolution quasi-elastic scattering instrument.

More than one analyzing crystal-detector assembly can be installed but in general not as many detector units are possible as for "direct" geometry (e.g., the instrument described previously).

In the notation used previously, we select the following parameters:

$$E_0 \sim 10 \text{ meV, so that the burst time } \tau_0 \sim 45 \text{ } \mu\text{s} ,$$

$$\text{sample} \sim 2.5 \text{ cm wide} \times 10 \text{ cm high} ,$$

$$l_0 \sim 50 \text{ m}$$

Then

$$\frac{\Delta n_0}{n_0} = \frac{\tau_0}{l_0 n_0} = \frac{45 \text{ } \mu\text{s}}{710 \text{ } \mu\text{s/m} \times 50 \text{ m}} = 1.27 \times 10^{-3}$$

$$\frac{\Delta E_0}{E_0} = 2.54 \times 10^{-3}$$

$$\Delta E_0 = 2.54 \times 10^{-2} \text{ meV}$$

The effective solid angle would be $\frac{25}{(5000)^2} = 10^{-6}$, which implies a very small neutron intensity at the sample. This can be increased by a bent guide tube with the length of l_0 . For 10 meV, $\Delta k_{\perp}/k_{\perp} \approx 10^2$, which would imply a solid angle of 10^{-4} . Imperfect transmission in the tube and the fact that the sample is smaller than the source may bring this down by a factor of 8, so that:

$$\text{Effective solid angle } \Delta\Omega \sim 1.25 \times 10^{-5} .$$

$$\text{Maxwellian factor } M \approx 0.11 \text{ for } 10 \text{ meV} .$$

Then the average intensity at the sample is given by

$$I_o = \hat{I}_{th} \Delta\Omega M \frac{\Delta\lambda_o}{\lambda_o} T ,$$

for neutrons with wavelengths in the range $(\lambda_o, \lambda_o + \Delta\lambda_o)$, where T is the duty cycle of the machine

$$= 45 \times 10^{-6} \times 60 = 2.7 \times 10^{-3} .$$

Then

$$\begin{aligned} I_o &= (0.89 \times 10^{16}) \times (1.25 \times 10^{-5}) \times 0.11 \times (1.27 \times 10^{-3}) \\ &\quad \times (2.7 \times 10^{-3}) \\ &= 4.2 \times 10^4 \text{ n/sec} . \end{aligned}$$

We now consider the scattering power of the sample. It would be highly desirable to be able to measure the diffusion of one metal through another, for example, at a concentration of $\sim 1\%$.

$$\begin{aligned} \text{Scattering power} &= n_{\text{defect}} \sigma_{\text{inc}} \\ &\sim 10^{-2} n_{\text{host}} \sigma_{\text{inc}} \\ &\sim 10^{-2} \times (5 \times 10^{22}) \times 10^{-24} \times 2.5 \\ &\quad \text{for a typical metallic sample} \\ &\sim 1.25 \times 10^{-3} . \end{aligned}$$

For the analyzer we also need a resolution given by

$$\frac{\Delta\lambda_1}{\lambda_1} \sim \frac{\Delta\lambda_o}{\lambda_o} = 1.27 \times 10^{-3}$$

and a range of scattering angle given by

$$\Delta\theta_1 \sim \Delta\theta_0 \sim 10^{-2} \quad .$$

Now

$$\frac{\Delta\lambda_1}{\lambda_1} \sim \cot \theta_1 \Delta\theta_1$$

so that

$$\cot \theta_1 \sim 0.127$$

$$\theta \sim 82.8^\circ \quad .$$

If the detector has the same dimensions as the sample and $\Delta\theta_1$ is achieved by a Soller collimator with vertical slits (perhaps 0.5 cm apart and 50 cm long), the vertical collimation $\sim \frac{10 \text{ cm}}{50 \text{ cm}} = 1/5$ radian. Thus the scattered solid angle = $10^{-2} \times 1/5 = 2 \times 10^{-3}$. If the crystal is 50% efficient and the detector 70%, number of neutrons scattered into the detector I_1 is given by

$$\begin{aligned} I_1 &= (4.2 \times 10^4) \times (1.25 \times 10^{-3}) \times \frac{2 \times 10^{-3}}{4\pi} \times 0.5 \times 0.7 \\ &= 2.9 \times 10^{-3} \text{ n/sec} \\ &= 0.175 \text{ n/min} \end{aligned}$$

For 10^3 cts in the quasi-elastic peak we would need 5.72×10^3 mins = 4 days, not an unreasonable counting time. We can expect the average background to be much below this counting rate. The energy resolution is given by

$$\begin{aligned} \sqrt{\Delta E_0^2 + \Delta E_1^2} &= 2.54 \times 10^{-2} \text{ meV} \times \sqrt{2} \\ &= 3.6 \times 10^{-2} \text{ meV} \end{aligned}$$

$$\begin{aligned}\text{In terms of frequency transfer, } \Delta\omega_{\text{res}} &= 5.46 \times 10^{-2} \text{ ps}^{-1} \\ &= 5.46 \times 10^{10} \text{ s}^{-1}\end{aligned}$$

In the range of diffusive behavior, we expect $\Delta\omega_Q \sim 2 DQ^2$.

In order to measure small diffusion constants D , we would like Q to be as large as possible. However, the range where the simple diffusive behavior holds normally lies below $1 \text{ \AA}^{-1}$, after which various jump mechanisms begin to dominate the incoherent scattering. However, one can expect to extract a line width from a clearly resolved quasi-elastic peak down to about one-fifth of the resolution width. Thus the minimum diffusion constant observable is given by

$$\begin{aligned}D &= \frac{\Delta\omega_Q}{2Q^2} = \frac{1/5 \Delta\omega_{\text{res}}}{2Q^2} \\ &\sim \frac{10^{10} \text{ s}^{-1}}{2 \times 10^{16} \text{ cm}^{-2}} \\ &\sim 5 \times 10^{-7} \text{ cm}^2 \text{ s}^{-1}.\end{aligned}$$

This is one to two orders of magnitude below those currently being measured in typical quasi-elastic scattering experiments.

Even smaller diffusion constants could be observed if the incident energy was reduced to smaller values. However, it is generally useful in such experiments to go up the first reciprocal lattice vector and beyond in order to test various models for jump mechanism and site occupation. For this purpose incident energies of 5 meV have been found to give too restrictive a range of momentum transfer.

(c) An "Inverted Geometry" Spectrometer for Paramagnetic Scattering Experiment

For paramagnetic scattering experiments on polycrystalline materials a design of a spectrometer using the "inverted geometry" has been considered. A layout of the instrument is shown in Fig. 2.3. The energy, E_s , of the scattered neutrons is determined by the Bragg condition:

Fig. 2.3. Schematic illustration of paramagnetic scattering spectrometer.

$$\lambda_s = 2d_{\text{Anal}} \sin \left(\frac{\pi}{2} - (\phi + \alpha) \right) . \quad (1)$$

where α is the angle between the incident beam and the normal to the reflecting planes.

