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1. INTRODUCTION

This quarterly progress report describes the work performed under Project Agreement No. 2 of Contract AT(04-3)-167 for the United States Atomic Energy Commission during the quarter ending March 31, 1966.

During this reporting period, the possibility of computing scattering models from first principles has been considered. This approach looks promising for describing the physics of the model in more detail than heretofore. More work has been done in calculating a phonon spectrum for BeO and it is expected that a scattering kernel for this moderator will be generated very soon. This will be of use in the analysis of thermal neutron spectrum measurements which were made during this quarter in beryllium oxide at elevated temperatures.

Further evaluation has been made of the scattering law data generated at the Brookhaven National Laboratory during last summer. The evaluation of the principal hydrogenous moderators is summarized in Appendix A. This evaluation has been aided considerably by the use of an automatic plotting routine, which has indicated some errors of a numerical nature in the scattering law code GASKET.⁽¹⁾ These mistakes have been rectified and the evaluated data have been added to the ENDF file. A list of evaluated $S(\alpha, \beta)$ decks submitted to ENDF is included in this report. Coupled to this work is the continued evaluation of our spectral data and presentation in the Spectrum Book (GA-5319) and in the EACRP version of this publication. In addition, a compilation documenting the scattering law data has been initiated. A writeup of the preliminary version of the code FLANGE,⁽¹⁾ which computes scattering kernels from the $S(\alpha, \beta)$

data is also being prepared for submission to the Argonne code center. These latter two items should facilitate the use of the ENDF system.

The ease with which a scattering kernel can be generated at any desired energy mesh from the scattering law data has prompted us to compile the results from other scattering models in that format. Heavy water and benzene are two particular moderators being so prepared, both in the incoherent approximation. The heavy water scattering kernel is needed to analyze heterogeneous spectral data presented previously.⁽¹⁾ The benzene scattering kernel is needed to analyze the neutron spectrum in diphenyl, which was measured as a function of poison concentration during this quarter. These scattering laws will also be most useful for an organic - D₂O interface experiment which is planned for the next quarter.

The improvement in scattering kernels over the years has led us to consider means of checking their validity other than by neutron spectra. Such checks include double differential scattering, angular scattering, and total cross sections. In the first of these categories we have been comparing the multiple scattering corrections to the measurements of other research groups. Our aim is to make the observed double differential cross section, the scattering kernel, and the multiple scattering correction all self consistent. In the second of these categories, we have evaluated our previous measurements⁽²⁾ of the angular scattering by zirconium hydride at elevated temperatures. The anomalies in the observed data are now believed to be related to that material's chemical instability above 300°C. Plans are underway to repeat the measurements under an inert atmosphere and with an improved angular resolution. In the third category, that of total cross sections, it has become evident that our present theoretical scattering models predict far more structure than is observed in such references as BNL 325. Since our facilities can give greatly improved total cross section detail over BNL 325, some

preliminary studies have been made on the common moderators. It is hoped that these measurements will give improved accuracy below the Bragg cutoff where our incoherent inelastic scattering models are not applicable.

The evaluation of the spectral data measured previously on a fast U^{235} metal ball reactor is continuing. The major problem in this evaluation has been in determining the efficiency of the two detectors involved. The efficiency of the intermediate energy detector has been computed elsewhere by the Monte Carlo methods and the final data measured in that energy range are presented in this report. Consequently emphasis has shifted from the measurements to the theory in which some calculational difficulties still remain. The research is described both in Section 3.8 and in Appendix C. Plans have been made to study the fast neutron spectrum as a function of position in a 20-in. diameter ball of depleted uranium.

Efforts have been made in this quarter to improve our data reduction methods, specifically the code ECTO for spectral data, and the code REDUCE⁽²⁾ for angular scattering data. The efficiency of the 16 m detector bank, which has direct bearing on the measurement of the thermal neutron spectrum, has been redetermined. A new efficiency curve has been obtained which shows improvements outside the normal range of our theory-experiment comparisons. This new curve covers a wider energy range than before and will be useful in our proposed intermediate energy spectrum measurements. Finally, the construction of the 200 m flight path and the new fast reactor spectrum cell have commenced.

The following papers derived from research performed under this contract were presented or prepared during this quarter for various scientific meetings.

American Nuclear Society Meeting in San Diego, February 1966

1. James A. Young, "Atomic Motion in Moderators"
2. J. R. Beyster, J. M. Neill and J. C. Young, "Recent Developments in Integral Neutron Thermalization."
3. J. U. Koppel, "Neutron Scattering by Hydrogeneous Moderators."

Conference on Nuclear Cross Sections in Washington, March 1966

1. J. R. Beyster and J. M. Neill, "The Influence of Neutron Cross Sections on Thermal Reactor Spectra."
2. J. L. Russell and A. E. Profio, "Adequacy of Fast and Intermediate Cross Section Data from Measurement of Neutron Spectra in Bulk Media."

American Nuclear Society Meeting in Denver, June 1966

1. A. E. Profio, H. M. Antunez, K. L. Crosbie, and J. L. Russell, "Time-of-Flight Measurement of the Neutron Spectrum from a Spherical U^{235} Fast Reactor."
2. J. M. Neill, J. R. Beyster, and W. M. Brouwer, "Thermal Neutron Spectra in Beryllium at Elevated Temperatures."

2. THEORETICAL STUDIES

2.1 NEUTRON SCATTERING PHYSICS

A scattering kernel presently exists for each of the reactor moderators, although the kernel for BeO is just now being evaluated. Most of the scattering models are relatively simple yet generally yield good agreement with experimental neutron spectra at room temperature and above. Nevertheless, for low temperatures and for small assemblies the current scattering kernels are bound to show serious inadequacies. One reason for this is the use of the incoherent approximation, which is widely used for the solid moderators. This approximation can be eliminated however, and previous work on this program⁽¹⁾ has calculated coherent effects in beryllium. The most serious problem, however, is to get more physics into the scattering model.

In the cases of BeO and even Be the present physical models are definitely not adequate to accurately describe the interaction of neutrons with the lattice, but it is possible to improve this situation. Since solids can be treated well in the harmonic approximation, it is possible to solve the many-body problem of the electrons and ions moving in their mutual coulomb fields. This calculation is being done for beryllium. The method consists in separating the nuclear and electronic motions by means of the Born-Oppenheimer adiabatic approximation to obtain an expression for the electronic contribution to the energy. At this point all quantities are expanded to second order in the nuclear coordinates (harmonic approximation), and the energy is calculated by perturbation theory. The Coulomb energy is evaluated by means of the Ewald technique as in our calculations

on BeO. The lattice dynamics can be obtained at this point in the standard way. While it is premature to include a detailed writeup of this work at this time, calculations are proceeding and the method looks promising indeed.

The above method of using first principles to compute scattering models is desirable since the neutron scattering experiments are so susceptible to multiple scattering that interpretation is very difficult.

2.2 SCATTERING BY BERYLLIUM OXIDE

During this reporting period the work done on BeO has been mainly numerical. The phonon spectrum has been calculated and is shown in Fig. 2.1. As explained in previous reports,^(1,2) the phonon spectrum alone is not sufficient to evaluate the neutron scattering cross section for a polyatomic moderator. The sums necessary over polarization vectors have been calculated and there remains only to run the machine code GASKET⁽¹⁾ to obtain the scattering kernel. In fact, sample curves of $S(\alpha, \beta)$, the scattering laws, have been calculated at particular α points to determine if everything appears to be correct. Aside from a few small changes found to be necessary in GASKET, after examining plots of the material being sent to Evaluated Nuclear Data File (ENDF), it seems a kernel can now be generated. Actually, four kernels must be obtained, one for each atom in the unit cell of the BeO lattice. A comparison of theoretical and experimental neutron spectra will be done very soon to evaluate the scattering model used for BeO.

2.3 MULTIPLE SCATTERING IN H₂O

The double-differential cross section for thermal and epithermal neutron scattering in water is important in the analysis of water-moderated

Fig. 2.1--Phonon spectrum of BeO

reactors. A substantial amount of experimental data and several theoretical studies concerning the water scattering kernel are available.⁽³⁾ However, the problem of selecting an appropriate kernel from this material is complicated by the following considerations:

1. No theoretical model shows better than qualitative agreement with double differential measurements.
2. No two theoretical models show better than qualitative agreement with each other.
3. Results of different experiments are frequently in disagreement.
4. Results of the same experiment are sometimes inconsistent, a fact brought to light when data is plotted in scattering law form.

A step toward reducing some of this confusion can be taken by considering the corrections for multiple scattering which should be applied to the data. It is to be hoped that such corrections will improve the internal consistency of results from individual experiments, resolve discrepancies between different experiments, and make theoretical comparisons more meaningful.

As a start, we have calculated some multiple scattering corrections for the double-differential H_2O experiment of Kirouac, Moore, and Seemann⁽⁴⁾ at RPI. Calculations were done with the program MUSE,⁽⁵⁾ a code which computes multiple scattering corrections for up to four collisions from transport equations. In the RPI experiment a neutron beam is incident at 45 degrees on a water sample 0.0205 in. thick held between plane parallel aluminum plates. Outgoing neutrons are observed at three angles corresponding to scattering angles of 25° , 60° , and 90° . MUSE corrections have been computed for this setup for incident energies of 0.153 eV and 0.232 eV, using five space points. The theoretical kernel input to MUSE was taken from GASKET results for water⁽³⁾ (based on a

frequency distribution obtained by Haywood⁽⁶⁾ with extrapolation techniques) with an added oxygen free gas kernel. A mesh of 30 energies and 21 angles was employed.

Figures 2.2 through 2.7 show the resulting correction factor, C_H , as a function of outgoing neutron energy. The effect of finite sample thickness on experimental double-differential cross sections is corrected for completely by multiplying by C_H . Since the RPI results have already been partly corrected for attenuation in the sample,⁽⁴⁾ this attenuation correction (a function only of incident energy) must be removed before correcting the data with the factor C_H . Also the correction factor, in principle, applies only to data which have been corrected for resolution broadening, whereas the RPI data are uncorrected. Moreover, the RPI data were taken relative to a vanadium standard cross section. The effects of multiple scattering in the vanadium target on the water cross sections are, of course, not included in the correction factors of Figs. 2.2 through 2.7. (From a Vineyard-type⁽⁷⁾ analysis of multiple scattering in vanadium it appears likely that these corrections will affect significantly only the scale of the RPI data and not the relative position of the data points.) For these reasons we are not, at present, exhibiting curves of corrected double-differential data.

The size and fluctuation of the correction factors shown in the figures confirms that multiple scattering effects cannot be neglected in double-differential experiments, particularly since the RPI experiments were done at a relatively high incident energy. Preliminary calculations of corrections for experiments of lower incident energies show even greater multiple scattering effects, notwithstanding the use of thinner targets. Appropriate to the consideration of target thickness is the dotted curve in Figs. 2.2 through 2.4. This curve shows the correction factor calculated for the same experimental arrangement as for a target half as thick

Fig. 2.2--Correction factors to observed $\frac{d^2\sigma}{dE d\Omega}$ at $E_o = 0.153$ eV, $\theta = 25^\circ$

Fig. 2.3--Correction factors to observed $\frac{d^2\sigma}{dE d\Omega}$ at $E_o = 0.153$ eV, $\theta = 60^\circ$

Fig. 2.4--Correction factors to observed $\frac{d^2\sigma}{dE d\Omega}$ at $E_o = 0.153$ eV, $\theta = 90^\circ$

Fig. 2.5--Correction factors to observed $\frac{d^2\sigma}{dE d\Omega}$ at $E_0 = 0.232$ eV, $\theta = 25^\circ$

Fig. 2.6--Correction factors to observed $\frac{d^2\sigma}{dE d\Omega}$ at $E_0 = 0.232$ eV, $\theta = 60^\circ$

Fig. 2.7--Correction factors to observed $\frac{d^2\sigma}{dE d\Omega}$ at $E_0 = 0.232$ eV, $\theta = 90^\circ$

as that actually used in the RPI experiment. The fluctuation and magnitude of the correction factor remains sufficiently large to show that multiple scattering effects could not reasonably have been avoided in this experiment by using thinner targets.

Since the correction factors computed by MUSE⁽⁵⁾ depend on the input theoretical kernel, the correction factors themselves are theoretical quantities which must, in some sense, be compared to experiment. Kirouac⁽⁸⁾ has pointed out that when the RPI data are plotted in scattering law form the deviation of particular points from the general trend of the data may give an experimental estimate of the multiple scattering correction. Correction factors taken from the curves of Figs. 2.2 through 2.7 do, in fact, improve significantly the self consistency of the RPI data and further improve its agreement with GASKET predictions.⁽³⁾

Investigation of multiple scattering in the RPI experiment and in other water double differential measurements is continuing.

2.4 SCATTERING KERNEL FOR D₂O

Neutron spectra in D₂O have been calculated in the past using a scattering kernel which is an adaption to deuterium by Honeck⁽⁹⁾ of the Nelkin kernel⁽¹⁰⁾ originally formulated for hydrogen bound in H₂O. Actually the results obtained with this incoherent, discrete oscillator model were really excellent. Coherent effects due to both inter- and intramolecular interference were shown⁽¹¹⁾ to be unimportant in most practical spectral problems. Since the code GASKET⁽¹⁾ has become available, scattering kernels have been generated in the form of the two-parameter scattering law, $S(\alpha, \beta)$, from which it is easy to obtain, by means of the code FLANGE,⁽¹⁾ a complete P_n -kernel for any arbitrary energy mesh.

This convenience made it desirable to obtain the scattering law for D_2O in the GASKET format. The frequency spectrum used for this purpose was made up of the following contributions: (a) two discrete oscillators with frequencies of 0.31 and 0.145 eV and weights of 1/3 and 1/6 respectively, (b) a rotational frequency band peaking at about 0.05 eV obtained from Haywood's⁽⁶⁾ measured frequency spectrum (the weight assigned to this band was 9/20) and (c) a δ - line at $\omega = 0$ with weight $\frac{1}{20}$ (free translation of molecules of mass 20). Figure 2.8 shows the frequency spectrum measured by Haywood. The dotted lines indicate the rotational band used in GASKET.

2.5 KERNELS FOR THE EVALUATED NUCLEAR DATA FILE

A substantial effort is being expended to make available all our calculations to the ENDF at Brookhaven⁽¹²⁾ and to calculate additional kernels in a standard format and temperature range for the data file. Table 1 lists the scattering kernels which have been placed on the ENDF tapes at present. Work is continuing toward checking the GASKET⁽¹⁾ computations so that all the kernels will very soon be available on the ENDF tapes.

2.6 EVALUATION OF GASKET RESULTS

A survey of the scattering law calculations made during the past two quarters has been started; more scattering models than were of immediate interest were computed at Brookhaven National Laboratory for use with the BNL END File.⁽¹²⁾ Analysis of these scattering law calculations has been speeded up by a machine program (SPLOT) for automatic plotting.