The wavelength, λ_s , (or the energy, E_s) as well as the scattering angle, ϕ , are determined by the position of the detector, and the energy of the incident neutrons is determined from the time of arrival of the scattered neutrons in the detector. The neutron energy and momentum transfers are given by

$$\Delta E = E_0 - E_s = \frac{\hbar^2 k_0^2}{2m} - \frac{\hbar^2 k_s^2}{2m} \quad \text{and} \quad (2)$$

$$Q^2 = k_0^2 + k_s^2 - 2k_0 k_s \cos \phi .$$

From Eqs. (1) and (2) $(Q, \Delta E)$ curves can be calculated as a function of the incident energy, E_0 , for fixed values of ϕ , α and $2d_{\text{Anal}}$. In Fig. 2.4 several curves are shown. These curves have been calculated for a (220)-Cu-crystal ($2d_{\text{Anal}} = 2.55 \text{ \AA}$) as analyzer. For both the two values of α the area that is covered in the $(Q, \Delta E)$ -plane for $5^\circ \leq \phi \leq 20^\circ$ is very useful for studying interactions in paramagnetic systems. Furthermore the incident energies that are utilized are in the vicinity of the peak of the Maxwellian Spectrum.

For an estimate of the resolution the point in $(Q, \Delta E)$ -space that corresponds to $\alpha = 20^\circ$, $\phi = 10^\circ$ and $E_0 = 25 \text{ meV}$ will be considered. For these initial values one has

$$\Theta_s = 90^\circ - (20^\circ + 10^\circ) = 60^\circ, \quad \Delta E = 8.2 \text{ meV} \text{ and } Q = .83 \text{ \AA}^{-1} .$$

For simplicity no collimators will be included between sample and detectors, and the width, w_{sample} , will be taken to be the same as the width, w_d , of a detector. Furthermore $L = L_1 = L_2$. The analyzing crystal will be assumed to be perfect. The following values of the widths and lengths have been chosen:

$$w_{\text{source}} = 10 \text{ cm}, \quad w_{\text{sample}} = w_{\text{detector}} = 2.5 \text{ cm}$$

$$L_0 = 1000 \text{ cm} \text{ and } L = L_1 = L_2 = 100 \text{ cm} .$$

The contribution to energy resolution from the analyzing system is given by:

$$\delta E_s = 2E_s \cot \Theta_s = 2 \times 16.8 \times \frac{1}{\sqrt{3}} \times \frac{2.5}{2 \times 100} \text{ meV} = .24 \text{ meV} .$$

Fig. 2.4. Regions of $(Q, \Delta E)$ space covered by instrument shown in Fig. 2.3.

For E_0 one has two contributions, the pulse width, δt_p of a 25 meV neutron and the time resolution δt_s of the analyzing system.

$$\delta t_p \approx 40 \mu s \quad (\text{from ANL/SSS-72-1}) \quad .$$

The time resolution, t_s , is much smaller than t_p , and an estimate of δt_s gives

$$\delta t_s \cong 11 \mu s .$$

The inverse velocity n_o of the incident neutrons is 457.2 $\mu s/m$ and so

$$t_o = n_o L_o = 4572 \mu s$$

$$\delta t_o = \sqrt{\delta t_p^2 + \delta t_s^2} \cong 41 \mu s .$$

Therefore

$$\delta E_o = 2E_o \times \frac{\delta t_o}{t_o} = 2 \times 25 \times \frac{41}{4572} = .45 \text{ meV}$$

and the energy resolution

$$\delta E = \sqrt{\delta E_o^2 + \delta E_s^2} \cong .5 \text{ meV} .$$

The derivation of the momentum resolutions is quite complex, and so the detailed derivation will not be given. A calculation of the uncertainty, δQ , in magnitude of Q gives

$$\delta Q \cong .05 \text{ \AA}^{-1} .$$

For many experiments the above values of δE and δQ are very good.

The length of a (220)-Cu crystal that is required to cover the range of ϕ between 5° and 20° for $L = 100 \text{ cm}$ is about 31 cm. For a 10 cm high sample a Cu-crystal that is 10 cm by 31 cm is adequate.

One great advantage of this particular spectrometer design is its simplicity. Approximately, 20 1-inch detectors are sufficient to cover ϕ between 5 and 20°, and the design does not require the sophisticated electronics that is needed for interphasing a rotating device (like a chopper) to the pulsed source. The cost of this spectrometer would therefore be modest.

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3. Diffraction from Single Crystals

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I. Introduction

The task given to this group was to evaluate the ZING-type spallation neutron source for investigations of elastic scattering from single crystals. Such investigations are commonly employed by a wide variety of scientists, in disciplines ranging from metallurgy to biology, to determine crystal structures that range from simple alloys to complex proteins. In principle, either X-ray or neutron diffraction can be employed, and indeed in most cases the two techniques are complementary. However due to the unavailability of intense neutron sources, X-ray investigations are much more common, although frequently the use of neutron diffraction would provide additional information and better results. A related area, and one in which neutron diffraction provides unique information, is the study of magnetic phenomena in single crystals.

Existing neutron diffraction techniques usually involve the use of crystal monochromatized beams for determining crystal structure factors from the measured Bragg reflections. In the light of our experience with this technique, commonly used at steady state reactors, we must ask what advantages, if any, a pulsed source would have for measuring single crystal Bragg intensities and compare the usefulness of such a source with that of presently available medium and high-flux reactors. Most important, pulsed sources are ideally suited to the adoption and use of white beam techniques which effectively utilize the full thermal neutron spectrum. This should be compared to the usual monochromatic techniques which make use of less than one percent of the usable source spectrum. Taking advantage of this feature will, we shall see, make a pulsed source such as ZING, which has a relatively low time-averaged neutron flux, fully competitive with the presently operating high-flux reactors for single crystal diffraction. Attention must therefore be focussed on the pulsed character of the source, and on the resultant ease of energy

separation that may be obtained with time-of-flight analysis.

The use of such 'white beams' (i.e., many wavelength components) is well known in X-ray diffraction, and is illustrated in Fig. 3.1 which is a simple Laue photograph. This photograph was taken with a stationary crystal and a white incident beam. The Laue photograph is clearly rich in detail, and contains almost all the information necessary for solving the crystal structure.^{1,2} Each Laue spot is made up of a number of Bragg reflection (orders) occurring at wavelengths λ , $\lambda/2$, $\lambda/3$ etc. The main disadvantage of the Laue technique, is its inability to separate the intensity of each spot into the individual integrated intensities of its component orders. Such a separation, however, can be readily achieved with time-of-flight analysis^{3,4,5} possible at the ZING pulsed source.

Using a single large area detector, identification and resolution of the various Laue reflections including the orders, might be accomplished merely by time-of-flight analysis in the case of simple crystals with sparsely populated reciprocal cells. However the resolution problems which arise with complex crystals and the intolerable peak to background ratios expected for hydrogen-containing crystals require the use of multiple detector systems. Large area position sensitive detectors⁶ appear to be best suited to the general problems encountered in structure studies of large complex molecules. Hence the optimum approach would match a large area position sensitive detector with time-of-flight analysis to the white beam Laue technique, to map large volumes of reciprocal space at a time.

II. Experimental Considerations

If such a white beam is used, the various Bragg intensity components are given by the Laue expression

Fig. 3.1. Laue photograph taken by back reflection of a white (polychromatic) X-ray beam incident on a stationary single crystal.

$$I_{\vec{h}} \propto \lambda^4 \varphi(\lambda) |F_{\vec{h}}|^2 T(\lambda) y(\lambda) / \sin^2 \theta ,$$

where λ is the neutron wavelength, $\varphi(\lambda)$ the incident flux on the sample, $F_{\vec{h}}$ is the structure factor, $T(\lambda)$ and $y(\lambda)$ are absorption and extinction corrections, and θ is the Bragg angle. Defining $\varphi_{\text{eff}} = \lambda^2 \varphi(\lambda)$ one obtains the expression

$$I_{\vec{h}} \propto \varphi_{\text{eff}}(\lambda) T(\lambda) y(\lambda) d_{\vec{h}}^2 |F_{\vec{h}}|^2 ,$$

for the typical Bragg component, where d_h is the interplanar spacing, and $\lambda = 2d_h \sin \theta$. The variation of $\phi_h(\lambda)$, the incident flux, from a linac pulsed-neutron source⁶ is shown in Fig.

3.2. A similar dependence would be expected for the thermal neutrons coming from the ZING moderator.

To obtain precise molecular parameters accurate values of

Fig. 3.2. Variation of neutron flux from a linac pulsed neutron source as a function of neutron wavelength. (Taken from Ref. 6).

$|F_h|^2$ need to be obtained. This immediately requires a knowledge of the incident spectrum, and the variation of absorption and extinction with wavelength. At present the latter functions present difficulties if very accurate crystal structures are required, but if the wavelength range $0.75 \leq \lambda < 3.0 \text{ \AA}$ is used, crystal structures can be determined with sufficient accuracy for most investigations.

The potential gains of the white-beam technique are evident from the construction shown in Fig. 3.3. Figure

3.3(a) depicts two Ewald spheres representing the wavelength limits discussed above. The sphere centered on the origin of reciprocal space represents that volume of reciprocal space containing wanted crystallographic information, and the shaded volume contains those lattice points which can be observed at this setting of the crystal. Figure 3.3(b) shows a similar construction for the monochromatic technique in which $\Delta\lambda/\lambda$ is commonly 0.02. All other things being equal, the white-beam technique samples reciprocal space approximately 200 times faster (in the case illustrated in Fig. 3.3) than any monochromatic-beam technique. Such a factor can be of decisive importance for the application of neutron diffraction to large biological molecules where many thousands of reflections must be measured before a molecular structure can be determined.

Unfortunately, all other things are not equal. The ratio of 200 must be modified by the time-average flux ratio corrected by a monochromator loss, which for ZING in comparison to the best high-flux reactor is $\sim 1/200$. One must also consider that peaks scanned with the monochromatic source require a number of measurements (background and intensity through the peak), and, on the other hand, that all peaks illuminated by the broad spectrum source do not experience the peak flux of the spectrum. These last two effects roughly cancel in comparing the two techniques. Assuming that the requirements of angular and wavelength resolution are similar for both methods, one finds that ZING could roughly match the data gathering power of the most advanced monochromatic technique at the best steady-state reactor. In making this comparison multiple detector systems are assumed in both cases.

The realization of these hopes, however, will require very sophisticated instrumentation. Most important is the construction of a large, high resolution, position-sensitive detector system; a greatly extended version of the resistance-wire counter^{7,8} or other type of multiple detector system.

Fig. 3.3. Constructions in reciprocal space to illustrate the use of white and monochromatic radiations in single crystal diffraction. In both figures the circle centered on the origin of reciprocal space represents the volume (or area in two dimensions) that must be measured to obtain a certain resolution in real space. In Fig. 3(a) the circles with radii $1/\lambda_{\min}$ and $1/\lambda_{\max}$ are drawn through the origin, and the direction of the radii represent the direction of the incident beam. All reciprocal lattice points within the shaded area will simultaneously diffract and may be sampled by a large position-sensitive detector. In Fig. 3(b) the conventional monochromatic system is illustrated. k_0 represents the incident beam, k_f the final beam diffracted through an angle of 2θ . With $\Delta\lambda/\lambda \approx 0.02$ only those reflections in the shaded area will satisfy the Bragg conditions.

The dimensions of such a system can be estimated from the angular and wavelength resolution requirements as given in Table 3.1. In the case requiring angular resolution (both

TABLE 3.1

Resolution requirements of TOF experiments on several unit cells.

Approx. unit cell dimensions $\text{\AA}$	$\Delta\theta^\circ$ vertical and horizontal	$\Delta\lambda/\lambda$
~ 4 (metals)	3	0.05
~ 15 (medium size organic compounds)	1	0.02
~ 60 (small proteins)	0.55	0.01
ZING (10 cm at 8 meters)	0.72	0.006

vertical and horizontal) of 0.55° and a wavelength resolution of $0.02 \text{ \AA}$ the 4π area detector must be divided into 10^5 individual elements, each connected to 100 to 300 time channels. Such an enormous system belongs at present in the realm of science fiction, and must certainly evolve from simpler systems.

The first crystallographic instrument at ZING could well be a 4-circle diffractometer with a single detector connected to a time-of-flight system. Such an instrument would collect data more efficiently than any instrument now operating at CP-5, although the loss in accuracy due to absorption and extinction functions might offset any potential gain in speed. We should emphasize here that resolution requirements and restrictions inevitably affect such comparison. For convenience we have assumed a flight path of ~ 8 meter, 7 meters from the source to sample, 1 meter from sample to detector. Longer flight paths result in intensity losses without any significant gains, since the resolution obtained with an 8 meter flight path is sufficient for most problems (see Table 3.1). However no consideration has been given to the use of beam pipes. A further requirement, again to avoid intensity losses, would be to evacuate, or fill with helium, the beam tube between the moderator and the sample. The second generation instrument should include a modest sized position sensitive detector and a computer connected to the time-of-flight system to allow the analysis of a number of Laue spots simultaneously. This would increase the effectiveness of the instrument and provide the experience necessary for the design and testing of the much larger system discussed above.

III. Magnetic Investigations

The investigation of magnetic phenomena by measuring the intensities of certain Bragg peaks is a very important subsection of neutron crystallography, because neutrons provide unique information via their interaction with magnetic moments

in solids. In this case no analogy exists with X-ray diffraction. Clearly, the development of a white-beam technique applies equally to magnetic investigations. To determine the ordered arrangement of magnetic moments in a crystal the first requirement is to determine the positions of any magnetic superlattice reflections. This suggests an immediate advantage for the white-beam technique since with it large volumes of reciprocal space (see Fig. 3.3 again) can be examined rapidly, thus eliminating the tedious scanning that is presently used in searching for superlattice reflections with a monochromatic beam. Experiments on magnetic systems usually require the sample to be at low temperature and a magnetic field is also occasionally applied to the sample. These factors are important in considering the design of a large position-sensitive detector.

Another important area of magnetic investigation depends on the availability of polarized neutrons. Thus whenever the intensity of Bragg reflection depends at least partially on the polarization of the incoming neutrons, the spin dependent cross section can be measured accurately by determining the intensity ratio for neutrons of opposite spins. Presently monochromatic polarized neutrons are obtained by diffraction from some magnetically saturated crystal. Although the efficiency of such crystals (e.g., Co-Fe) is quite low the polarized beam technique has proved very valuable for the determination of magnetic moment distributions.