The machine plots for each scattering law deck are of three types: (1) a log-log plot of $S(\alpha, \beta)$ versus α with 20 curves per plot covering most of the β range, (2) a series of semi-log plots of $S(\alpha, \beta)$ (linear)

Fig. 2.8--Frequency spectra for D_2O

Table 1

SCATTERING LAW DECKS AVAILABLE FOR END FILE

GA ID	Moderator	Temp ^o K	$\alpha \times \beta$	Description	
1	H (H ₂ O)	296	80x80	BNL-2 2 oscillators + Haywood $\rho(\omega)$	$E_{\max} = 2.0 \text{ eV}$
2		350	"	"	"
3		400	"	"	"
4		423	"	GA	"
5		450	"	BNL-2	"
6		500	"	"	"
7		600	"	"	"
10	H (poly)	296	80x80	BNL-3 2 oscillators + $\rho(\omega)$	"
12	H(ZrH90)	296	40x150	BNL-1 Mass 90 acoustical model	$E_{\max} = 1.0 \text{ eV}$
13		400	"	BNL-2	"
14		500	"	"	"
15		550	"	"	"
16		600	"	"	"
17		700	"	"	"
18		750	"	"	"
19		800	"	"	"
20	H(ZrH360)	296	"	" Mass 360 acoustical model	"
21		1273	"	NBS-2a	"
22		1473	"	"	"
24	Be	296	50x80	BNL-1 JAY frequency spectrum	"
29		400	"	"	"
30		500	"	"	"
31		600	"	"	"
32		700	"	"	"
42		1000	"	"	"
43		1200	"	"	"
33		800	"	"	"
34	C(graphite)	296	40x80	"	"
35		400	"	"	"
36		500	"	"	"
37		600	"	"	"
38		700	"	"	"
39		800	"	"	"
40		1000	"	"	"
41		1200	"	"	"

versus α (log) with three curves per plot, progressing through the β range, and (3) a similar series of $S(\alpha, \beta)$ versus β plots for a series of α values. Typical plots for water at room temperature, using the Haywood⁽⁶⁾ distributed frequency spectrum, are shown in Figs. 2.9, 2.10, and 2.11.

The systematic plotting (at room and at higher temperatures) of all the scattering law models calculated has revealed two errors in the calculations. The first was a program mistake in the treatment of the portion of the scattering law calculated in the short collision time approximation. For models with a distributed frequency spectrum and no discrete oscillators, the code gave results at the fixed α at which the switch from exact calculations was made instead of going to higher values of α . This error produced large effects only in beryllium. The second error was in the calculation of zirconium hydride, where the entire calculation was made without switching to the short collision approximation. The time mesh chosen for the integration which was checked for small and intermediate values of α proved to be inadequate at the very large α 's usually treated in short collision approximation. The results for the time mesh used would not have been bad but for a peculiarity of the integrands $Q(t)$ and $R(t)$ for ZrH. These functions show structure at small t , and then have alternating intervals of essentially zero behavior and damped structure. To save computing time, a test was included in the code to terminate the time integration at some time when the expected contribution from higher times becomes negligible. This test was satisfied, unfortunately, by the first stretch of zeros after the initial structure (Fig. 2.12) omitting the second structured region which seems to produce the typical oscillation of $S(\alpha, \beta)$ (see Fig. 2.13). The results from the integration of the first region only are very similar to the short collision time results. The switching to this behavior occurs at sufficiently large α to make the present results acceptable for reactor spectrum calculations; we will, however, recalculate both ZrH and Be for distribution on the END File.

Fig. 2.9--Computer log-log plot of $S(\alpha, \beta)$ against α at various β for H_2O using Haywoods Frequency Spectrum

Fig. 2.11--Computer plot of $S(\alpha, \beta)$ against β at various α for H_2O using Haywoods Frequency Spectrum

Fig. 2.12--Irregular behavior of $Q(t)$ and $R(t)$ for zirconium hydride

Fig. 2.13--Irregular behavior of $S(\alpha, \beta)$ for zirconium hydride at large α

2.7 SPECTRUM BOOK

During the past three months, 17 new cases have been completed for the Spectrum Book⁽¹³⁾ and have been forwarded to all holders of the book. Neutron spectra and other pertinent information are now presented in the new format as described in Ref. 1. A common reference list and a listing of the cross section data of the absorber nuclei are now also included in the data sheets.

The new cases, which also include spectra reported in Ref. 14, have been analyzed or reanalyzed using the most up to date neutron cross sections. An attempt has been made to include a realistic value of the leakage in all these calculations. The buckling (B^2) for each calculation was obtained from experimental neutron flux plots whenever available. A simple computer code was written to determine the energy-dependent diffusion coefficients ($D(E)$), which were used in the calculation of "infinite medium" neutron spectra with a negative buckling. The use of diffusion theory to describe the leakage is satisfactory where $DB^2 \ll \Sigma_a$. In fact, where DB^2/Σ_a is very small, the inclusion of a leakage term causes a negligible difference in the spectrum. However, in cases where DB^2 is not small relative to Σ_a , the diffusion theory approximation to the leakage does not apply. These cases are often unanalyzable and will not be presented.

Although the Spectrum Book does not contain all our experimental work on pure and poisoned water, it now represents a fairly complete range of temperatures and poison concentrations of this moderator, for resonance absorbers as well as $1/v$ absorbers. Consequently, emphasis will be shifted toward other moderators for additional cases in the future.

We have received layout sheets of the modified format of the EACRP publication and intend to submit in the next quarter our latest cases for inclusion in the European version of the Spectrum Book.

3. EXPERIMENTAL AND ANALYTICAL STUDIES

3.1 THERMAL NEUTRON SPECTRA IN BeO AT ELEVATED TEMPERATURE

Spectral measurements in beryllium oxide were made as a function of temperature. The BeO assembly and its geometrical arrangement were described previously.⁽²⁾ Figure 3.1 gives most of the pertinent details. The BeO was poisoned by borated stainless steel to about 0.5 barns/Be atom at 2200 m/sec. The results of these studies are shown in Fig.

3.2. No comparative analysis has yet been performed because the complete new scattering kernel for BeO has not yet been generated (see Section 2.2). The temperatures shown on the figure are average values and, in fact, there were large variations across the block amounting to 50°C for the 586°K case. Nevertheless, the spectral data exhibit a behavior that is consistent with the temperature indicated. The small flux peaking below the Bragg cutoff diminishes as the temperature is increased.

3.2 THERMAL NEUTRON SPECTRA IN DIPHENYL AT VARIOUS POISON CONCENTRATIONS

Interest in organic-cooled, D₂O-moderated reactors provides motivation for the study of neutron thermalization in the organic coolant. Typically the coolant may consist of a mixture of polyphenyls, principally diphenyl and terphenyl. It was decided to study the infinite medium neutron spectrum in diphenyl since that material has the simplest chemical structure and so presumably would allow a scattering model to be formulated with the least difficulty.

Fig. 3.1--Geometrical arrangement used for spectral measurements in BeO

Fig. 3.2--Thermal neutron spectra in BeO as a function of temperature

The geometry used for the spectral studies is illustrated in Fig. 3.3. Heating of the tank was necessary since diphenyl melts at about 60°C and thermalization in the liquid state was to be studied. Methyl borate was used as poison because it is a liquid and mixes uniformly into the molten diphenyl. In fact the methyl borate boils at a temperature lower than the melting point of the diphenyl, but the poison is believed to form a complex which is retained at the higher temperature. This was checked in the laboratory where retention of the poison in the molten diphenyl was demonstrated. A further check was made by repeating a poisoned spectrum measurement since there would have been a change of shape had much methyl borate been liberated. Identical results were obtained in both checks. Chemical analysis of each batch provided a further check, but these analyses have not yet been completed.

Neutron spectra were measured at about 75°C in the molten diphenyl at the following poison concentrations, natural $-(0.33 \text{ b/H})$, 1.30 b/H , 2.19 b/H , and 3.04 b/H , these values being at 2200 m/sec . The measured data are shown in Fig. 3.4. Flux plots were made at the beginning and end of these spectral studies in the axial direction of the tank. Cadmium covered indium foils and dysprosium-aluminum foils were utilized. The spatial dependence of the indium resonance flux changed little with poison concentration, not so the thermal flux as Fig. 3.5 illustrates. The slopes of the indium resonance and dysprosium-spatial activities at the measurement point are approximately the same for the 3.04 b/H case but are different for the pure diphenyl. This indicates that the buckling is energy dependent for the latter case so that there will be difficulties in its analysis.

No analysis of these data has yet been performed since no scattering model for diphenyl is available. Its formulation is quite formidable. It has been suggested that the scattering model used for hydrogen in benzene

Fig. 3.3--Geometrical arrangement used for spectral measurements in diphenyl

Fig. 3.4--Thermal neutron spectra in diphenyl as a function of poison concentration

Fig. 3.5--Axial flux plots in 10 in. x 10 in. x 10 in. tank of diphenyl

will be adequate for hydrogen in the higher polyphenyls since their chemical structure is similar, e.g.,

Benzene

Diphenyl

A scattering model for benzene has been developed by Boffi, et al.,⁽¹⁵⁾ using the computer code SUMMIT.⁽¹⁶⁾ This scattering kernel utilized the vibrational frequency distribution due to Crawford and Miller.⁽¹⁷⁾ Since we plan to use this scattering kernel later in a D_2O -organic interface experiment we would like to get the scattering described in the $S(\alpha, \beta)$ form; this allows us to vary the energy mesh of the kernel conveniently. We have also received a request for the scattering law for benzene. In addition, the present measurements were made at about $75^\circ C$ instead of at $296^\circ K$, at which temperature the present benzene kernel was generated. Consequently the scattering by benzene is being recomputed by means of the code GASKET.⁽¹⁾ The experiment-theory comparison should be completed during the next quarter.

3.3 THERMAL NEUTRON SPECTRA IN BERYLLIUM METAL AT ELEVATED TEMPERATURES

Neutron spectra have been measured at elevated temperatures in beryllium poisoned mildly with samarium to 0.10 barns/Be atom at 2200 m/sec. The measurements have been performed utilizing the standard time-of-flight techniques.⁽¹⁸⁾ The purpose of these studies was to check the bound scattering kernel for the prediction of thermal neutron spectra in beryllium at temperature. The measurements were expected to help answer the question that the crystal frequency spectrum should be changed when the scattering kernels are generated at higher temperature. In this case the frequency spectrum for beryllium is that due to Young.⁽¹⁹⁾

The beryllium was stacked into a cube of approximately 20 inches edge. The poisoning was introduced heterogeneously by means of samarium loaded foils. The assembly was heated by means of 16KW strip heaters. It was excited by an externally located source of photoneutrons. Cadmium covered indium foils were activated along the three axes of the assembly in order to determine the local buckling and obtain an epithermal source distribution for the comparative calculation. The neutron flux was extracted from a 1 in. by 1-7/16 in. reentrant hole extending into the center of the beryllium block. Measurements were made at room temperature (288°K), 452°K and 605°K and the results are compared to preliminary theoretical values in Fig. 3.6. Several conclusions may be drawn from this figure. The differences between free gas and bound model predictions are small at room temperature and negligible at 600°K. The difference between the bound and free atom calculation is largest at the low energies of the 288°K measurement. This suggests that the use of a bound kernel will be most important for beryllium at very low temperatures such as in a nuclear rocket reflector or in the design of cold neutron sources. However, accuracy there will involve the development of a scattering kernel in which the incoherent approximation is no longer used.

Fig. 3.6--Neutron spectra in beryllium as a function of temperature

At room temperature the agreement between theory (bound kernel) and experiment is reasonable. The agreement at higher temperature is not as satisfactory. The discrepancies are believed to arise from three sources:

1. Nonuniformities in the temperature distribution across the beryllium assembly.
2. Differences between the average metal temperature and the temperature at which the scattering kernel was generated.
3. Inadequate treatment of the leakage, a factor which becomes significant only because the poisoning is low.
($DB^2/\Sigma_a = 29\%$ at 2200 m/sec).

Plans have been initiated to make spectral measurements at liquid nitrogen temperature, which should test more severely our present scattering kernel for beryllium.

3.4 NEUTRON SPECTRUM IN H_2O

A measurement of the thermal neutron spectrum in pure water was made previously⁽¹⁾ in a 10 in. by 10 in. by 10 in. tank. The measurement was difficult to analyze because the tank was not large enough to provide a quasi infinite medium geometry at the point of measurement. Using the GATHER⁽²⁰⁾ code for the analysis would therefore be of questionable accuracy. Unfortunately the tank was too large to allow the use of our S_n transport theory code GAPLSN⁽²¹⁾ with proper spatial, angular, and energy representation. Since our previous studies⁽¹⁾ showed that the local buckling at the measurement point was energy dependent, our earlier analyses of the measurement were unsatisfactory. The spectrum has therefore been recalculated using the method employed by Parks⁽²²⁾ in predicting the neutron spectra in graphite.

In Park's method the flux per unit energy, $\phi(E, r)$ at the spatial point r in the axial dimension is a solution of

$$-D(E)\nabla^2\phi(E, r) + C\phi(E, r) = f(E) Q(r)$$

where

$$\begin{aligned} f(E) &= (E - \alpha E_0)/E & E \geq \alpha E_0 \\ &= 0 & E < \alpha E_0 \end{aligned}$$

and $Q(r)$ is the spatial distribution of the indium resonance flux in the axial dimension

$$\alpha = \left(\frac{M - 1}{M + 1} \right)^2$$

C is defined by

$$\begin{aligned} C\phi(E, r) &= \left[\Sigma_a(E) + \Sigma_s(E) + D(E) B_L^2 \right] \phi(E, r) \\ &\quad - \int_0^E \Sigma(E' - E) \phi(E', r) dE' \end{aligned}$$

where B_L^2 is the lateral buckling.

The method is to assume a solution of the form

$$\phi(E, r) = \phi_0 Q(r) + R(E, r)$$

where $\phi_0(E)$ is chosen to satisfy

$$D(E) B_0^2 \phi_0(E) + C \phi_0(E) = f(E)$$

with

$$B^2(r) = - \frac{\nabla^2 Q(r)}{Q(r)} = B_0^2 + \delta B^2(r)$$

B_o^2 is the value of $B^2(r)$ at the measurement position r_o .

The expansion below is made.

$$R(E, r) = R_o(E) \bar{Q}(r) + R_1(E) L^2 F(r)$$

where

$$\bar{Q}(r) = -L^2 \delta B^2(r) Q(r)$$

$$L^2 = \frac{\bar{D}}{\bar{\Sigma}}$$

F satisfies

$$-L^2 \nabla^2 F + F = \bar{Q}$$

$$R_o(E) = C \bar{\Sigma} \phi_o(E)$$

and

$$R_1(E) = C \frac{D(E)}{L^2} R_o(E)$$

We compute F by integrating over the tank length a

$$F(r) = \int_o^a G(r/r') \bar{Q}(r') dr'$$

where the actual Green's function is taken as that appropriate to an infinite medium

$$G(r/r') = \frac{1}{2L} e^{-(|r-r'|)/L}$$

This correction was applied to the calculation at the 17.8 cm position in the tank. In Fig. 3.7 the measured and calculated thermal neutron spectra in pure water are compared. The Parks method⁽²²⁾ gives an improved fit to the measured data. Nevertheless discrepancies as high as 30%

Fig. 3.7--Thermal neutron spectra in pure H_2O

remain between 0.08 and 0.25 eV. To improve the comparison it will be necessary either to repeat the measurement in a larger tank or use a transport theory code on a digital computer having a larger fast memory than our present IBM 7044 machine.