Efficient utilization of a pulsed source for polarized neutron studies requires the production of highly polarized white beams. The most promising methods for this appear to be: a) the use of filters such as polarized ^1H or ^3He , b) the Stern-Gerlach technique and c) the use of polarizing mirrors or guide tubes. The use of polarizing guide tubes seems most promising since the long flight paths (8-10 meters) contemplated for ZING may well require guide tubes for most experiments. For a mirror or guide tube the critical angle θ_c for neutron reflection in the two spin states is given by

$$\theta_c^\pm = \lambda \sqrt{N(b \pm p)/\pi}$$

where N is the number of atoms per unit volume and b and p are the nuclear and magnetic scattering amplitudes respectively. To obtain a fully polarized beam, $p > b$. A suitable polarized neutron guide with a Dy surface, for example, could produce at nearly 100% efficiency a beam of polarized white neutrons for which $\Delta\theta = 0.10^\circ$ for the 1 Å component. It should be pointed out that the beam pipe would have to be maintained in a magnetic field (~ 2 kOe) at 78°K and has not been tested as yet. The resulting intensity, however, would be superior to anything presently available at high flux reactors and such a beam should be useful for a variety of solid state experiments.

IV. Mode of Governance

The development of a large position-sensitive detector with time-of-flight analysis capability, coupled with a pulsed source such as ZING, opens up unprecedented data-collection rates for single-crystal neutron diffraction. Rates of 150 Bragg reflections a day, presently obtained with a single detector 4 circle instrument at CP-5, could be increased, in principle, by several orders of magnitude. Clearly, an instrument with such capabilities can handle, and is justified only by, problems of large magnitude and a large number of users. It seems most appropriate that such a facility be developed, from the beginning, either as a regional or national facility. A large reservoir of potential interest in, and users for, such a facility exists among the Midwestern Colleges and Universities, and the main pool of users would certainly come from this area. However, the potential for wider national use should be recognized.

The proper development and utilization of such an

instrument would certainly require a strong and active group of Argonne based scientists, who would both oversee the original and continued instrument development, and carry out their own research programs. The proper division of instrument time between the local staff and visitors would, in our opinion, be best handled on the basis of a fixed formula allotting perhaps 50% of the time to visiting scientists. The actual time allocations would be made by a user committee similar to that set up for ZGS, and other national facilities. Inexperienced users would be required to gain initial experience through a cooperative project, but those with demonstrated competence would be accepted as independent users. An important objective would be to build up and maintain the widest possible group of enthusiastic users.

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4. Diffraction from Isotropic SamplesParticipants

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I. Introduction

The working group attempted to consider some aspects of the following questions:

1. What experiments in this classification are well suited to the proposed facility?
2. What are the scientific (including engineering) problems for which this facility has unique capability?
3. Who are those dedicated users of neutron scattering techniques that are most likely to participate fully in the proposed facility?
4. How should this facility be adapted to meet the need for the particular experiments indicated by the consideration of scientific problems and scientists?
5. What are the design criterion for the instruments needed to carry out diffraction studies in isotropic samples?

This broad classification of isotropic samples includes almost all materials. The consideration has been somewhat restricted by the removal of those fields where the emphasis is on energy transfer and from the separate category of single crystal diffraction. The overlap, however, is nonzero. In the case of powder diffraction one is attempting to find information also sought in single crystal diffraction. In the case of samples in which the recoil from the neutrons is not negligible, it is necessary to properly take into account the inelasticity to arrive at the static information. The following classifications by materials were used in our deliberations.

1. Polycrystalline materials
 - a. Single phase
 - b. Multiphase
 - c. Multi-components

2. Amorphous Solids
 - a. glasses
 - b. semiconductors
 - c. metals
3. Liquids
 - a. Water and solutions
 - b. Metals
 - c. Organic Solvents
 - d. Salts
 - e. Liquid Crystals
4. Polymer and other larger molecules

We attempted to identify those features of a pulsed neutron source which should be mapped onto the classification list to single out useful or unique features for studying these materials or more precisely the scientific or technical problems associated with them. This led to the following queries.

1. How useful is the extension to short wavelength beyond the range of conventionally moderated reactors?
2. How useful is the simultaneous availability of the wide range of wavelengths overlapping and extending beyond the reactor range?
3. As a pulsed system naturally leads to time-of-flight methods what are the advantages of
 - a. time-of-flight
 - b. time-of-flight at a pulsed facility?
4. What advantages arise from the time scale of the repetition rate?

Rather than systematically consider all the combinations and permutations of these questions and classification we have sampled the possible areas of application using a Monte Carlo approach of doubtful statistical validity, some of the results of which are presented here.

II. Water and Solutions

Water is undoubtedly an important scientific and technological material. The currently appearing four volume series edited by F. Franks titled, "Water, A Comprehensive Treatise", published by Plenum Press, gives ample documentation of this. In the study of water neutrons have played an important role in structure determination and the dynamics of the molecular motions. The study of water at room temperature and atmospheric pressure is still not complete. The structural changes with the physical variables temperature and pressure are yet to be investigated by neutrons. X-ray work indicates dramatic changes in the pair correlation functions with temperature. Studies on solutes in water are only recently being undertaken. Our hypothesis is that the field of water research is more than ever today an exciting and important scientific field. We conclude that neutron diffraction studies combined with x-ray studies are important tools in this field. The wavelength region with the most important structural information is 1 to $.5 \text{ \AA}^{-1}$, though even shorter wavelengths would be important in the study of solutions.

There is no doubt that the ZING facility would be well suited to such studies. The optimum design of a spectrometer for this purpose is open to further study. Two approaches have been suggested. One was equal flight paths before and after the sample to give an integration of the scattering intensity which approximates $S(Q) = \int S(Q, \omega) d\omega$. This arrangement makes full use of the white beam, without chopping. To check the validity of this approximation on a particular sample it would be sufficient to add a coarse chopper. The alternative approach is to use an $S(Q, \omega)$ machine with relaxed ω resolution.

III. Structure Determinations under High Pressure

The importance of pressure as a thermodynamic variable has been mentioned for water. For materials in general, the much more frequent use of temperature as variable is simply because the techniques are much less difficult. The number of problems in which pressure determination of structural changes are of scientific interest is obviously very large. The important point to be made here is that there are unique advantages for time-of-flight methods using a pulsed neutron source with a broad range of wavelengths. In addition to the scientific and technical importance of pressure studies there is the very important consideration that ANL already has an active program in time-of-flight methods applied to pressure investigations. This work would benefit by 2 to 3 orders of magnitude in useful count rate. In diffraction studies under pressure it is necessary for the radiation to penetrate through some of the pressure containment material. Despite the greater intensity of X-ray sources, the greater penetrability of neutrons makes the use of neutrons desirable. The advantage of time-of-flight methods is that with the wide range of wavelengths it is possible to obtain the diffraction pattern with a single exit port. Not only does this simplify the containment problem, but also makes it easier to collimate against the background from the pressure medium. These things will become more important when techniques are pushed from their present range of near 50 kilobars to above 100 kilobars. Higher pressures generally necessitate decrease in sample size. It is anticipated that most pressure work will be on isotropic samples, that is polycrystalline, amorphous or liquid materials.