3.5 ANGULAR SCATTERING BY ZIRCONIUM HYDRIDE AT ELEVATED TEMPERATURES

Analysis of measurements of angular scattering of $\text{ZrH}_{1.85}$ at 750°K has shown anomalies not explained by theory. Theoretical values of $d\sigma/d\Omega$ have been calculated using our broadened Einstein oscillator model⁽²³⁾ and the codes GASKET⁽¹⁾ and FLANGE⁽¹⁾ at temperatures of 750°K and 296°K . Figures 3.8, 3.9 and 3.10 show these results at three different scattering angles. It can be seen that the greatest temperature dependence is at the largest scattering angles. The curves for the two temperatures nearly converge for the lowest scattering angles.

The theoretical calculations predict more nearly isotropic scattering at large scattering angles. However, analysis of the observed 750°K data shows more anisotropic scattering. The plot of measured $d\sigma/d\Omega$ against scattering angle was found to be more sharply peaked than the theoretical: the magnitude of the observed $d\sigma/d\Omega$ was less. The sample was analyzed and was found to have suffered a change in composition after being subjected to high temperature for long periods of time. The chemical analysis showed that a large fraction of the hydrogen had been driven off the zirconium hydride sample. This reduction in the amount of hydrogen in the sample would explain the observed drop in scattering cross section. Previous data taken at 525°K also indicated evidence of a loss of bound hydrogen. These data follow the same trends as the 750°K data and are also unusable.

Fig. 3.8--Angular scattering cross section of zirconium hydride at $\theta = 60^\circ$

Fig. 3.9--Angular scattering cross section of zirconium hydride at $\theta = 120^\circ$

Fig. 3.10--Angular scattering cross section of zirconium hydride at $\theta = 154^\circ$

Work is now in progress to construct a sealed sample holder. The dislocation pressure at 750°K is less than 5×10^{-2} atmospheres and a sealed sample holder should be possible without explosion. It can be seen from Figs. 3.8, 3.9 and 3.10, that if certain angles and energies are chosen the temperature effect should be observable. Since the model for zirconium hydride⁽²³⁾ is a phenomenological one only, the observed differences with temperature may exceed these calculated values (see, for example, Fig. 3.12). When a good reliable sample and sample holder are constructed, new measurements will be taken at large scattering angles and near the Einstein oscillator resonance points. These data should give results which can be used to study the model for $\text{ZrH}_{1.85}$.

It is anticipated that future angular scattering experiments will make use of the data reduction code REDUCE.⁽²⁾ This code has been checked against hand calculations of 13-mil H_2O data and has been found to be in good agreement.

3.6 TOTAL CROSS SECTION STUDIES

The total cross sections of the common moderators are important data to know accurately. They are useful for reasons other than knowing the attenuation of a neutron beam. These reasons are that σ_T provides a valuable integral check of the theoretical scattering kernel, σ_T at higher energies provides the free atom scattering cross section for the theoretical studies, and the measured σ_T is commonly used in double differential measurements for normalization purposes. The scattering kernels are presently generated with precision using such codes as GASKET⁽¹⁾ and FLANGE⁽¹⁾ to compute the inelastic incoherent scattering. The elastic coherent scattering may be added by means of codes such as HEXSCAT.⁽²⁴⁾ These codes consist of dynamical models which have been developed over the last few years and embody numerous refinements.

The state of the art appears to be such that within the limitations of the moderator models, the total cross section can be predicted above the Bragg cutoff to accuracies and with detail not afforded in measurements such as those in BNL 325. In addition the subBragg cutoff values of σ_T allow the determination of the magnitude of the coherent inelastic scattering cross section. These data are necessary to complete an understanding of the physics of slow neutron scattering and also to determine accurately the thermalization in cold moderators. These latter situations arise in practice in reflectors for a nuclear rocket reactor, and in cold neutron sources for research on systems having large periodic spacings, e.g., organic molecules.

Recently we have performed a few simple total cross section measurements in the geometrical arrangement shown in Fig. 3.11. Some of the results are slightly anomalous due to the presence of room return neutrons. Two samples gave somewhat more consistent results than the others and are discussed here. One of these, zirconium hydride, was studied to complement the angular scattering measurements that we have made on that moderator. In Fig. 3.12, the measured total cross section of $\text{ZrH}_{1.87}$ is compared to the theoretical values. The calculation is based on a model⁽²³⁾ having a single harmonic oscillator having a broadened width at 0.13 eV for the optical modes and a Debye spectrum for the acoustical modes with a cutoff at 0.02 eV. The discrepancy between the measured and calculated σ_T is reduced when the calculation uses a mass 360 for the long wave length acoustical modes instead of mass 90. In addition, the calculated σ_T show many bumps above 0.1 eV corresponding to quantized excitation of the proton in the harmonic potential. The measured σ_T shows only three such bumps indicating that the harmonic well has a finite depth. Below 0.006 eV, the measurements appear to display characteristics that one would expect from coherent scattering by the zirconium. It can certainly be concluded that this crude σ_T measurement affords useful comments on the model for zirconium hydride.

Fig. 3.11--Geometrical arrangement for total cross section studies

Fig. 3.12--Total cross section of $ZrH_{1.87}$

The other sample for which good data were obtained was copper; this was done in order to investigate the non $1/v$ behavior of our bank of low pressure BF_3 detectors which have copper walls. This measurement is compared to the BNL 325 values in Fig. 3.13. Since the low energy and high energy parts of our measurements agree with BNL 325, we have some confidence in our data in the middle energy range where they have better energy resolution than the other source. Our data show the BNL 325 bump at 0.006 eV to be split into two sharp spikes, and the presence of a large bump at 0.02 eV not shown in the other reference. While our copper sample was not polycrystalline (a piece of rod was used), the observation is consistent with the pattern we have measured with the low pressure BF_3 detectors.

There are reservations about these two sets of data, but despite this the measurements are quite informative. Further studies are anticipated during the next quarter to improve our knowledge of the total cross sections for the common moderators.

3.7 LEAKAGE SPECTRA FROM A FAST U^{235} METAL BALL REACTOR

The experiments performed with the Accelerator Pulsed Fast Assembly (APFA) were described and discussed in the previous annual report, ⁽¹⁾ in the section devoted to Fast Reactor Studies.

A Monte Carlo calculation for the intermediate energy (1.5 MeV to $\sim$ keV) neutron detector was performed recently. ⁽²⁵⁾ It determined the spatial distribution of the (n, γ) conversion in the B_4C /paraffin slab. Its results, combined with those of the measurement of the gamma attenuation in the slab and detection by the gamma sensitive liquid scintillator, provided the values of the intrinsic efficiency $G(E)$ for that detector. These were needed to reduce the APFA time-of-flight data.

Fig. 3.13--Total cross section of copper

During the last few months, considerable effort has been made to increase the understanding of the behavior of the detectors used in the APFA spectra measurements. The detectors are normally gated off; they are gated on for some 150 μ sec after the end of the gamma flash originated by each Linac electron burst; this procedure avoids undesirable slow-recovery saturation effects due to the intense radiation field. In a careful checkup of the experimental techniques, it was found that the effect of the transient when gating-on affected the calculated intrinsic efficiency $G(E)$ for the detectors. As a consequence, a correction has to be made to those $G(E)$ functions for properly reducing the data collected. Meanwhile, the electronics of the detectors are being modified to minimize the aforementioned transient effect. This correction depends on the time interval between the Linac burst and the instant in which the detectors are gated on. The correction has already been measured for the intermediate energy neutron detector by determining the time distribution of random pulses immediately after the detector is gated on.

In Fig. 3.14, APFA intermediate energy leakage neutron spectra are shown. They were measured at different angles from the normal to the surface with a 0.87 in. precollimator. Obviously, they supersede Fig. 8.5 of Ref. 1. Error bars indicative of the standard deviation ΔN of the neutron flux per unit energy $N(E)$ values for different energy regions are shown on some points. The point at 426 keV was too high in all the cases when first calculated. This has been attributed to a very prominent dip in the product of the intrinsic efficiency of the detector (due to the boron σ_T resonance) and the flight path transmission (due to the oxygen and nitrogen σ_T resonances). This dip is narrower than the actual time (and hence, energy) resolution for the neutron time-of-flight determination, which in these experiments is also affected by the time spread in the emission from the U^{235} sphere (~ 112 nsec) and in the capture in the

Fig. 3.14--Leakage neutron spectra at intermediate energies from a U^{235} metal ball reactor

B_4C /paraffin slab (~ 150 nsec). The spurious effect resulting from this dip was corrected by replacing that point and its two neighbors by their average.

The calculated $G(E)$ curve for the fast neutron detector is presently in process of being adjusted to render final $N(E)$ curves in the range 0.8 to 15 MeV. The theoretical predictions are still being calculated and present some difficulty due to the large anisotropy of the flux. Various calculational approaches are being tried. A preliminary comparison is shown in Fig. 3 of Appendix C.

3.8 PROPOSED FAST SPECTRAL STUDIES IN URANIUM

The large discrepancies between the calculated and experimental values of neutron lifetimes and boron control reactivities in fast-spectrum assemblies are well-known and adequately documented.⁽²⁶⁾ As these quantities are dependent on the neutron energy distribution, direct measurements of the spectra of these fast-spectrum assemblies are in order. Numerous such measurements have been made⁽²⁷⁻²⁹⁾ and compared with calculated spectra for the assemblies; the usual outcome is that the low energy end of the spectrum is being incorrectly described by the computations.

There are two possible causes of these spectral inconsistencies. One is the possibility that the nuclear cross sections may be in error. Especially suspect are the cross sections for inelastic scattering of high energy neutrons to lower energies, and the capture cross sections in the unresolved resonance region (see Appendix C). Another possible error, although not as suspect as the cross section data, is the computational method involved. Specific questions here involve the adequacy of the energy-space-angular mesh, the treatment of the excitation source spectrum and the adequacy of the resonance absorption treatment.

An idea of the fit between computed and experimental spectra may be seen in Fig. 1 of Appendix C. Experimental points are taken from the work of Kato and Russell⁽²⁸⁾ while computations have been made with a one-dimensional transport code GAPLSN⁽²¹⁾ and various neutron sources (photoneutron, fission neutron). The typical underestimate of the low energy end of the spectrum is apparent.

The desirability of constructing a fast-spectrum assembly which will allow an accurate intercomparison of theory and experiment is evident. Such an apparatus should be capable of explicit description, without approximations, in a computational scheme and should be flexible enough to allow measurement of internal spectra as well as leakage spectra. Additional constraints include (1) reasonable emergent beam intensity, (2) capability of measuring scattered neutrons at varying angles, and (3) a short dieaway time for fast neutron pulses initiated by the Linear Accelerator. Provision for heating the entire assembly to several hundred degrees Centigrade for measurement of the Doppler effect is also desirable.

The preliminary design of such an assembly has been completed and is shown in Fig. 3.15. As presently envisioned it consists of a 20-in. diameter sphere of depleted uranium metal (.2 wt. % U-235) with appropriate beam reentry holes at varying radii and central target space for the standard Linac target. Considerable care has been exercised to eliminate void spaces and streaming cracks which could invalidate experimental measurements. Plugs are provided for filling the reentry holes not being used. The effect of possible anisotropy of the neutron source, with peaking in the electron beam forward direction, has been minimized by locating the reentry holes at 90° to the beam. The flux can be examined at various angles by using plugs to fill the reentry hole to a partial depth, thus varying the angle between the radius vector from the neutron source and the viewing direction. Standard time-of-flight neutron spectroscopy is planned for the spectral measurements.

Fig. 3.15--Proposed assembly for fast spectrum measurements
in depleted uranium

Computational methods will utilize the cross sections from GAM II (or, if available, a later version of this code) in an explicit geometric representation with either the GAPLSN⁽²¹⁾ or DTF-IV⁽³⁰⁾ transport codes. The excitation source utilized in the calculation will be modified to match the leakage spectrum of the Linac target. This leakage spectrum will be measured both with and without the surrounding 20-in. diameter ball so that the effect of the larger sphere on the source spectrum can be estimated. Previous computations⁽³¹⁾ with various methods of source insertion (e.g., as a homogeneous, zero-density source with a leakage spectrum distributed over the volume; as a thin spherical shell with a leakage spectrum surface source, or as a point source) have shown slightly different results. The appropriate method is the one which enables the calculated leakage spectrum to match the experimentally measured spectrum for the Linac target. Work is in progress on this aspect of the computation.

4. EXPERIMENTAL AND CALCULATIONAL TECHNIQUES

4.1 MODIFICATIONS TO THE ECTO TIME-OF-FLIGHT DATA REDUCTION CODE

Several improvements have been made to the time-of-flight data reduction code ECTO.⁽³²⁾ The resulting new version is called HECTO; it maintains all the capabilities of ECTO

4.1.1 Energy Dependent Energy Resolution

The option of using an energy dependent energy resolution $R(E)$ is now available. R is used by the code for deciding when to group adjacent data channels until $\Delta E/E \geq R$. Up to now only provision for a constant R value for all energies existed. The time-of-flight T , the energy E and their uncertainties are related to the length of the flight path L by

$$R = \Delta E/E = 2\Delta T/T \propto \Delta T E^{1/2}/L$$

For a ΔT independent of energy it is convenient to express

$$R(E) = R_{NF} (E/E_{NF})^{1/2}$$

where R_{NF} is the energy resolution at energy E_{NF} . This expression is used in HECTO to account for the energy dependence of R , with R_{NF} and E_{NF} as input parameters. For $E_{NF} = 0$, the code groups channels with a constant $R = R_{NF}$.

4.1.2 Increased Energy Resolution in the Detector Sensitivity Used by the Code

ECTO has provisions for forming a sensitivity table $S(E)$ by multiplying the intrinsic efficiency $G(E)$ of the detector by the attenuation calculated from the thicknesses of the materials present in the flight path and the total cross section tables included in the input. This $S(E)$ table is calculated for the values of E for which $G(E)$ is given also in a table included in the input. Finally, the code interpolates the sensitivity from the $S(E)$ table for the energy value corresponding to each channel for which data are reduced.

This procedure obviously results in a loss of the energy resolution of the total cross section tables through the intermediate formation of an $S(E)$ table; this loss might be minimized by the painstaking procedure of preparing more dense $G(E)$ tables, taking into consideration the energy ranges in which any of the cross sections that are likely to be used show a strong energy dependence.

In HECTO the transmission is calculated from the total cross section tables directly for every energy value corresponding to a channel for which data are reduced. This avoids any loss of resolution.

4.1.3 Calculation of the Standard Deviation of the Number of Counts

Changes have been made to the method of computing the statistical uncertainty. HECTO now calculates the standard deviation of the net (constant background per channel CONB subtracted) number N of counts with the formula from Ref. 33.

$$\frac{\Delta N}{N} = \frac{\left[\sum_{\nu} S_j + \frac{\nu^2 \text{CONB}}{\text{NB2} - \text{NB1} + 1} \right]^{1/2}}{\sum_{\nu} S_j - \nu \text{CONB}}$$

where ν is the number of grouped channels whose gross counts S_j are used to calculate N ; and CONB has been averaged from channel NB1 to channel NB2.

4.1.4 Other Changes

The interpolation in the intrinsic efficiency of the detector $G(E)$ and mean emission time tables was linear in ECTO. HECTO now provides for a second degree Lagrangian interpolation for both functions.

E/v^2 , the energy divided by the square of the velocity, was recalculated for nonrelativistic neutrons with improved data,⁽³⁴⁾ and inserted in HECTO:

$$E/v^2 = (.52268 \pm .00002) \text{ eV } \frac{\mu\text{sec}^2}{\text{cm}^2}$$

A correction for relativistic neutrons (which amount to 3.1% at 18.8 MeV) will be included late.

Some of the aforementioned changes resulted in a reduction of the memory space available for input tables. This was circumvented by providing for the tables with the intrinsic efficiency $G(E)$ to be written on tape and to be recalled to the fast memory of the computer one at a time when the particular problems to which they correspond are being reduced. The present input capacity for $G(E)$ tables is no longer limited, and each can now contain up to 450 points.

4.2 SENSITIVITY OF THE 32 BF_3 DETECTOR BANK AND OTHERS

The accuracy with which thermal neutron spectra are determined depends directly on how accurately the sensitivity of the detector measuring the spectra is known. For present thermal neutron spectra, the detector in question is a bank of 32 BF_3 detectors. This bank comprises five rows of detectors having a 12-in. active length and containing BF_3 -enriched in B^{10} to 96% at 70 cms Hg pressure. It is very difficult to

calculate directly the sensitivity of this detector bank due to its geometry. It has therefore been standard practice to calibrate the bank by comparing its response to a given flux with that from a known detector.

We discuss here a recent calibration of this BF_3 bank using as standard detectors a set of low pressure BF_3 counters and a Li^6 glass scintillator. This new calibration has been motivated by the strong interest in making spectrum measurements outside the energy range (.001 eV to 100 eV) for which previous calibrations have been made. In addition there was some question as to whether the low pressure BF_3 counters really do have a $1/v$ response. Furthermore, prior calibrations⁽³⁵⁾ resulted in unresolved discrepancies between the values obtained with the two different standard detectors. In the following sections, the response of each detector is discussed in turn.

There is at General Atomic a set of 10 stacked BF_3 counters, each 1-in. diameter containing natural BF_3 at 20 cms of mercury. These detectors have been assumed to have a $1/v$ response, but some doubt on this exists. A mathematical model has been set up to calculate the efficiency of this detector bank by treating a single counter simply when placed in a direction perpendicular to the incident beam. The counter is split into discrete segments parallel to the beam; the average wall thickness and BF_3 path length are calculated for each segment. The wall is allowed to scatter neutrons half in the forward direction and half in the backward direction. There will, therefore, be some secondary absorption from the neutrons backscattered from the rear wall. The contributions from each segment are added up and normalized. In addition this response is divided by a $1/v$ response so that the departure therefrom may be easily observed. Table 4.1 gives this efficiency. It should be noted that the calculated departure from a $1/v$ behavior can be related entirely to the variation of the scattering cross section of copper at its Bragg cutoff. The following specifications were used in the computation.

Table 4.1
EFFICIENCY OF LOW PRESSURE BF₃ DETECTORS

<u>Energy</u>	<u>Sensitivity</u>	<u>Sensitivity $\times \sqrt{\frac{E}{E_o}}$</u>
1.00000-03	5.63414-02	1.12013-02
2.00000-03	4.19830-02	1.18040-02
3.00000-03	3.48286-02	1.19932-02
4.00000-03	3.04499-02	1.21075-02
5.00000-03	2.74048-02	1.21829-02
6.00000-03	2.51230-02	1.22345-02
7.00000-03	2.33476-02	1.22809-02
8.00000-03	2.19068-02	1.23187-02
9.00000-03	2.07072-02	1.23504-02
1.00000-02	1.96886-02	1.23781-02
1.20000-02	1.80353-02	1.24209-02
1.50000-02	1.61919-02	1.24676-02
2.00000-02	1.40812-02	1.25197-02
3.00000-02	1.15554-02	1.25830-02
4.00000-02	1.00374-02	1.26209-02
6.00000-02	8.22489-03	1.26662-02
8.00000-02	7.13816-03	1.26932-02
1.00000-01	6.39389-03	1.27117-02
1.50000-01	5.23246-03	1.27406-02
2.00000-01	4.53758-03	1.27579-02
3.00000-01	3.71087-03	1.27784-02
4.00000-01	3.21679-03	1.27906-02
6.00000-01	2.62948-03	1.28052-02
8.00000-01	2.27874-03	1.28139-02
1.00000-00	2.03911-03	1.28198-02
1.50000-00	1.66612-03	1.28290-02
2.00000-00	1.44352-03	1.28345-02
3.00000-00	1.17923-03	1.28410-02
4.00000-00	1.02155-03	1.28449-02
6.00000-00	8.34393-04	1.28495-02
8.00000-00	7.22757-04	1.28522-02
1.00000-01	6.46551-04	1.28541-02
1.50000-01	5.28023-04	1.28570-02
2.00000-01	4.57345-04	1.28587-02
3.00000-01	3.73481-04	1.28608-02
4.00000-01	3.23475-04	1.28621-02
6.00000-01	2.64145-04	1.28635-02
8.00000-01	2.28771-04	1.28643-02
1.00000-02	2.04630-04	1.28650-02
1.50000-02	1.67090-04	1.28658-02
2.00000-02	1.44711-04	1.28664-02

Copper Wall Thickness	0.056 cm
Counter Internal Radius	1.27 cm
Copper Wall Atom Density	0.08408 atoms/barn-cm
B ¹⁰ Number Density	1.373 x 10 ⁻⁶ atoms/barn-cm
B ¹¹ Number Density	5.106 x 10 ⁻⁶ atoms/barn-cm
Cross sections are from BNL 325.	

There are several Li⁶ glass scintillators at General Atomic. The Li⁶ content of these scintillators was suspected as being less than the value quoted by the supplier. Consequently another one was ordered in which a cullet was taken from the ingot from which the fluor was cut. This cullet was analyzed by both the supplier and by General Atomic with the following results.

Supplier Nominal Specification (Crystal)	6.98% Li ⁶
Supplier Analysis (Cullet)	6.15% Li ⁶
General Atomic Analysis (Cullet)	6.87% Li ⁶

To resolve the above anomalies a transmission experiment through the scintillator was performed. The constituents of the fluor consist for the most part of nuclei having negligible absorption and constant cross section. This makes the analysis easier.

$$\text{Reciprocal transmission} \quad R = \exp \{ T (\Sigma_a + \Sigma_s) \}$$

where $T = \text{fluor thickness}$

$$\ln(R) = \frac{T \Sigma_{a0} \sqrt{E_0} + T \Sigma_s}{\sqrt{E}}$$

Thus by plotting $\ln(R)$ against $1/\sqrt{E}$, we should get a straight line whose slope is $T \Sigma_{a0} \sqrt{E_0}$ and whose intercept on the Y axis is $T \Sigma_s$, provided the

absorption is all $1/v$. This is what is shown in Fig. 4.1. From the slope of this line, the Li^6 content of the glass was computed to be 5.93%. This value is believed to be the most reliable and has been used to compute the detector efficiency. For reference the following specifications have been used for the scintillator. It would be most desirable to calculate

Diameter	2.529 cms		
Thickness	1.2743 cms		
Density	2.4764 gms/cc		
Composition (% Wt)	Li_2O	13.8	(95.4% Li^6)
	MgO	8.7	
	Al_2O_3	12.1	
	Ce_2O_3	2.7	
	SiO_2	62.7	

the efficiency of this detector by Monte Carlo methods. This calculation is anticipated at a later time. Meanwhile a simple computation has been performed which will be accurate at least up to 2 eV and will indicate the magnitude of multiple scattering corrections at much higher energies. The mathematical model for this detector is as follows.

The detector is considered with its circular face perpendicular to the incident beam. The thickness is divided into discrete increments. The primary absorption term is computed simply taking into account also the attenuation through one mil of aluminum foil at the leading edge. For absorptions additional to the primary, the scattered neutrons are presumed to be isotropic in the laboratory system with no change of energy. At each spatial point along the cylindrical axis a series of discrete angles is considered and path lengths are computed either to the crystal edge or to a limit of one complete radius. The corresponding absorption is calculated, and the angular and energy integration is carried out for each initial neutron energy. For secondary absorption, the source neutron distribution comes from the primary attenuation. For tertiary and higher

Fig. 4.1--Neutron transmission through 1/2" thick lithuim glass

absorption, the source neutron distribution is presumed to be a cosine to the extrapolation length on each side of the fluor. The radial spatial distribution is ignored, and hopefully the limitation of one scattering path equal to one crystal radius will account for the leakage of neutrons. The absorption due to collision terms higher than the third have a fixed ratio at each energy to the absorption for the preceding collision source. This makes for a simple summation. The calculated efficiency of the detector is given in Table 4.2.

Although the calculation of the 32 BF_3 bank sensitivity is difficult except by Monte Carlo methods, a simplified model may be useful since it will be accurate where the bank is almost totally absorbing, and it will indicate where the bank takes on a $1/v$ response. The latter will be helpful when measurements at higher energy are made. The mathematical model assumed here is as follows, with the geometry indicated in Fig. 4.2. The basic element is split into 22 segments and the average wall thickness and path length for each counter is calculated for each segment. The neutrons are considered to be scattered half in the forward direction and half in the backward direction at each wall. This is further approximated by ignoring all scattering beyond the first copper wall and before the last. Therefore, only straight absorption attenuation is considered. The back-scattering from the last wall is allowed to be attenuated through the whole stacked array. Contributions from each segment are added up and normalized. The results of this calculation are given in Table 4.3

The calibration of the 32 BF_3 bank is quite simple in principle. A standard spectrum $\Phi(E)$ is measured by the bank to give count $C_o(E)$. For the standard detector with efficiency $S_1(E)$, counts $C_1(E)$ are obtained.

$$\text{Hence } \Phi(E) = \frac{C_o(E)}{S_o(E)} = \frac{C_1(E)}{S_1(E)}$$

Fig. 4.2--Calculation geometry for sensitivity of the 16-meter detector system

Table 4.2
EFFICIENCY OF LITHIUM6 GLASS SCINTILLATOR

<u>Energy (eV)</u>	<u>Contribution to Sensitivity From Primary Absorption</u>	<u>Total Sensitivity</u>
0.10000-02	0.99582-00	0.99750-00
0.15000-02	0.99504-00	0.99728-00
0.20000-02	0.99438-00	0.99710-00
0.30000-02	0.99326-00	0.99681-00
0.40000-02	0.99231-00	0.99658-00
0.60000-02	0.99072-00	0.99622-00
0.80000-02	0.98938-00	0.99593-00
0.10000-01	0.98820-00	0.99568-00
0.15000-01	0.98570-00	0.99515-00
0.20000-01	0.98359-00	0.99472-00
0.30000-01	0.98007-00	0.99400-00
0.40000-01	0.97713-00	0.99340-00
0.60000-01	0.97221-00	0.99239-00
0.80000-01	0.96807-00	0.99151-00
0.10000-00	0.96440-00	0.99068-00
0.15000-00	0.95634-00	0.98855-00
0.20000-00	0.94907-00	0.98613-00
0.30000-00	0.93543-00	0.98021-00
0.40000-00	0.92232-00	0.97316-00
0.60000-00	0.89730-00	0.95721-00
0.80000-00	0.87398-00	0.94046-00
0.10000+01	0.85242-00	0.92390-00
0.15000+01	0.80544-00	0.88539-00
0.20000+01	0.76647-00	0.85160-00
0.30000+01	0.70513-00	0.79593-00
0.40000+01	0.65836-00	0.75179-00
0.60000+01	0.59014-00	0.68515-00
0.80000+01	0.54152-00	0.63611-00
0.10000+02	0.50434-00	0.59778-00
0.15000+02	0.43920-00	0.52891-00
0.20000+02	0.39562-00	0.48159-00
0.30000+02	0.33876-00	0.41833-00
0.40000+02	0.30196-00	0.37643-00
0.60000+02	0.25524-00	0.32213-00
0.80000+02	0.22571-00	0.28714-00
0.10000+03	0.20481-00	0.26203-00
0.15000+03	0.17103-00	0.22089-00
0.20000+03	0.15013-00	0.19506-00
0.30000+03	0.12457-00	0.16307-00

Table 4.2 (Contd.)

0.40000+03	0.10893-00	0.14326-00
0.60000+03	0.89968-01	0.11900-00
0.80000+03	0.78452-01	0.10414-00
0.10000+04	0.70501-01	0.93818-01
0.15000+04	0.57987-01	0.77471-01
0.20000+04	0.50439-01	0.67550-01
0.30000+04	0.41398-01	0.55605-01
0.40000+04	0.35963-01	0.48391-01
0.60000+04	0.29472-01	0.39742-01
0.80000+04	0.25580-01	0.34538-01
0.10000+05	0.22914-01	0.30966-01

Table 4.3

CALCULATED EFFICIENCY OF THE 32 BF₃ DETECTOR BANK

<u>Energy</u>	<u>Sensitivity</u>	<u>Sensitivity $\times \sqrt{\frac{E}{E_o}}$</u>
1.00000-04	6.25491-01	3.93243-02
1.50000-04	6.68151-01	5.14470-02
2.00000-04	6.94145-01	6.17170-02
3.00000-04	7.24737-01	7.89189-02
4.00000-04	7.42298-01	9.33357-02
6.00000-04	7.61685-01	1.17298-01
8.00000-04	7.72061-01	1.37289-01
1.00000-03	7.78390-01	1.54752-01
1.50000-03	7.86433-01	1.91491-01
2.00000-03	7.89661-01	2.22022-01
3.00000-03	7.98073-01	2.72337-01
4.00000-03	7.88547-01	3.13543-01
4.50000-03	7.86521-01	3.31708-01
5.00000-03	7.73633-01	3.43922-01
5.50000-03	7.77079-01	3.62315-01
6.00000-03	7.73634-01	3.76748-01
6.50000-03	7.62418-01	3.86447-01
7.00000-03	7.66009-01	4.02923-01
7.50000-03	7.65183-01	4.16616-01
8.00000-03	7.62948-01	4.29022-01
9.00000-03	7.57938-01	4.52058-01
1.00000-02	7.51897-01	4.72714-01
1.30000-02	7.28436-01	5.22159-01
1.60000-02	7.07798-01	5.62871-01
1.90000-02	6.88436-01	5.96596-01
2.20000-02	6.77709-01	6.31967-01
2.50000-02	6.67341-01	6.63373-01
3.00000-02	6.50968-01	7.08860-01
3.50000-02	6.32276-01	7.43670-01
4.00000-02	6.14955-01	7.73237-01
4.50000-02	5.99513-01	7.99548-01
5.00000-02	5.86193-01	8.24072-01
5.50000-02	5.72844-01	8.44613-01
6.00000-02	5.60622-01	8.63347-01
7.00000-02	5.39327-01	8.97100-01
8.00000-02	5.21259-01	9.26912-01
9.00000-02	5.04761-01	9.52021-01
1.00000-01	4.89690-01	9.73556-01

Table 4.3 (Contd.)