IV. Glasses

The use of neutron diffraction as a means for studying the structures of glassy solids lies in two directions. The large $\vec{q}$ range accessible makes possible greater precision in determining relative positions of atoms in the disordered array. The weighting of scattering by different species according to their neutron scattering cross section makes the method a much needed complement to X-ray diffraction in these studies.

For the purposes of extending the measured range of q 's to highest values ZING would be an ideal source since the spectrum is relatively rich in high energy neutrons and since the pulse width decreases with increasing energy making the required resolution easily obtainable.

V. Service Powder Diffractometer for Chemists, Metallurgists and Material Scientists

Arguments were assembled to support the hypothesis that the utilization of powder diffraction with neutrons as a tool in chemical research has not been in keeping with its potential utility. Unlike such tools as x-ray, optical, infrared, nuclear magnetic and mass spectrometry, neutron spectrometers have not been packaged by engineers for the general market. One attempt to do this at the U.K. Atomic Energy Research Establishment at Harwell has been reported to be quite successful in creating a steady demand for such service.

It is contended here that a time-of-flight powder spectrometer is a good candidate for making routine use attractive. The entire procedure would be computer controlled including correction for spectrum, resolution, absorption and extinction. With the use of a statistical chopper just before the sample one could also correct for background and inelasticity. The sophistication of such a spectrometer combined with its simplicity to the user would be very likely to create a large

demand for its use.

We envision the use of a high resolution machine. The full moderator block should be used as a source. The moderator block should be cooled, e.g., solid or liquid CH_4 , to give a wide spectrum. Time focusing should be employed using detectors which cover shell segments in space. Three detector angles should be considered; back, 90° , and small angle. The 90° detector should be broken into several segments to allow determination of the isotropy of the sample.

VI. Numerical estimates

It is interesting to compare the intensity which could be achieved at ZING for the CP-5 High Pressure Neutron Diffraction Facility with that which could be achieved under similar conditions at CP-5.

In order to simplify intensity estimates we will assume that both set-ups will have the same angular divergence of the incident beam. At CP-5 the angular divergence is defined by the collimator between the reactor core and by the chopper itself to be 0.3° . Assuming this angular divergence and desiring to view the whole source at ZING we must set the flight path before the sample to be

$$L_1 = \frac{10 \text{ cm}}{\tan(0.3^\circ)} = 19.1 \text{ m}$$

The burst time defined by the chopper at CP-5 is $7 \mu\text{sec}$. To achieve the same $\Delta t/t$ at ZING we must have a total flight path which gives the same $\Delta t/t$ at the given wavelength. The burst time at ZING can be approximated by $\Delta t = 20 \mu\text{sec} \times \lambda(\text{\AA})$. We will match resolutions for neutrons of wavelength $3.5 \text{\AA}$ which is the range of best resolution for the CP-5 facility. For this wavelength $\Delta t \approx 70 \mu\text{sec}$ so the path length at ZING would need to be ten times as long as at CP-5 or $10 \times 2.1 \text{ m} = 21 \text{ m}$. This would give the same resolution for the two

instruments for 3.5 Å neutrons. However, at 1 Å or less the resolution on the instrument at ZING would be 3 to 4 times as good. Since the small lattice spacing lines are closest together the usable resolution would probably be increased by this same amount in most experiments.

We can now compare intensities by comparing the peak flux at ZING with the CP-5 flux since we have kept the same angular divergence of the incident beam. Then the gain in intensity is given by

$$1.5 \times 10^{15} / 3 \times 10^{13} = 50.$$

But there will be an additional factor of four because we can make the beam width 1/4" to match the sample at ZING while the CP-5 beam has been restricted to 1/16" in order to achieve a sufficiently short burst time. Thus we conclude that the gain in intensity at ZING would be a factor of 200 if we match the resolutions in the region where the CP-5 resolution is best. In the region of smaller lattice spacings the ZING instrument would have much superior resolution. It should be mentioned that because of the long distance from the source and because the source itself is only on for a small fraction of the time there would be a dramatic gain in signal to noise ratio which would make it possible to distinguish peaks much more clearly for the same number of counts in the peak.

We have not considered a number of improvements which could be made in this comparison but we will mention some of these things briefly here. Guide tubes could be used to bring the beam from the source to the sample. Guide tubes would make it possible for more experiments to be set up at the same source since each could look at a smaller area of the moderator without losing intensity through small angular divergence. This small area for the end of the beam tube would also make it easier to construct choppers to eliminate delayed

neutrons and shape the time pulse. The availability of a cold source would be a great advantage for these experiments because the cold neutrons are essential for the study of large lattice spacings if small scattering angles cannot be used and these might not be available in a very high pressure experiment. If we were to use a cold source we would envision adding a spectrum shaping chopper midway between the source and the sample to eliminate frame overlap. This chopper would run at the same frequency as ZING but would have an adjustable phase and a 50% duty cycle. It could then select the appropriate range of wavelengths for the particular experiment. The proposed set up is shown in Fig. 4.1. The beam tube is kept under vacuum or filled with helium gas to avoid attenuation of the beam.

Finally we might discuss the need for this improvement in intensity and resolution. At present it takes about two days at the CP-5 High Pressure Neutron Diffraction Facility to obtain adequate counting statistics on samples which have good scattering cross sections and comparable absorption cross sections. Many experiments are not attempted because of the long count times which would be involved. (At pressures about 100 kbar, the useful sample volume is one-hundredth of that at 40 kbar). Others are just not possible because of the low signal to noise ratio. Sample conditions also limit the count times, especially at very high temperature, so the higher intensity would allow us to reach more extreme sample conditions. The present resolution is adequate for the large lattice spacings but resolution is lost rapidly in going to smaller lattice spacings. Greater resolution in the small lattice spacing region would make many more experiments possible. At present one is limited to working with relatively simple samples and sample conditions must be maintained constant for the length of the run--perhaps two days or more. These are serious limitations which would be eliminated with a source such as ZING.

Fig. 4.1. Proposed High Pressure Neutron Diffraction Experiment at ZING (without Neutron Guides).

5. Radiation EffectsParticipants

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I. Introduction

Radiation effects is the field of study of the changes in properties of solids induced by the bombardment with energetic nucleons. The special case of neutron bombardment is obviously of special interest. Unfortunately, neutron sources with high flux and the good instrumentation required for fundamental studies are in short supply. Indeed there is some danger that in the foreseeable future most, if not all, the research reactors will be phased out.

It should be noted that irradiation studies with other types of energetic nucleons have been widely performed and have been very useful. For example, low energy electron bombardments have been the main source of information for displacement energies and for the properties of single vacancies and single interstitials. On the other hand, simulation of high dose neutron bombardment has been performed by heavy ion bombardment. This technique provides very high displacement rates making it possible to simulate in hours some of the radiation effects obtained in years of bombardment in the most intense neutron sources. Neither of these techniques, however, duplicates neutron irradiation. Firstly, charged particle irradiations cannot generally yield the concentration of damage in displacement cascades as produced by fission neutron irradiations. Interactions between radiation induced defects are of prime importance in determining the defect state resulting from the irradiation and these interactions are highly sensitive to the local defect concentrations. Secondly, in addition to displaced atoms, neutron irradiation produces impurity atoms and noble gases by nuclear reactions. The interaction of mobile lattice defects with such reaction products is extremely important in many considerations as, for example, void formation. Such synergistic effects are almost impossible to simulate by other techniques. Thirdly, charged bombarding particles have limited range in solids; only with neutrons can radiation damage in structural members be

studied. Fourthly, there is increasing evidence, both experimental and theoretical, that primary lattice defects produced by high energy displacements have different properties from those produced by near threshold displacements so that electron irradiations do not even qualitatively simulate neutron irradiations. It is therefore necessary to continue radiation effect studies with neutrons.