1.50000-01	4.32719-01	1.05364-00
2.00000-01	3.93536-01	1.10647-00
3.00000-01	3.41051-01	1.17441-00
4.00000-01	3.06409-01	1.21835-00
6.00000-01	2.61649-01	1.27419-00
8.00000-01	2.32906-01	1.30968-00
1.00000-00	2.12330-01	1.33490-00
1.50000-00	1.78702-01	1.37599-00
2.00000-00	1.57645-01	1.40164-00
3.00000-00	1.31627-01	1.43333-00
4.00000-00	1.15551-01	1.45293-00
6.00000-00	9.59056-02	1.47693-00
8.00000-00	8.38846-02	1.49165-00
1.00000+01	7.55436-02	1.50189-00
1.50000+01	6.23479-02	1.51812-00
2.00000+01	5.43460-02	1.52800-00
3.00000+01	4.47191-02	1.53990-00
4.00000+01	3.89091-02	1.54711-00
6.00000+01	3.19468-02	1.55576-00
8.00000+01	2.77594-02	1.56097-00
1.00000+02	2.48858-02	1.56455-00
1.50000+02	2.03920-02	1.57016-00
2.00000+02	1.76979-02	1.57353-00
3.00000+02	1.44871-02	1.57755-00
4.00000+02	1.25654-02	1.57996-00

The following specifications were used in the computation:

Copper Wall Thickness	0.056 cms
Counter Internal Radius	2.54 cms
Copper Wall Atom Density	0.08408 atoms/barn-cm
B ₁₀ Number Density	2.223×10^{-5} atoms/barn-cm
B ₁₁ Number Density	2.709×10^{-6} atoms/barn-cm

Cross Sections from BNL 325, except copper which used the data of Fig. 3.13.

Thus the BF_3 bank sensitivity is given by

$$S_o(E) = \frac{C_o(E)}{C_1(E)} S_1(E)$$

We can obtain $C(E)$ from the output of our spectral data reduction code ECTO⁽³²⁾ so as to correct properly for count rate losses, etc. The data are reduced initially taking a detection sensitivity as constant at 1.00.

Before checking the three calibrations it is worth while to plot (in Fig. 4.3) the low energy counts $C(E)$ for the three detector systems (each reduced with $S(E) = 1.0$). We observe that the response of the Li^6 detector is a smooth curve. The response of the $1/v$ bank has some significant departures from a smooth curve. These departures can be observed, though to a much lesser extent in the response of the 32 BF_3 detector bank. These departures are believed to be due to the coherent elastic scattering by the copper and that its $\bar{\mu}$ becomes negative in the vicinity of the Bragg cutoff. These departures are certainly not reflected in the computed efficiency of the low pressure BF_3 detectors (see Table 4.1). This effect was one reason for measuring the total cross section of copper as reported in Section 3.6. The results shown in Fig. 3.13 indicate considerably more structure than that shown in BNL 325. However, it was discovered that even if the new total cross section for copper were to be used in the efficiency calculation, the low pressure BF_3 bank sensitivity would still be computed incorrectly since there is no angular scattering allowed for in its simplified mathematical model. The conclusion to be drawn from Figs. 3.13 and 4.3 is that the efficiency of the low pressure BF_3 detectors is not likely to be $1/v$ in behavior below 0.03 eV. In Fig. 4.4 the sensitivities of the 32 BF_3 bank determined by the three methods are compared with each other. The agreement between the low pressure

Fig. 4.3--Neutron spectra uncorrected for detector sensitivity

Fig. 4.4--Measured and calculated sensitivity of the
16-meter detector system

BF₃ bank and the Li⁶ glass scintillator has been much improved by the more exact determination of the latter's Li⁶ content. If we ignore the low pressure BF₃ bank values below 0.03 eV (due to the incorrect treatment of the elastic coherent scattering by the copper walls), we can say that the two detectors give excellent agreement from that energy to 20 eV. The calculated sensitivity of the 32 BF₃ bank is in surprisingly good agreement with the "measured" calibrations except above 20 eV. The discrepancy above 20 eV is believed to be due to neutrons from pulsing overlap, in which case the computed sensitivity will be more accurate. It has therefore been decided to use this new computed sensitivity as the standard efficiency calibration of the 32 BF₃ detector bank. In Fig. 4.5, a comparison is made of this new calibration with the previous one measured on December 18, 1962. The old one has more structure, reflecting that it was a "measured value". There is little difference between the old and new set of values between 0.025 and 20 eV so that our older spectral data will be satisfactory over that energy range. Fortunately that energy range is the dominant one in comparing measured with calculated thermal neutron spectra. Below 0.025 eV and above 20 eV, significant discrepancies occur. Theoretical comparison of neutron spectra have not usually been made above 10 eV, so that we cannot make much of the discrepancy at the higher energy. Below 0.025 eV our measured and calculated neutron spectra have not always been in good agreement; hopefully this newer calibration will be an improvement in that area.

4.3 STATUS OF THE 200-METER FLIGHT PATH

The construction contract for the 200-meter flight path was let March 21, 1966. Construction of the flight path has begun and most of the excavation is completed. The schedule submitted by the contractors indicate that the flight path should be installed by mid-June. It is

Fig. 4.5--New and old sensitivities of the 16-meter detector system

anticipated that the acceptance test and evacuation of the flight tube will be completed shortly thereafter. If this schedule is maintained the flight path should be ready for installation of experimental apparatus by July 1966.

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APPENDIX A

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NEUTRON SCATTERING BY HYDROGENEOUS MODERATORS

by

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NEUTRON SCATTERING BY HYDROGENEOUS MODERATORS

I. INTRODUCTION

The theory of binding effects on neutron thermalization by hydrogen is a particularly interesting subject because of the great variety of bound states in which hydrogen occurs in nature. In order to get a simple qualitative idea of the diversity of these binding effects it suffices to take a look at two integral quantities which are representative of the energy exchange and transport properties of different materials. These two quantities are the slowing down power, usually defined by

$$\xi \sigma_s(E_0) = 2\pi \int_0^\infty \int_{-1}^1 \sigma(E_0 \rightarrow E; \theta) \ln \frac{E_0}{E} dE d(\cos \theta)$$

and the average cosine of the scattering angle

$$\bar{\mu}(E_0) = \frac{2\pi}{\sigma_s} \int_0^\infty \int_{-1}^1 \sigma(E_0 \rightarrow E; \theta) \cos \theta dE d(\cos \theta)$$

where $\sigma(E_0 \rightarrow E; \theta)$ is the double differential scattering cross section from initial energy E_0 to final energy E through the angle θ , and σ_s is the total scattering cross section. In fact the diversity of hydrogen binding is clearly illustrated by Figs. 1, 2, 3, and 4 in which we compare $\xi \sigma$ and $\bar{\mu}$ as calculated for some typical hydrogenous moderators such as water, polyethylene, liquid hydrogen and zirconium hydride.

Fig. 1--Slowing down power $\xi\sigma_s$ for hydrogen bound in water and zirconium hydride

Fig. 2--Slowing down power $\xi\sigma_s$ of para- and ortho-hydrogen at 20°K

Fig. 3--Average cosine of scattering angle for polyethylene and zirconium hydride (scattering by C, respectively Zr is included)

Fig. 4--Average cosine of scattering angle for para- and ortho-hydrogen at $20^\circ K$

In the following we shall analyze with some detail the state-of-the-art of calculating slow neutron scattering from each one of these materials and compare the theoretical results with the most recent measurements. All calculations are made in the incoherent approximation which for scattering by H is excellent ($\sigma_{\text{inc}}/\sigma_{\text{coh}} \sim 40$). The only exception is liquid hydrogen where intramolecular interference as well as spin correlations are taken into account.

II. WATER

1. Historically H_2O was the first moderator for which a physically realistic scattering model, namely the Nelkin model⁽¹⁾ was given. As is well known, in this model the scattering units are freely recoiling H_2O molecules. Each molecule can undergo torsional harmonic oscillations (hindered rotations) as a whole with a single frequency of 0.06 eV, as well as internal vibrations producing a (symmetric or antisymmetric) change of length of the O-H bonds (frequency of 0.48 eV) and of the angle between them (0.20 eV). The original Nelkin model for water was a considerable improvement over the free monoatomic gas model by which it was preceded, and it could predict quite satisfactorily integral quantities such as the total scattering cross section, the transport mean free path, and neutron spectra in water moderated assemblies. It also was suited to the capabilities of digital computers available at the time of its formulation. However, as more and more accurate experimental data, both differential and integral, became available, it was recognized that a more sophisticated theory which also would take advantage of modern high speed computers, was required.

2. The simplest possible improvement over the Nelkin model was the analysis of the spatial anisotropy of the proton vibration in each H_2O molecule. The first step in this direction was the formulation of a model⁽²⁾ in which the following harmonic system of forces acting on a proton displaced

from equilibrium were explicitly postulated: a) a force along the direction of the O-H bond, proportional to the change of length of the bond; b) a force perpendicular to the bond and coplanar with the H₂O molecule, proportional to the change of angle between the two O-H bonds; c) aside from these internal restoring forces, it was assumed that the rotation of the whole molecule about its three principal areas of inertial are completely hindered by a harmonic torsional potential. This extremely simplified picture of the molecular dynamics in water yields six normal modes of vibrations (besides the three translational degrees of freedom corresponding to freely recoiling molecules). The calculations are considerably simplified if one neglects the displacements of the oxygen atom as compared to the vibrational amplitudes of the two protons. In this approximation the three rotational modes and the H-O-H bending mode correspond to each proton moving in a plane perpendicular to its O-H bond, whereas in the two stretching modes (which have the same frequency if oxygen displacements are neglected) the motion is in the direction of the O-H bond.

With the described anisotropic force system and further assuming that all three rotational modes have the same frequency and that the oxygen only moves in the translational and rotational modes the following expression is obtained for the double differential cross section:

$$\frac{d^2\sigma}{d\Omega d\epsilon} = \frac{a_b^2}{2\pi} \sqrt{\frac{E}{E_o}} \int_{-\infty}^{\infty} \exp(-i \frac{\epsilon t}{T}) \chi(\kappa^2, t) dt \quad (1)$$

$$\begin{aligned} \chi(\kappa^2, t) = < \exp \left\{ \frac{\kappa^2}{2m} \left[-\frac{it + Tt^2}{18} + g_s \cos^2 \theta_x + \frac{1}{2g_b} \cos^2 \theta_y + \right. \right. \\ \left. \left. + \frac{1}{2.25} g_r (\cos^2 \theta_y + \cos^2 \theta_z) \right] \right\} >_{\Omega} \end{aligned} \quad (2)$$

Here $\epsilon = E - E_0$, E_0 and E being the initial and final neutron energies, κ is the momentum exchange, T the temperature, m the neutron mass and a_b the bound scattering length. Planck's and Boltzmann's constants are set equal to unity. The function g_q is the familiar expression

$$g_q = \frac{1}{\omega_q} \left[(n_q + 1)(e^{-i\omega_q t} - 1) + n_q(e^{i\omega_q t} - 1) \right] \quad (3)$$

where

$$n_q = \left(\exp \frac{\omega_q}{T} - 1 \right)^{-1}$$

and ω_q is the frequency of mode q . The indices s , f and r refer to stretching, bending and rotational modes. θ_x , θ_y and θ_z are the directional angles of the vector $\vec{\kappa}$ in a cartesian coordinate system where the x axis is parallel to the OH bond, the y -axis also is coplanar with the H_2O molecule and the z axis is perpendicular to it. Finally $\langle \chi \rangle_\Omega$ indicates an average over all molecular orientations.

It is clear that for $\kappa^2 / 2m\omega_r \ll 1$ (ω_r is the lowest frequency) the average can be performed in the exponent and in this case Eq. (2) reduces to

$$\chi(\kappa^2, t) = \exp \left\{ \frac{\kappa^2}{2m} \left[-\frac{it + Tt^2}{18} + \frac{g_s}{M_s} + \frac{g_b}{M_b} + \frac{g_r}{M_r} \right] \right\} \quad (4)$$

with

$$M_s = 3, \quad M_b = 6, \quad M_r = 2.25 \quad (5)$$

which are practically the same as Nelkin's original masses ($M_s = 2.92$, $M_b = 5.85$, $M_r = 2.32$). It should be noted however, that within the harmonic approximation the masses given by Eq. (5) are extreme values

(maximum for M_r , minimum for M_s and M_b) since the only mode in which the oxygen is truly at rest is the rotation about the symmetry axis of the molecule.

For larger values of $\kappa^2/2\omega_r$, however, a more accurate method of averaging is in order. Since the exact averaging procedure requires considerable computing time for evaluation of a complete scattering kernel between, say 0 to 1 eV it is reasonable to make the following approximation for assessing the importance of anisotropy in Eq. (2): assume that the two-dimensional oscillations of the proton in the bending and torsional modes are isotropic in the plane perpendicular to the O-H bond (exact for $\omega_b = \omega_r$) but treat the one-dimensional stretching mode exactly. This leads to the expression

$$\chi(\kappa^2, t) = \frac{1}{2} \exp \left[-\frac{\kappa^2}{2m} (it + Tt^2) \right] \int_{-1}^1 d\mu \exp \left\{ \frac{\kappa^2}{2m} \left[g_s \mu^2 + \left(\frac{g_b}{4} + \frac{g_r}{1.5} \right) (1 - \mu^2) \right] \right\} \quad (6)$$

from which $d^2\sigma/d\epsilon d\Omega$ is easily evaluated using a modification of the code GAKER⁽³⁾ originally written for computing the Nelkin kernel.

3. A more radical modification of the Nelkin model, which, however, required the development of new, more elaborate computing techniques, was the introduction of a distributed frequency spectrum. As is well known, for a harmonic isotropic and incoherent crystalline scatterer, the Fourier transform of the scattering law, the quantity we previously called $\chi(\kappa^2, t)$ is expressed by

$$\chi(\kappa^2, t) = \exp \left\{ \frac{\kappa^2}{2m} [\gamma(t) - \gamma(0)] \right\}$$

with

$$\gamma(t) = \int_{-\infty}^{\infty} \frac{f(|\omega|) \exp \left(\frac{\omega}{2T} - i\omega t \right)}{2\omega \sinh \frac{\omega}{2T}} d\omega \quad (7)$$

M being the mass of the scatterer and $f(\omega)$ the frequency spectrum. It follows from Eq. (7) at least in principle that $f(\omega)$ can be obtained experimentally from the double differential cross section at small values of κ^2 through the relation:

$$f(\omega) = 2\omega \sinh \frac{\omega}{2T} \lim_{\kappa^2 \rightarrow 0} \left[\frac{2M}{\kappa^2} S(\kappa^2, \epsilon) \right] \quad (8)$$

with

$$S(\kappa^2, \epsilon) = \frac{T}{2} \sqrt{\frac{E_0}{E}} \exp \frac{\epsilon}{2T} \frac{d^2 \sigma}{d\Omega d\epsilon} \quad (9)$$

It was Egelstaff⁽⁴⁾ who first used this result for calculating scattering kernels from measured frequency spectra. He further showed that it can also be applied to liquids undergoing diffusive motion, at least if their χ -function is still reasonably well approximated by a gaussian like Eq. (7). In this case $f(\omega)$ no longer represents a true frequency spectrum but has a more generalized meaning. In fact it can then be shown⁽⁵⁾ that

$$\frac{3}{4} \frac{\omega}{\sinh \frac{\omega}{2T}} f(\omega)$$

is the Fourier transform of the velocity autocorrelation function taken at the complex time $t - i/2T$. Furthermore, for a liquid, this generalized $f(\omega)$ no longer vanishes at origin, $f(0)$ being proportional to its internal diffusion coefficient. In this case both $\gamma(t)$ and $\gamma(0)$ diverge but their difference remains finite for any finite time t . *

* When $f(0) \neq 0$ we have $\chi(\kappa^2, t) \rightarrow 0$ for $t \rightarrow \infty$. Hence there is no elastic scattering in this case.