II. Neutron Spallation Sources

Sources of neutrons for irradiations have mostly been limited to nuclear reactors; however, Livermore currently has a source of neutrons produced by the D-T reaction. This machine is specifically designed to produce 14 MeV neutrons which are of interest to the thermonuclear program. The neutron flux is relatively low, 5×10^{11} neutrons/cm² sec. Machines of higher intensity are currently being planned.

Machines producing neutrons by spallation from bombardment by energetic protons have been under consideration for some time. Los Alamos LAMPF and Brookhaven's BLIP are the only facilities currently being constructed. At LAMPF it is anticipated that a flux of the order 10^{13} n/cm² sec will be produced at the beam stop when the machine becomes fully operational. The neutron spectrum from these machines is largely determined by the target and the energy of the bombarding particle. In general (for bombarding energies higher than 100 MeV) the spectrum will consist of evaporation and spallation neutrons.

The spallation neutrons are a small portion of the total yield and are of high energy. From 2 MeV to near cutoff at the bombarding energy the number of neutrons with a given energy is roughly proportional to the inverse square of that energy. The evaporation neutrons have a distribution very similar to that of a fission neutron spectrum, i.e., an average energy of about 2 MeV. The spectrum can be tailored to some degree by target selection as the fraction of

evaporation neutrons increases with the atomic number, and by specimen placement since the high energy neutrons are peaked in the direction of the bombarding particles. About 10% of the neutrons will be at energies above the fission spectrum for a light metal beam stop such as graphite whereas only 3% will be above the fission spectrum for heavy metal beam stops. In summary, the neutron spectrum from spallation machines is basically a fission neutron spectrum with a high energy tail.

The spallation machines have a large advantage over nuclear reactors in regard to sample heating as the ratio of gammas to neutrons is reduced by almost two orders of magnitude.

Spallation machines are by nature pulsed. In the cases currently considered the number of displacements per pulse is marginal for the effects of a single pulse to be studied in metals. Radiation effects can be detected in semiconductors with greater sensitivity and these are more attractive candidates to utilize the pulsed feature; however existing burst reactors offer much larger neutron pulses than does ZING and most single experiments will probably be conducted in such machines. The ZING will generally be used for radiation effect studies as if it were a steady state reactor with large numbers of pulses integrated between measurements.

III. A Proposed Irradiation Effects Facility in ZING

ZING can be considered as a dual purpose machine i.e., as a source of neutrons for in situ radiation effect studies and for scattering experiments where pulsed neutron beams are to be used. These two requirements conflict in several respects. Perhaps the most important of these is the neutron spectrum. An intense flux of thermal neutrons is desired for scattering experiments whereas radiation effects experiments require a large flux of fast neutrons with minimal thermal neutron and gamma ray components. There is a strong bias toward scattering experiments in the original ZING proposal. A depleted uranium target is planned which doubles the neutron flux due

to fission but increases the gamma flux by a factor of 30. In addition, the moderators needed to thermalize the ZING neutron spectrum would increase nuclear heating in the vicinity of the beam stop to unacceptable levels for many radiation effects studies. The scattering studies require long uninterrupted operation while frequent periods of down time are needed for sample insertion, measurements, and sample retrieval in radiation effects studies. These conflicting requirements make a dual beam-stop facility necessary to make ZING useful to both groups.

A two target facility does not pose great technological difficulties. A bending magnet is required to switch from one source to another. A mode of operation with a minimum of a few hours operation at a target would probably be desirable. The fact that no beam facilities are required would greatly simplify shielding as the machine could be put in a pit and earth used for shielding. The target would be greatly simplified as tungsten would be used rather than uranium. The selection of tungsten as a target material is determined by its density, its high thermal conductivity, its elastic isotropy, and its high melting point. The former property is needed to produce a high neutron current without fission while the latter three are important factors in handling the energy deposited in the target.

By careful target design it is anticipated that a flux up to 10^{14} neutrons/cm² sec may be obtained at the target surface. This design includes reducing the target size to a diameter of 3" and using a tungsten reflector. It is clear that much more detailed study of this second target must be made in order to verify this flux estimate. Transport calculations of the proton beam and neutron flux are clearly needed as well as detailed calculations of the neutron energy distribution and target cooling. Nevertheless this seems to be a very promising machine for neutron irradiation studies.

IV. Uses of the Irradiation Facility

As previously outlined we should expect a flux of 10^{14} neutrons/cm²sec. The neutron spectrum is basically a neutron fission spectrum with a high energy tail and the nuclear heating is considerably lower than in the usual neutron sources. What kind of experiments can this machine be used for?

The low gamma ray flux makes the machine ideal for those experiments which require closely controlled temperatures. The influence of neutrons on the creep of materials has never been adequately studied primarily because of the difficulty of maintaining temperature control. It would be relatively easy to conduct creep experiments in this machine and it would probably represent the best available conditions for these studies. Low temperature experiments would fall in the same category. The neutron flux is more than an order of magnitude greater than any existing facility. It is clear that low temperature experiments in a steady state reactor with this flux would be difficult if not impossible. It should be possible to examine the effects of irradiation on superconductors including the measurement of radiation effects on properties of superconducting coils. In addition, there are many basic problems which the enhanced flux could solve. Studies of voids could also well be made.

The high energy tail in the neutron spectrum offers the possibility with the proper material selection to study the simultaneous effects of displacement cascades and helium production by (n, α) reactions.

Perhaps the most important aspect of this machine is that it will be a dedicated radiation effects machine. It will thus be possible to run the machine to conform to the needs of the radiation effects experimentalist. This advantage is very adequately illustrated by the situation at the beam stop area of the LAMPF. If this machine were adopted for

irradiation effects studies it would be an ideal machine. As it now stands, the requirements of other users makes difficult the tailoring of the target to radiation effects studies with the result that the flux will be only 1/10 that of ZING and 1/100 of that obtainable with optimum target design at LAMPF.

In conclusion, the building of the ZING radiation effects facility as outlined above would result in new and exciting results in radiation effects research.

6. Prospects and ProblemsParticipants

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I. Introduction

Even though the general layout of the proposed ZING facility was suggested in Ref. 1, it seemed important to discuss the detailed design of all the components of the facility and the performance required to meet the stringent demands of the other five working groups.

We have found ourselves able to meet these requirements, at least to a first approximation. The shield thickness, for example, of about five meters of heavy material, which is required from the biological as well as from the counter background point of view, does not conflict with the required flight path lengths for various spectrometers. We can also provide enough neutron beam holes, with very clean beams, with very few background neutrons between pulses, and also with wide flexibility as regards neutron spectrum. It seems that the final limitations of the facility will come only from the absolute intensity of the proton beam available.

The mode of beam sharing between the neutron scattering facility and the radiation effects research facility was discussed. One means is by frequent switching of the proton beam from one spallation target area configured as a slow neutron source, to another designed for radiation effects studies. Another means is to switch on a long term basis for periods up to one-third year per year. This would make possible a mode of interchanging target assemblies. Further investigation, however, is required before selecting the most convenient and desirable switching mechanism.

II. Targets and Moderators

Because of the crowding of the beam tubes, the proton beam must enter the target directed vertically downward. Directing the beam upward will produce "skyshine".

It is suggested that a neutron scattering target assembly will be replaced by a radiation effects target assembly for

about one-third of the year. In this way, both parties will get the maximum flux. This may require, however, some sophisticated switching mechanism. The problem may be alleviated by putting the radiation effects target below the beam tubes and by making the neutron scattering target assembly in small pieces to fit in small coffins.

For the neutron target, we should consider depleted U-Zr alloy clad in Zr, because of the formation of a stainless steel-uranium eutetic at $650^{\circ} - 700^{\circ}\text{C}$. Moreover, uranium metal will suffer from anisotropic growth in its α phase.

There may be some restrictions imposed by safety considerations of the interaction of water and sodium coolants for the background rotors and the target. The background rotors to cut off the unusable slow neutron tails will be sited about two meters from the target and will have about 1 kw heat production for each rotor.

As we use a U^{238} target, some 30% of the neutrons are fission neutrons, of which 1.5% will be delayed. Thus the total delayed neutron fraction will be about 0.45% averaged over the entire period. We can safely neglect the accumulation of delayed neutrons due to the small reactivity of the system. This delayed neutron background level is a very small value compared with the case of pulsed reactor in which over 30% is inevitable.

Using a background rotor about two meters from the target, we can expect a signal to background ratio of several thousand or greater, assuming a flight path to the measuring point of 10 m and making reasonable assumptions concerning the range of energies accepted.

III. Beam Tubes

The beam tubes should not look at the neutron target. This results in an "ear-like" moderator arrangement. Three moderators may give the best performance, as shown very schematically in Fig. 6.1. The beam tubes should terminate at

the inner wall so that beam tubes do not intersect.

Beam tubes, in general, should not be inclined at large angles with respect to the normal to the surfaces of the moderators. Considering the flight path variation across the inclined source and ignoring possibilities of focussing, then we find that independent of neutron energies, an angle no greater than 8° will not hurt the resolution. It is thought that four beam tubes will "look at" each moderator "ear". Their axes will diverge at an angle of 15° . (Each axis 7.5° with respect to the normal to the moderator surface.) Hence, a beam tube will not look at structure; it must see only the opposite beam tube. (Each pair of beam tubes can be regarded as a "through tube" with a moderator in its center.) Figure 6.1 shows schematically a possible arrangement, drawn assuming bare (unreflected) moderators.

Fig. 6.1. Schematic illustration of source and beam-tube arrangement.

There should be two vertical tubes intersecting each beam tube. One as close to the target as possible, and the other just within the shield. Figure 6.2 shows a possible arrangement.

The beam port windows might possibly be composed of incoherently-scattering material to maintain a smoothly varying spectrum.

Fig. 6.2. Arrangement of beam-tubes and vertical tubes.

IV. Reflected Moderator Beam Current

For the reference moderator, 2" CH_2 reflected by beryllium, the mock-up measurements of ANL-SSS-72-1 provide the ratio of the time averaged total thermal neutron leakage beam current per unit solid angle, per unit time average source intensity

$$\frac{I_{Th}}{\bar{S}} = 4.1 \times 10^{-3} \quad n_{Th}/ster/n_f \text{ produced} \quad .$$

Since for ZING the time averaged source intensity is

$$\bar{S}_{ZING} = 5.4 \times 10^{15} \quad n_f/sec \quad ,$$

the ZING thermal neutron beam current is

$$\begin{aligned} \bar{I}_{Th} &= \left(\frac{I_{Th}}{\bar{S}} \right) \bar{S}_{ZING} = 4.1 \times 10^{-3} \times 5.4 \times 10^{15} = \\ &2.2 \times 10^{13} \quad n_{Th}/ster/sec \quad . \end{aligned}$$

The thermal neutron beam current per pulse is then

$$\begin{aligned} I_{Th} &= \frac{1}{f} \bar{I}_{Th} = \frac{2.2 \times 10^{13}}{.9 \times 60} \\ &= 4.1 \times 10^{11} \quad n_{Th}/ster/pulse . \end{aligned}$$

The energy dependence of the slow neutron beam current is expected to be independent of the presence of the reflector at least up to several eV. Since the moderator-reflector interface is covered with Cd, the moderator appears unreflected to thermal neutrons. Since the effect of the reflector is mainly to reduce high energy neutron leakage, its effect upon the spectrum in the narrow interval from 0.5 eV to several eV is small. Therefore, the shape of the energy spectrum is nearly independent of the presence of the reflector, for energies below a few eV, and can be taken to be the same shape

as for a bare 2" CH₂ moderator. Consequently, the energy dependent beam current is

$$I(E) = \left(\frac{J(E)}{J_{Th}} \right) I_{Th}$$

where $J(E)$ is the measured current for a bare moderator, with normalization J_{Th} .

The shape of the spectrum might be taken, for example, from the work of Graham except that that work included no spectrum measurement for a 2" CH₂ slab. Taking the measured spectrum 1-1/2" thickness as a realistic approximation (curve a in Figure A-IV of ANL-SSS-72-1) and the total thermal current J_{Th} from Table IV of Graham and Carpenter, N.S. & E., 49, 418 (1972), one obtains, on a per-pulse basis,

$$\begin{aligned} I(E) &= J(E) \frac{I_{Th}}{J_{Th}} \\ &= J(E) \times \frac{4.1 \times 10^{11}}{2.44 \times 10^{-3}} \\ &= 1.68 \times 10^{14} J(E) \text{ n}_{Th}/\text{eV-ster/pulse} \end{aligned}$$

This function is plotted below as figure 6.3.

V. Pulse Widths for Be-Reflected CH₂ Moderator

The pulse width for a Be-reflected moderator is the result of the pulse width for a bare moderator, with the width of the distribution of epithermal neutrons "set" into the moderator by the beryllium reflector. This latter has been taken as the mean slowing-down time of neutrons in the beryllium reflector calculated below. A less conservative, possibly more realistic estimate of the broadening due to the reflector might be the FWHM but the more conservative approach

Fig. 6.3. Energy spectrum emerging from a 2" thick polyethylene slab embedded in a beryllium reflector.

is adopted here. The bare (unreflected) moderator pulse widths were taken from the data of Graham, extrapolated to 2" overall thickness as in Figure 6.4.

For the reference moderator 2" thick, 4" x 4" CH₂ moderator, reflected by beryllium, the pulse widths were calculated by the following recipe:

$$\Delta t_{FWHM} = \sqrt{(\Delta t(E)_{Bare})^2 + \tau_{Reflector}^2}$$

τ_{Ref}

is to be evaluated at energy E or at $E_{Cd} = 0.5$ eV, whichever is greater.

Fig. 6.4. Pulse FWHM vs thickness for neutrons of various energies leaking from $10.2 \times 10.2 \times t$ cm³ blocks of 300°K polyethylene.