Applying this extrapolation technique to the case of water, Haywood and Thorson⁽⁶⁾ found the frequency distribution shown in Fig. 5. They added the high frequency part peaking at 0.44 eV (which is the frequency actually observed in water as compared to the value 0.48 used by Nelkin and which corresponds to vapor) in order to account for the bond stretching vibrations of the H_2O molecule, and normalized the area under the peak to 1/3 in accordance with Eq. (5). It is seen that $f(0) \neq 0$ and that there are some low frequency wiggles, somewhat similar to what one would expect from translational vibrations in ice. However, the general features of the Nelkin model can still be recognized, namely the narrow peak at 0.205 eV due to the internal bond bending modes and a much broader peak centered at about 0.07 eV corresponding to the hindered rotations of the H_2O molecules.

The knowledge that water has a broad band of torsional oscillation is not recent. It was first inferred from Raman scattering⁽⁷⁾ and early calculations of the torsional potentials were made by Magat⁽⁸⁾ in 1936. This broad band has been explained by two main causes: a) the different potentials and moments of inertia of a single molecule for rotation about its three principal axes of inertia and the strong anharmonicity due to the shallow nature of these potentials; b) the clustering through hydrogen bonding of many molecules into quasicrystalline aggregates of different sizes.⁽⁹⁻¹⁴⁾ While the internal modes of the quite rigid individual molecules would practically not be affected by this rather weak coupling (similarly to the case of polyethylene to be considered in Section III), the rotational modes, however, would go over into a broad band as a consequence of the interaction between neighboring H_2O groups.

The second interpretation has led to still another modification of the Nelkin model, namely the treatment given recently by McMurry, et al.⁽¹⁵⁾ These authors consider that water is a mixture made up of 10% free H_2O

Fig. 5--Haywood's spectrum for H_2O at
 room temperature ($\beta = \frac{\omega}{kT}$)

molecules, 45% aggregates of mass 75 and another 45% aggregates of mass 150. They also assume that the internal vibrational modes of each molecule are unaffected by the clustering. Then they introduce a torsional and translational frequency spectrum for each one of the freely recoiling aggregates, represented by a system of 17 properly weighted discrete harmonic oscillators with frequencies between 0 and 0.125 eV. The weights or amplitudes of these oscillators which must satisfy a number of normalization conditions, were then chosen to give the best fit of Brugger's experimental scattering law data. ⁽¹⁵⁾

4. Computer Programs

The introduction of distributed frequency spectra naturally requires the development of suitable computer programs to deal with the increased complexity of the equations to be evaluated. The code SUMMIT⁽¹⁶⁾ developed at General Atomic as well as the English program LEAP⁽¹⁷⁾ use the familiar phonon expansion of the exponential in Eq. (7), which gives $S(\kappa^2, \epsilon)$ (as defined by Eq. (9)) in the form

$$S(\kappa^2, \epsilon) = \exp \left[-\frac{\kappa^2}{2M} \gamma(0) \right] \left[\delta(\epsilon) + \sum_{n=1}^{\infty} \frac{1}{n!} \left(\frac{\kappa^2}{2M} \right)^n G_n(\epsilon) \right] \quad (10)$$

with

$$G_1(\epsilon) = \frac{\exp\left(\frac{-\epsilon}{2T}\right) f(\epsilon)}{2\epsilon \sinh \frac{\epsilon}{2T}} \quad (11)$$

and

$$G_{n+1}(\epsilon) = \int_{-\infty}^{\infty} G_1(\epsilon - \omega) G_n(\omega) d\omega \quad (12)$$

A more recent program called GASKET⁽¹⁸⁾ uses a modern numerical Fourier transformation technique for evaluating the scattering law $S(\kappa^2, \epsilon)$. This latter method has the advantage of higher flexibility since it can handle practically any shape of $f(\omega)$ and does not have to cope with the convergence

problems of the phonon expansion. In its final version GASKET will also be able to handle anisotropies of the form of Eq. (6) for distributed frequency spectra.

5. Numerical Calculations and Comparison with Measurements

It is clearly unfair to compare double-differential cross sections calculated from either the isotropic or the anisotropic Nelkin model with the values obtained from the Haywood-Egelstaff or even the McMurry model. The details of the energy transfer between neutron and moderator can of course not be reproduced by a model in which the very complicated dynamics of a molecular liquid is approximated by a system of freely recoiling molecules only interacting with their neighbors through a single frequency torsional potential. Integral and single differential quantities, however, can be surprisingly well predicted by the three discrete oscillator models. In fact, Fig. 6 shows that the discrete oscillator models predict total scattering cross sections which agree with experiment to within a few percent for all energies down to about 0.005 eV. This should be compared to the cross sections calculated with different distributed frequency spectra shown in Fig. 7. Curve 1 was obtained using a frequency spectrum which is essentially the one measured by Haywood, as represented in Fig. 5, but modified in the following way: The area under the vibrational peak at 0.45 eV was approximated by a discrete line of the same frequency and weight $1/M_s = 1/3$ in accord with Eq. (5). Furthermore, an ω^2 behaviour was introduced at the low frequency end, as shown by the dashed line in Fig. 5, and the area between the dashed and solid lines, which was adjusted to 1/18 of the total area, was then treated in the short collision approximation (free gas of mass 18). Hence diffusive effects were crudely approximated by free recoil of the H_2O molecules. We shall designate this frequency spectrum as Haywood I. It is seen in Fig. 7 that Haywood I gives an overestimate of the cross section in the range 0.02 - 0.2 eV, which may be attributed to excessive weighting of the 0.205 eV peak relative to the area corresponding to the torsional modes. In fact Curve 2, which fits the experimental data

Fig. 6--Scattering cross section for hydrogen bound in H_2O ; comparison of discrete oscillator models with experiment

Fig. 7--Scattering cross section for hydrogen bound in H_2O ; comparison of distributed frequency spectrum calculation with experiment

much better, was obtained by cutting off the distributed part of the Haywood I spectrum at 0.165 eV, the bottom of the valley between the torsional and first vibrational peaks. The suppressed part was then replaced by an additional discrete line at 0.205 eV with weight 1/6 as required by Eq. (5). We shall call this modified spectrum Haywood II. Also, shown for comparison is Curve n^o3 representing the cross section calculated with a frequency spectrum consisting of the same two discrete oscillators as used in the previous case, plus a rotational band peaking at 0.07 eV, of the form:

$$\omega^2 \exp \frac{(\omega - \omega_0)^2}{-\sigma^2}$$

and with $\sigma = 0.03$ eV. This last curve fits the data best of all.

The average cosine of the scattering angle, $\bar{\mu}(E)$, is also reasonably well predicted by the discrete oscillator models. This is shown in Fig. 8 where it is seen that a better fit of the experimental data is obtained with the anisotropic model. The improvement is most significant in the energy region around 0.06 eV (rotational peak), where it reaches about 7 or 8%. The same figure also shows that the modified Haywood II spectrum described above gives the best fit over the whole range of energies relevant for thermal neutron spectra calculations. Also shown in Fig. 8 are the results obtained by McMurphy which are slightly worse than all previous calculations, with the exception of the very low energy range (0.001 - 0.010) where the other calculations break down because of their poor treatment of the diffusive or translational modes.

After having discussed the calculation of integral cross sections, we now should turn to differential quantities. Let us first consider the single differential cross section $d\sigma(E)/d\Omega$. This quantity can be measured with very good accuracy in the thermal range using detectors which are black (efficiency $\eta = 1$) for all neutron up to say 1 eV. It turns out that for most of this range all frequency spectra described above, discrete and distributed, give quite similar results. Only near the rotational resonance, between,

Fig. 8--Average cosine of scattering angle for H_2O
(scattering by oxygen is included in all calculations)

say 0.03 and 0.1 eV is there an appreciable difference between the different models, the best fit of Beyster's measurements⁽¹⁹⁾ being obtained with the Haywood II spectrum. This is shown in Fig. 9 for $E = 0.057$ eV. Actually the fit obtained with this model is excellent for all energies and all angles. The anisotropic discrete oscillator model fits the data nearly equally well but the isotropic Nelkin model tends to underestimate $d\sigma/d\Omega$ for the back-scattering angles.

Now turning to the comparison of measured and predicted double differential quantities, the discrepancies we find become much more severe. This can be attributed to shortcomings of both theory and experiment, as put in evidence by the large scattering of the experimental results. In fact, the experiment is faced with the major difficulty of multiple collision correction, which for a double differential measurement becomes extremely important and hard to account for accurately. In Figs. 10-15 we compare some of Haywood's data⁽²⁰⁾ for the scattering law $S(\alpha, \beta)$ as given by Eq. (9) (with $\alpha = \kappa^2/2MT$ and $\beta = \epsilon/T$) with the results obtained from GASKET, using the Haywood II frequency spectrum.* Here again the agreement is worst at low values of β , which is due to the incorrect treatment of diffusive effects. Also shown are some measurements by Kottwitz⁽²¹⁾ and calculations by McMurry. It is seen that for very low β the latter give a much better agreement than the GASKET calculations. This, of course, is not surprising since McMurry's parameters were actually adjusted to fit Brugger's data at small energy exchange.

For larger values of energy transfer we have compared the GASKET calculations with measurements performed by Kirouac, et al.⁽²²⁾ These data are presented in the form of $d^2\sigma/d\epsilon d\Omega$ for fixed initial energy and fixed scattering angle. Figures 16-21 show this comparison for initial energies of 0.23 and 0.65 eV and scattering angles of 25° , 60° and 90° . Again it is seen that qualitatively the agreement is reasonable but that quantitatively the

* Scattering by oxygen was not included in calculated double differential quantities.

Fig. 9--Single differential scattering cross section $d\sigma/d\Omega$ for H_2O atom initial energy $E_0 = 0.057$ eV

Fig. 10--Scattering law for H_2O at $\beta = 0.2$

Fig. 11--Scattering law for H_2O at $\beta = 1.00$

Fig. 12--Scattering law for H_2O at $\beta = 2.40$

Fig. 13--Scattering law for H_2O at $\beta = 2.81$

Fig. 14--Scattering law for H_2O at $\beta = 3.50$

Fig. 15--Scattering law for H_2O at $\beta = 5.96$

Fig. 16--Double differential scattering cross section for hydrogen bound in H_2O : $E_0 = 0.232 \text{ eV}$, $\theta = 25^\circ$

Fig. 17--Double differential scattering cross section for hydrogen bound in H_2O : $E_0 = 0.232 \text{ eV}$, $\theta = 60^\circ$

Fig. 18--Double differential scattering cross section for hydrogen bound in H_2O : $E_0 = 0.232$ eV, $\theta = 90^\circ$

Fig. 19--Double differential scattering cross section for hydrogen bound in H_2O : $E_0 = 0.65$ eV, $\theta = 25^\circ$

Fig. 20--Double differential scattering cross section for hydrogen bound in H_2O : $E_0 = 0.65 \text{ eV}$, $\theta = 60^\circ$

Fig. 21--Double differential scattering cross section for hydrogen bound in H_2O : $E_0 = 0.65 \text{ eV}$, $\theta = 90^\circ$

discrepancies are important, especially for small angles. For $E_0 = 0.65$ eV we also show the results obtained from the anisotropic Nelkin model. Since in this latter calculation the short collision approximation was made for the rotational modes above initial energies of 0.32 eV, the exaggerated structure due to the lumping of these modes into one discrete oscillator does not appear and the agreement with experiment at 0.65 eV is about as good as obtained with GASKET (Haywood II spectrum).

Finally we have also compared with experiment⁽²³⁾ some neutron spectra in poisoned H_2O calculated with the different models. The result of this comparison is shown in Figs. 22 and 23 for a solution of 5.2% borated H_2O , and confirms our previous conclusions namely that the best results are obtained with a distributed frequency spectrum, and that the inclusion of anisotropic effects in general seems to improve the agreement between theory and experiment.

In summary, as a result of the preceding analysis we might say that at present there is no model which fits all the integral and differential data equally well, although in general integral and single differential quantities calculated with GASKET and using the Haywood II frequency spectrum are in satisfactory agreement with experiment. It is hoped that by including in GASKET the anisotropic as well as the diffusive effects some overall improvement will be achieved. An alternate possibility of improvement could be expected from McMurry's idea of considering a mixture of several aggregates containing different number of H_2O molecules each one with its own frequency spectrum which, however should be continuous rather than discrete in order to simplify the calculations with a GASKET like program.

III. POLYETHYLENE

1. Crystalline polyethylene is known to be formed of very long molecules having a planar zig-zag shape with a CH_2 radical at each vertex.⁽²⁴⁾ The lattice unit cell is orthorhombic with its c-axis parallel to the axis of the molecular chain. There are four CH_2 groups per unit cell, which however belong to two different molecules. Since the coupling between neighboring chains is rather weak, it is quite accurate to calculate the

Fig. 22--Ratio of predicted to measured spectrum in 5.2% borated H_2O
(scattering by oxygen is included)

Fig. 23--Infinite medium neutron spectrum in 5.2% borated H_2O

frequency spectrum of polyethylene from a model of infinitely long, non-interacting molecules. Only at very low frequencies will the results be altered by cross links between molecules. The vibrational behavior of such a system of completely uncoupled chains is radically different from that of an ordinary three dimensional solid. In fact, analyzing its normal modes of vibration in terms of plane waves $C_n \exp(i \vec{k} \cdot \vec{r}_n)$, since only atoms of the same molecule are supposed to interact, it is clear that the frequency of a mode can only depend on $\vec{k} \cdot \vec{e}_c$, the projection of the wave vector on the c axis of the crystal, which is parallel to the chains. This means that for any arbitrary direction of $\vec{k}$, the frequency can only depend on the phase difference 2θ between the vibration of corresponding atoms in two consecutive unit cells belonging to the same molecular chain. Furthermore, since two consecutive CH_2 repeat units are equivalent to each other under a screw (helical) symmetry operation (translation $\vec{c}/2$ plus rotation of 180° about the molecular axis) it is possible to reduce the order of the secular determinant to 9×9 corresponding to the three atoms in each repeat unit. Thus the solution of the secular equation with θ as a parameter yields nine different branches. The results of such a calculation done by Lin and Koenig⁽²⁵⁾ are shown in Fig. 24. The nine branches can be assigned to the following modes of vibration.*

1	T	C-C-C-C torsion
2	B	} C-C-C bending and stretching
3	S	
4	R	CH_2 rocking
5	Tw	CH_2 twisting
6	W	CH_2 wagging
7	Ib	H-C-H bending
8	Ss	C-H symmetric stretching
9	As	C-H antisymmetric stretching

* This assignment is only qualitative for some of the branches. Tasumi, et al, ⁽²⁰⁾ actually find that the mode R corresponds exactly to rocking at $\theta = \pi$ and to twisting at $\theta = 0$, the reciprocal holding for mode Tw.

Fig. 24--Frequency distribution for polyethylene as calculated by Lin and Koenig

It is seen that the branches T and B are acoustical since for both of them $\omega \rightarrow 0$ as $\theta \rightarrow 0$. Actually T(0) corresponds to a rotation of the molecule about its axis (parallel to $\vec{c}$) whereas B(0), B(π) and T(π) correspond to simple translations. Of the remaining seven optical branches, Ib, Ss and As correspond to internal vibrations of the CH₂ radicals, which are equivalent to the bond stretching and bending modes found in the case of H₂O. Their frequencies are nearly constant, in contrast with the other optical branches which are strongly phase dependent as a consequence of the coupling between neighboring repeat units.

The force system leading to the described dispersion relations is of the valence type, namely forces which are proportional to the stretching of the C-C and C-H bonds or to the change of angle between consecutive bonds (C-C-C, C-C-H, H-C-H and C-C-C-C, the torsional angle defined by four consecutive C atoms). The force constants were obtained in part from measured infrared and Raman spectra which give the frequencies of the optical branches for $\theta = 0$ or $\theta = \pi$, and in part, from known force constants for some finite n-paraffins.

Another calculation with a somewhat different force system was performed by Tasumi, et al.⁽²⁴⁾ with results which are quite similar to the preceding.*

Since a frequency spectrum is defined as the fraction of modes per unit frequency interval, in the case of our non-interacting infinite chain model for polyethylene we simply have

$$f(\omega) = \frac{1}{9\pi} \sum_{j, \eta} \left| \frac{d\theta_{j\eta}(\omega)}{d\omega} \right| \quad (13)$$

*They used a Urey Bradley force system which includes central forces between unbonded atoms in addition to the valence force field.

where the sum is over all intercepts of all branches j having modes with frequency ω . Hence $f(\omega)$ becomes singular at all those points where $d\omega/d\theta = 0$. However, if the weak coupling between neighboring chains is considered, these peaks become broadened, the singular behavior at $\omega = 0$ disappears and the familiar ω^2 behavior typical of three dimensional crystals is recovered.*

The lattice property relevant to neutron scattering, however, is not the unweighted frequency spectrum $f(\omega)$. In fact, as is well known, the Fourier transform of $d^2\sigma/d\epsilon d\Omega$, which we called $\chi(\kappa^2, t)$ in Section II, is given in the incoherent approximation for the atom ℓ , by:

$$\chi_{\ell}(\kappa^2, t) = \exp \left\{ \sum_{\vec{q}, j} \frac{|\vec{\kappa} \cdot \vec{C}(\ell | \vec{q}_j)|^2}{2NM_{\ell}} g[\omega_j(\vec{q}), t] \right\} \quad (14)$$

where $g[\omega_j(\vec{q}), t]$ is given by eq. (3), $\vec{C}(\ell | \vec{q}_j)$ is the polarization vector (or modal amplitude) of the atom ℓ corresponding to the wave vector $\vec{q}$ and the branch j , N is the total number of unit cells and M_{ℓ} is the mass of atom ℓ .

Only for a cubic lattice with one atom per cell can this result rigorously be reduced to the much simpler form of Eq. (7). However Eq. (14) can still be somewhat simplified for polycrystals if the exact average over crystal orientation is replaced by averaging in the exponent.

Then

$$\chi_{\ell}(\kappa^2, t) \approx \exp \left\{ \frac{\kappa^2}{6NM_{\ell}} \sum_{\vec{q}, j} |\vec{C}(\ell | \vec{q}_j)|^2 g[\omega_j(\vec{q}), t] \right\} \quad (15)$$

*It might be interesting to point out that since the frequency spectrum for infinite uncoupled chains does not vanish at $\omega = 0$, the elastic cross section of such a system must be zero.

The polarization vectors satisfy the orthogonality and closure relations

$$\sum_{\ell} \vec{C}^*(\ell | \vec{q}_j) \cdot \vec{C}(\ell | \vec{q}_j) = \delta_{jj}, \quad (16)$$

$$\sum_j C_{\alpha}^*(\ell | \vec{q}_j) C_{\beta}(\ell' | \vec{q}_j) = \delta_{\alpha\beta} \delta_{\ell\ell'} \quad (17)$$

Hence if the ratio of the amplitudes corresponding to H and C atoms were constant for all $\vec{q}$'s and j 's, one could still reduce Eq. (15) to the form of Eq. (7). However, this is far from being correct, and unless a detailed calculation of the polarization vectors is available one must resort to rather crude approximations. The simplest assumption one can make is that the CH_2 move like rigid units in the modes involving skeletal deformation (T, B and S) and that in the remaining modes the C atoms are nearly at rest, which implies, invoking the symmetry between the two H atoms, that for these modes the square of the H atom amplitude $|\vec{C}(\text{H} | \vec{q}_j)|^2$ is a constant. The only branch for which this approximation is not adequate is mode S where skeletal deformation and CH_2 wagging are strongly coupled.⁽²⁴⁾ Disregarding this fact and making the described assumption we find:

$$R_H = \frac{|\vec{C}(\text{H} | \vec{q}_j)|^2}{|C(\text{H} | \vec{q}_{j'})|^2} = \frac{2}{13} \quad \text{with} \begin{cases} j = 1 - 3 \\ j' = 4 - 9 \end{cases} \quad (18)$$

Introducing this weighting factor into Eq. (15) and recalling Eq. (13) we finally recover the simple form of Eq. (7) by means of the weighted frequency spectrum⁽²⁶⁾

$$f_H(\omega) = \frac{1}{84\pi} \left[2 \sum_{i=1}^3 \sum_{\eta} \left| \frac{d\theta_{i\eta}}{d\omega} \right| + 13 \sum_{i=4}^9 \sum_{\eta} \left| \frac{d\theta_{i\eta}}{d\omega} \right| \right] \quad (19)$$

A coarse histogram representing Eq. (19) is shown in Fig. 25. From the same assumptions one obtains a second weighted frequency spectrum for scattering by C-atoms:

$$f_C(\omega) = \frac{1}{42\pi} \left[12 \sum_{i=1}^3 \sum_{\eta} \left| \frac{d\theta_{i\eta}}{d\omega} \right| + \sum_{i=4}^9 \sum_{\eta} \left| \frac{d\theta_{i\eta}}{d\omega} \right| \right] \quad (20)$$

As we said earlier, the frequency spectrum for unlinked chains is altered by the interaction between neighboring molecules in the frequency range extending from 0 up to values of $\hbar\omega$ of the order of the interaction energy. Theoretical calculations of the perturbed frequency spectrum were undertaken by several authors^(27, 28) and gave additional peaks between 0 and 0.025 eV corresponding to the fact that the branches T and B are each split in two and shifted in the neighborhood of $\theta = 0$ and $\theta = \pi$. The remaining branches are practically not affected by the perturbation. Only for two of the perturbed low frequency branches does ω vanish for $\theta \rightarrow 0$ and only for one of them does this happen for $\theta \rightarrow \pi$. This clearly corresponds to the existence of three acoustical branches in a regular three dimensional crystal. The perturbed spectrum obtained by Miyazawa and Kitagawa is shown in Fig. 26 for the range 0 - 0.062 eV.

The additional structure at low frequency introduced by the coupling between chains has been confirmed by neutron scattering experiments. One of the most interesting of these experiments⁽²⁹⁾ was performed on stretch oriented polyethylene. In this technique the microcrystals are lined up preferentially with their c-axis parallel to the stretching direction. This permits one, as is seen from Eq. (14) to excite independently modes with polarization vectors either parallel or perpendicular to the c-axis and should provide a good check on the correct assignment for the low frequency modes which are optically inactive. Unfortunately there still seem to remain some ambiguities which have not yet been explained.

Fig. 25--Frequency spectrum for polyethylene

Fig. 26--Miyazawa and Kitagawa's frequency spectrum for polyethylene
(low frequency range)

2. Comparison of Calculations and Measurements

Because of the large uncertainty of the correct frequency spectrum in the range between 0 and 0.025 eV, it is reasonable to treat this low frequency part, in the short collision approximation. In the histogram of Fig. 25 these low frequency modes have a total weight of 0.0345.

Using this modified phonon spectrum in GASKET^{*} yielded a total scattering cross section in good agreement with Armstrong's⁽³⁰⁾ measurements above initial energies of 0.01 eV, as seen in Fig. 27.^{**} For lower energies the calculated cross section starts deviating from experiment but not exceeding a difference of 10% with the average over experimental points. However, the experimental error in this range also increases. On the other hand, in view of the rather crude treatment of the low frequency modes, a much better agreement could hardly be expected. Nevertheless, better agreement was obtained in this energy region with an earlier calculation⁽²⁶⁾ in which the frequency spectrum of Fig. 25 was lumped into four discrete oscillators plus a δ -function at origin with weight $\frac{1}{14}$ (freely recoiling CH₂ radicals); this is also shown in Fig. 27.

There is some reason to believe that this better agreement of the lumped calculation in the low energy region is due to the fact that it gives more weight to the low frequency modes. This seems to be borne out by the comparison of predicted and measured double differential cross sections. Figures 28 through 33 show this comparison between measurements by Kirouac, et al.⁽²²⁾ and calculations with GASKET using the $f(\omega)$ of Fig. 25. It is seen that at least qualitatively most of the structure predicted by the theory is also found in the measurement, but in a considerably smoother form. Measurements by Whittemore⁽³¹⁾ confirm this conclusion. If more weight were to be given to the low frequency modes, it is clear that also the predicted structure would be smoothed out. On the other hand, it is also possible that some structure is lost in the experiment due to multiple scattering and resolution effects.

* The cross hatched area in Fig. 25 was approximated by two discrete oscillators of frequencies 0.182 and 0.36 eV.

** The experimental points are corrected for scattering by carbon.

Fig. 27--Scattering cross section for hydrogen in polyethylene

Fig. 28--Double differential scattering cross section for hydrogen bound in polyethylene: $E_0 = 0.232 \text{ eV}$, $\theta = 25^\circ$

Fig. 29--Double differential scattering cross section for hydrogen bound in polyethylene: $E_0 = 0.232$ eV, $\theta = 60^\circ$

Fig. 30--Double differential scattering cross section for hydrogen bound in polyethylene: $E_0 = 0.232 \text{ eV}$, $\theta = 90^\circ$

Fig. 31--Double differential scattering cross section for hydrogen bound in polyethylene: $E_0 = 0.41$ eV, $\theta = 25^\circ$

Fig. 32--Double differential scattering cross section for hydrogen bound in polyethylene: $E_0 = 0.41 \text{ eV}$, $\theta = 60^\circ$

Fig. 33--Double differential scattering cross section for hydrogen bound in polyethylene: $E_0 = 0.41$ eV, $\theta = 90^\circ$

Neutron spectra in poisoned polyethylene are also well predicted by scattering kernel obtained using the frequency spectrum of Fig. 25. As an example we show in Fig. 34 a calculated and measured neutron spectrum in 1% borated polyethylene.⁽²³⁾

Summarizing the preceding discussion we might say that although the present status of the theory is satisfactory for calculating integral quantities, more work is required to improve the fit of double differential measurements. The most straightforward of future improvements is to make a detailed calculation of the polarization vectors and to use Eq. (15) for the intermediate scattering function χ rather than the simplified form of Eq. (7).

IV. ZIRCONIUM HYDRIDE

The crystal structure of gamma phase zirconium hydride consists of a face centered cubic lattice of zirconium atoms with interstitial hydrogen atoms forming a second cube of side $\frac{a}{2}$ centered inside the zirconium cube of side a . In this arrangement each hydrogen atom is at the center of a tetrahedron formed by four zirconium atoms. Because of the large mass ratio of Zr to H (91:1) one would expect to find in the frequency spectrum of zirconium hydride a rather narrow optical band corresponding to the proton oscillating isotropically in the potential set up by its practically stationary nearest Zr neighbors. Such an optical band centered at 0.137 eV is actually observed in neutron scattering experiments but its width of about 15%⁽³¹⁾ exceeds by far the one associated with the simple model of a proton harmonically bound in a spherically symmetric potential due to nearest neighbors of mass 91. Several reasons have been suggested for this broadening such as H-H coupling, deviation from spherical symmetry or anharmonic effects. But none of these explanations is completely satisfactory. For instance the introduction of H-H coupling seems to contradict the experimental evidence⁽³²⁾ that the width of the optical band does not depend on the hydrogen concentration in ZrH_n .

Fig. 34--Neutron spectrum in an infinite medium of 1% borated polyethylene

Neutron scattering experiments further show that in addition to the optical band just mentioned the frequency spectrum of zirconium hydride has an acoustical band extending up to about 0.02 eV. Without having any further details of the lattice dynamics and since in the acoustical modes the H atoms essentially move together with their Zr neighbors, it is reasonable to assign a weight between $\frac{1}{90}$ and $\frac{1}{360}$ to these modes in a scattering law calculation. Thus for neutron less energetic than the optical phonons, the moderating properties of ZrH are very poor. This is clearly seen in Fig. 1.

Numerical computations were performed with the code GASKET using a phonon spectrum consisting of a Debye spectrum with cutoff at 0.02 eV and the two extreme weights mentioned above, plus a gaussian centered at 0.137 eV having a width of .02 eV at half maximum. Some of the results are shown in Figs. 1, 3 and 35 through 37. Whereas the scattering cross section is found to be rather insensitive to the weight of the acoustical modes, somewhat contradictory conclusions are reached from comparing experimental and theoretical values of the average scattering angle cosine⁽¹⁹⁾ and neutron spectra in borated ZrH_{1.75}⁽²³⁾. It is apparent, however, that both calculated spectra and average cosines show too much structure associated with optical multiphonon transitions. Scattering by Zr has been treated in the free gas approximation in all cases. From the preceding results it must be concluded, that although the described model for zirconium hydride is basically correct, further improvement is still required.

V. LIQUID HYDROGEN

1. The theory of neutron scattering by hydrogen at very low temperatures is becoming increasingly important in several fields, one of the most relevant being related to the design of nuclear rockets. Cold neutron research is another area of interest where the neutron slowing down properties of hydrogen at low temperature are important.

Fig. 35--Scattering cross section of hydrogen bound in zirconium hydride

Fig. 36--Average cosine of scattering angle for $\text{ZrH}_{1.85}$

Fig. 37--Infinite medium neutron spectrum in borated $\text{ZrH}_{1.75}$
(3.4 barns per H atom)

The hydrogen molecule is probably the simplest of all molecular scattering systems and hence one should be able to develop a theory essentially free of approximations. The results of this theory compared with experiment should thus yield a severe check of our understanding of neutron interactions with more complicated systems.