The resulting pulse widths will thus be over-estimated and a very pessimistic approach is represented by these data. The bare moderator pulse widths serve as a lower limit on the pulse width. Pulse widths calculated from this recipe are shown in Table 6.1 and Fig. 6.5.

Table 6.1 Pulse Width vs Energy for the ZING Reference Moderator.

E_0 eV	v_0 m/sec	λ_0 Å	Δt μ sec	Δt_{bare}	$t_{\text{Ref1.}}$
2.1	2.00×10^4	.20	5.6	1.27	5.5
1.0	1.38×10^4	.29	8.2	1.80	8.0
.5	9.78×10^3	.41	11.3	2.55	11.
.1	4374	.92	18.	14.	11.
.05	3093	1.29	31.	29.	11.
.02	1956	2.05	45.	44.	11.
.01	1383	2.89	49.	48.	11.

Fig. 6.5. Pulse FWHM vs energy of neutrons emerging from 4" x 4" x 2" CH₂ moderator, reflected by Be, decoupled by cadmium.

VI. Beam Current from a Bare CH Moderator

Here, the calculation of ANL-SSS-72-1 is essentially repeated to obtain estimates of minimum realizable beam intensities.

$$\text{Subtended area} = 10 \times 7.65 = 76.5 \text{ cm}^2$$

$$\text{Fraction of source subtended} = \frac{76.5}{4 (7.5)^2} = 0.108$$

$$\text{Neutrons in source} = 6 \times 10^{15} \times 0.90 = 5.4 \times 10^{15} \text{ n/sec.}$$

$$\text{Neutrons into moderator} = 5.4 \times 1.08 \times 10^{-1} \times 10^{15} = 5.84 \times 10^{14} \text{ n/sec.}$$

$$\begin{aligned} \text{Neutrons into moderator/pulse} &= \frac{5.84 \times 10^{14} \text{ n/sec.}}{54 \text{ pulses/sec.}} = \\ &1.08 \times 10^{13} \text{ n/pulse} \end{aligned}$$

Thus, multiplying the data of Figure A-IV of ANL-SSS-72-1, by 1.08×10^{13} n/pulse, one obtains the realizable beam current

per pulse for the moderators tabulated there. The resulting beam currents are shown in Figure 6.6 for several choices of unreflected moderators. Multiply the number read from the scale, by the multiplicative factor on the curve, to obtain the beam current for each case. This is a minimum current

Fig. 6.6. Energy spectra emerging from a bare polyethylene moderator of lateral dimensions 10 cm x 10 cm.

estimate, since the data represent 14 MeV incident neutron energy, while the spallation source neutrons (of lower average energy) have greater collision probability. Also, recent LASL calculations indicate that substantial factors of improvement (2.7) can be obtained by using TiH_2 moderators, for example. Pulse widths for these moderators are shown in Figures 6.7 and 6.8. The pulse widths have been extrapolated toward higher energies, to approach the value $\text{FWHM} = 1.8/\sqrt{E_{\text{ev}}}$ which is approached in large CH_2 moderators

Fig. 6.7. Measured FWHM vs energy for five 296°K polyethylene moderators of 10 cm x 10 cm lateral dimensions.

above about 1 eV.

VII. Beam Currents from Bare Cryogenic Moderators

Proceeding exactly similarly, the beam current emerging from bare, 4" x 4" x 2.9", cryogenic moderators, solid methane (CH_4) and solid ammonia (NH_3) at 77°K, has been estimated. The resulting beam currents are shown in Figure 6.9. Pulse widths for these moderators are shown in Figure 6.10, taken from the work of Graham, and from the work of Fluharty. There is seen to be a substantial intensity advantage for neutrons of energy less than .020 eV, while the pulse widths are generally substantially narrower, when the cryogenic moderators are compared with those at room temperature.

Fig. 6.8. Measured FWHM vs energy for 296°K polyethylene moderators of 10 cm x 10 cm lateral dimensions. Measurements by Fluharty et al also shown.

Fig. 6.9. Energy spectra emerging from 77°K moderators 7.3 cm x 10 cm x 10 cm.

Fig. 6.10. Measured FWHM vs energy for bare, 77°K solid methane (CH₄) and solid ammonia (NH₃) moderators.

EXTERNAL REPORTS

1. Pulsed Neutron Radiography

by

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Neutron radiography using a short burst source of neutrons has already been successfully demonstrated.¹ In these experiments a fast burst facility produced enough thermal and epithermal neutrons in a single 40 msec burst to permit gated photomultiplier imaging of the object. The purpose of gating the detector is to select neutron energies that will reveal selectively one or another material in an object to be radiographed.

A flight path of 6-7 meters permitted sufficient energy resolution of the neutron beam so that neutrons utilizing the 4.9 eV resonance of gold, the 1.1 eV and 2.38 eV resonances of hafnium, and the 0.45 eV resonances of erbium could be selected by the gated electronic imaging system. Although the number of neutrons per burst will be greatly reduced for ZING compared to a fast burst assembly, the repeated pulsing of ZING (30 or 60 cycles/sec) should permit useful experimentation with a similar gated electronic imaging system.

2. A Neutron-Neutron Scattering Experiment

A series of detailed calculations (as yet unpublished) has been performed for the n-n scattering experiment. The goal of this experiment is to measure with high accuracy the scattering length (or scattering cross section) for the scattering of thermal neutrons by thermal neutrons. In general, acceptable ratios of signal to noise are obtainable for this

experiment with a steady state thermal neutron flux of 10^{15} nv or with a pulsed thermal neutron fluence of $> 10^{18}$ nvt.

For the ZING facility, one predicts 10^{14} nvt per burst in a time 50 μ sec, short compared to the neutron lifetime. For a pulsed annular core TRIGA, one can obtain 6×10^{18} nvt from a 100 Mw-sec pulse in a time of the order 10 millisecc, long compared to the mean life of a thermal neutron. Comparing these two neutron sources on the same time scale, we find that the TRIGA produces for a comparable pulse

$$\frac{.05}{10} \times 6 \times 10^{18} = 3 \times 10^{16} \text{ nvt} .$$

This is approximately 300 times greater than for ZING. Accordingly, the scattering rate in a burst would vary as the square of this ratio and would be 9×10^4 greater for TRIGA than ZING. Pulsing ZING at 60 cycles/sec for a 24-hour day would produce the same signal as a single pulse of the TRIGA reactor; however, ZING would produce much more background. Since the reactor can be pulsed 10 times per hour, it appears that ZING is a poor choice by comparison as a neutron source for the n-n scattering experiment.

The geometry at ZING as planned for the TOF experiments would completely defeat the needs of the n-n experiment. Far too few of the 10^{14} neutrons per pulse would be utilized. There is not enough moderator available for insertion of the large through-hole required for the n-n experiment. Further, the need for a very symmetrical source current of thermal neutrons around the through-hole would also be difficult to obtain with fast neutrons produced in a single proton target. The latter objection could be removed by a completely redesigned moderator-source assembly. However, one concludes that both the intensity per burst and the geometry of ZING make it unsuitable for the n-n scattering experiment.

¹"A Technique for Obtaining Neutron Radiographs in the Resonance Region", by L. Forman et al., Rev. Sci. Inst., 41, Dec 1970 (1900-1901).