Since very little is known about the intermolecular forces in liquid hydrogen we consider the molecules to be completely free. Hence rigorously this theory only applies to a dilute molecular hydrogen gas. However, the results should also hold for the liquid phase, at least for neutron energies not too small compared to the intermolecular binding characterized by a Debye temperature of the order of 0.007 eV.

2. Review of the Theory

There are two features which make the hydrogen molecule a unique scattering system. The first one is its small moment of inertia, causing the rotational levels to be widely separated; and second there is the fact already mentioned that the incoherent scattering cross section is about 40 times larger than its coherent cross section. Due to the first of these two properties, the standard approximations made in scattering kernel calculations fail for H_2 . On the other hand, because of the smallness of σ_{coh} , we do not have to consider intermolecular interference scattering.

On account of the preceding considerations, the following features have been incorporated into the theory:

- a) Free rotations and translations of the H_2 molecules,
- b) Harmonic H-H bond stretching vibrations,
- c) Correlation between spins of the two protons in a hydrogen molecule.

The details of the calculations have been given in a recently published article.⁽³⁴⁾ Here we want only to point out the most relevant aspects of the theory. *

* References on previous work on neutron scattering by molecular hydrogen are given in (34).

First we discuss the effects of spin correlations. It is well known that the spins of the two protons can be either parallel or antiparallel corresponding to ortho- and para-hydrogen. The spin of the ortho and para states being respectively 1 and 0, the relative abundance of ortho- and para-hydrogen in thermodynamical equilibrium must be 3 to 1 at sufficiently high temperatures. At 20°K, however, 99.79% of the molecules are in the ground rotational state which is para. In fact, because of symmetry requirements all states with even angular momentum quantum numbers are para, whereas all odd states are ortho.

In the calculation of a double differential scattering cross section transitions of the initial system to all possible final states compatible with energy and momentum conservation contribute. An interesting result is that for para-hydrogen, the probability of transitions involving a change of spin is much larger than that for transitions conserving nuclear spin. In fact in this case, the spin conserving terms are proportional to the coherent hydrogen cross section, whereas the spin flip contributions are multiplied by the incoherent cross section which as we said earlier is about 40 times larger. On the other hand, when the initial state is an odd rotational state, that is in the case of ortho-hydrogen, it is the spin conserving transitions which are favored but only by a ratio of 2 to 1. This dependence on initial and final spin states is clearly shown by the following expressions for the double differential cross sections of para- and ortho-hydrogen:

$$\left(\frac{d^2\sigma}{d\Omega d\epsilon}\right)_{\text{para}} = \frac{k}{k_o} \sqrt{\frac{M}{\pi\kappa^2 T}} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega}\right)^n \sum_{J=0, 2, 4, \dots} P_J \left[a_c^2 \sum_{J'=0, 2, 4, \dots} + a_i^2 \sum_{J'=1, 3, 5, \dots} \right] \cdot$$

$$\cdot (2J'+1) \exp \left\{ -(\epsilon + \Delta E + \kappa^2/4M)^2 / \left(\frac{\kappa^2 T}{M}\right) \right\} \sum_{\ell=|J'-J|}^{J'+J} C^2(JJ'\ell; 00) |A_{n\ell}|^2 \quad (21)$$

$$\left(\frac{d^2\sigma}{d\Omega d\epsilon}\right)_{\text{ortho}} = \frac{k}{3k_o} \sqrt{\frac{M}{\pi\kappa^2 T}} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega}\right)^n \sum_{J=1, 3, 5, \dots} P_J \left[a_i^2 \sum_{J'=0, 2, 4, \dots} + (3a_c^2 + 2a_i^2) \sum_{J'=1, 3, 5, \dots} \right]$$

$$(2J' + 1) \exp \left\{ - (\epsilon + \Delta E + \kappa^2/4M)^2 / \left(\frac{\kappa^2 T}{M} \right) \right\} \sum_{\ell=|J'-J|}^{J'+J} C^2(JJ'\ell; 00) |A_{n\ell}|^2 \quad (22)$$

In these expressions, ϵ is the neutron energy transfer, k_0 , k the initial and final neutron momenta, M the proton mass, T the temperature, $|\vec{\kappa}|$ the neutron momentum transfer, P_J the statistical weight of the angular momentum state J , ω the level spacing of the vibrations (0.54 eV), a_c and a_i the coherent and incoherent scattering lengths respectively. Also $\Delta E = E_{J'} - E_J + n\omega$, where $E_J = 0.0147 J(J+1)/2$, $C(JJ'\ell; 00)$ is a Clebsch-Gordan coefficient, and

$$A_{n\ell} = \int_{-1}^1 d\mu \mu^n \exp \left(- \frac{\kappa^2 \mu^2}{8M\omega} + \frac{i\kappa a \mu}{2} \right) P_\ell(\mu), \quad (23)$$

in which a is the equilibrium separation of the two protons.

At temperatures of the order of 20°K practically all of the H_2 molecules are in the ground state with $J=0$. Hence only the first term in the sum over J in Eq. (21) contributes to $\left(\frac{d^2\sigma}{d\Omega d\epsilon} \right)_{\text{para}}$. Similarly at such a low temperature only the term with $J=1$ will contribute to $\left(\frac{d^2\sigma}{d\Omega d\epsilon} \right)_{\text{ortho}}$.

Although in a state of complete thermodynamical equilibrium, the relative concentration of ortho to para-hydrogen at 20°K is nearly zero, it still is possible to obtain (by fast cooling of a room temperature equilibrium mixture) large ortho concentration because of the smallness of the ortho-para spontaneous transition probability. However, it is important to notice that the scattering kernel obtained for the metastable moderator will not (and should not) satisfy detailed balance, which can only hold for the true equilibrium mixture of para- and ortho hydrogen.

The rotational energy levels of the H_2 molecule are expressed by the formula

$$E = 0.0074 J(J+1) \text{ eV}$$

Since we are assuming free translation of the molecules, the resonances shown by the differential cross section in correspondence with these levels, are Doppler broadened and shifted because of the considerable recoil energy taken up by the relatively light H_2 molecule. Since the recoil energy increases with the scattering angle, the rotational spectrum is best seen by looking at the small angle scattering cross sections. Figures 38 and 39, for instances show this structure for a scattering angle of 32° and initial neutron energies of 0.22 and 1.0 eV.

Figures 40 and 41 represent the cross section for the same initial energies but for a scattering angle of 55° . It is observed that because of the larger recoil, all peaks are broader and shifted towards smaller final energies. Coming back to Fig. 39 we notice that both for para- and ortho-hydrogen, the rotational peaks are superimposed on a much broader fundamental structure. This structure is the contribution from the zero and one phonon terms of the vibrational phonon expansion.

It is important to realize that for the exact treatment of the internal vibrations of the H_2 molecule, the vibrational matrix elements must be evaluated before averaging over molecular orientation. Once the rotational and vibrational matrix elements for a given initial state have been found, the average over orientation is done quantum mechanically by averaging over all initial rotational states. This essentially exact procedure leads to the functions A_{nl} of Eq. (23).

It should be mentioned that the only approximations involved in this calculation is the neglect of rotational vibrational coupling effects and the assumption that the restoring forces of the H-H bond stretching are harmonic in nature. Both assumptions are certainly justified up to 1 or 2 eV since the vibrational quantum is 0.546 eV and only 2/3 of the energy in the laboratory system is available in the center-of-mass system.

Fig. 38--Double differential scattering cross section of para- and ortho-hydrogen at $T = 20^\circ\text{K}$, $E_0 = 0.22 \text{ eV}$ and $\theta = 32^\circ$

Fig. 39--Double differential scattering cross section of para- and ortho-hydrogen at $T = 20^\circ\text{K}$, $E_0 = 1.0 \text{ eV}$ and $\theta = 32^\circ$

Fig. 40--Double differential scattering cross section of para- and ortho-hydrogen at $T = 20^{\circ}4\text{K}$, $E_0 = 0.22 \text{ eV}$ and $\theta = 55.2^{\circ}$

Fig. 41--Double differential scattering cross section of para- and ortho-hydrogen at $T = 20.4\text{K}$, $E_0 = 1.0 \text{ eV}$ and $\theta = 55.2^\circ$

3. Integral Properties of the Scattering Kernel

The most remarkable consequence of the spin correlations in the H_2 molecule is the peculiar energy dependence shown by the total scattering cross section of para-hydrogen. For very low neutron energies, the only contribution comes from spin conserving elastic scattering and according to our previous considerations this is rather small. But when the neutron energy reaches the threshold for exciting the first rotational level, a spin flipping transition becomes possible and the cross section suddenly rises from its minimum of about 1.6 to over 25 barns. This is seen in Fig. 42 which shows the calculated total cross section* of para- and ortho-hydrogen at $20.4^\circ K$. The experimental points shown on the curves are due to Squires and Stewart⁽³⁵⁾ and are in very good agreement with the calculations. It is seen that for large energies both cross sections approach the asymptotic free atom value of 20.4 barns after executing more or less pronounced oscillations due to the alternation of favored (spin flipping for para and spin conserving for ortho) and unfavored transitions.

The slowing down power $\xi\sigma_s$ defined in Section I shows the same structure as the total cross section as can be seen in Fig. 2. In the plot for para-hydrogen, the peaks corresponding to favored transitions are even more pronounced than those shown by σ_s . The three peaks appearing in Fig. 2. correspond to $J' = 1, 3$ and 5 , the threshold for exciting these levels being (in the lab system) $E = 0.0222, 0.133$ and 0.333 eV. The maxima of the peaks, however, are considerably shifted towards higher energies because of the predominant probability of collisions with large recoil energies. Another interesting feature is the large negative $\xi\sigma_s$ of ortho-hydrogen at very low energies, as compared with para-hydrogen. This is clearly due to the transition $J = 1 \rightarrow J = 0$ which leads to upscattering of the neutron.

* Because of a now corrected numerical error in the machine calculation of Ref. (34), all curves for ortho-hydrogen given in the present paper differ from those appearing in that reference.

Fig. 42--Scattering cross section for para- and ortho-hydrogen
at $T = 20.4\text{ K}$

The energy dependence of the average cosine of the scattering angle is represented in Fig. 4. Again it is seen that the curves for para- and ortho-hydrogen are quite different, although the overall trend of increasing anisotropy with increasing energy is common to both cases. Beyond 1 eV both curves approach the free atom value of $2/3$.

4. Thermal Neutron Spectra

A code for the IBM 7044⁽³⁶⁾ was written which calculates the P_0 and P_1 scattering kernels for para- and ortho-hydrogen from Eqs. (21) and (22). Both space dependent and space independent spectra were then calculated using these kernels. Some of the results are shown in Figs. 43 to 46. The space independent calculations for ortho-hydrogen were performed in the B_1 approximation using the code GATHER.⁽³⁷⁾ However, since convergence in a reasonable time could not be achieved for para-hydrogen media with $B^2 > 0.005 \text{ cm}^{-2}$ (probably because of the strongly fluctuating P_1 kernel), in this case, the so called B_0 option of the code was used, which replaces the P_1 kernel by a diagonal kernel giving the same integrated cross section. As a check, the B_0 and B_1 calculations were compared for an ortho-hydrogen medium with $B^2 = 0.04 \text{ cm}^2$. The comparison showed that the discrepancies are smaller than 3% at all energies, the B_0 calculation always being higher and thus slightly underestimating the thermal leakage.

Figures 43 and 44 show the results obtained respectively for para- and ortho-hydrogen in the buckling range $0 \leq B^2 \leq 0.1 \text{ cm}^2$. The two sets of spectra are compared with each other in Figs. 45 and 46, and it is seen that the difference between para and ortho spectra is remarkable, even in the case of infinite media ($B^2 = 0$). The reason for this is clearly that ortho-hydrogen is not a system in thermal equilibrium and hence that even for zero leakage and negligible absorption, the neutron spectrum will differ from a Maxwellian because of the considerable upscattering of low energy neutrons due to the unbalanced ortho-para ($1 \rightarrow 0$) transitions. For comparison, we also show in Figs. 45 and 46, the spectra calculated from a hydrogen free gas kernel at a temperature of 20°K .

Fig. 43--Neutron spectra in para-hydrogen
(bucklings B^2 are expressed in cm^{-2})

Fig. 44--Neutron spectra in ortho-hydrogen

Fig. 45--Comparison of neutron spectra in para-hydrogen, ortho-hydrogen and free proton gas ($B^2 = 0$)

Fig. 46--Comparison of neutron spectra in para-hydrogen
ortho-hydrogen and free proton gas ($B^2 = 0.04 \text{ cm}^{-2}$)

Because of its much larger total cross section at low energy, the leakage from ortho-hydrogen is much smaller than from a medium of para-hydrogen of the same size. The larger cross section also causes a better energy transfer, despite the important upscattering due to the $1 \rightarrow 0$ transition. As a result of all this, it appears that ortho-hydrogen would be a much better source of cold neutrons than para-hydrogen. This statement is clearly borne out by Fig. 46. Unfortunately, the inherent instability of ortho-hydrogen makes this advantage somewhat problematic.

Finally a few (one dimensional) space dependent spectra for para-hydrogen slabs were calculated using the discrete angle code GAPLSN⁽³⁸⁾ with group cross sections averaged over GATHER spectra of equivalent buckling. The scalar flux at the center and the angular flux for $\theta = 0^\circ$ at the surface of a 5-cm slab are shown in Fig. 43 together with the previously mentioned GATHER spectra for para-hydrogen.

In all the above spectral calculations, the poisoning taken was the natural absorption cross section of the proton.

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APPENDIX B

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RECENT DEVELOPMENTS IN INTEGRAL
NEUTRON THERMALIZATION*

by

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RECENT DEVELOPMENTS IN INTEGRAL NEUTRON THERMALIZATION

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Introduction

For a number of years extensive programs of measurement and theoretical analysis have been conducted in the various phases of integral neutron thermalization. These investigations have been undertaken at General Atomic and at other laboratories primarily to establish how well various average quantities characterizing the influence of given moderators on thermal neutron scattering can be predicted. There is a rather fine distinction between the areas of differential and integral neutron thermalization. Generally, in differential thermalization investigations, the full scattering process, namely the double differential scattering cross section $d^2\sigma/d\Omega dE$, is studied. In integral studies one or more integrals, including those over final energy and scattering angle, are taken (with various weighting functions) over the double differential scattering cross section. From the standpoint of reactor technology, it has often been advisable to compare calculated integral thermal neutron quantities with experiment to establish in an appropriate manner the adequacy of various approximate scattering descriptions employed in the many diffusion and transport theory codes now in use. Thus, for example, one would expect a scattering description for a moderator to give the correct total cross section (σ_T), the correct transport cross section (σ_{tr}), and the correct neutron spectrum under a wide variety of experimental conditions. In addition one can test other important properties of the theoretical scattering description by comparison with experimental thermal diffusion

constants (D), diffusion cooling constants (C), the second energy moment of the scattering cross section (M_2), and time dependent or asymptotic spectra.

In the integral thermalization program at General Atomic, the primary emphasis has been placed on measurement of neutron spectra in various moderators under conditions appropriate to reactor design, since this measurement tells quite rapidly whether a problem exists in describing the thermalizing properties of the moderator. In addition programs of measurement of total cross section (σ_T) and single differential scattering ($d\sigma/d\Omega$) have also been conducted to establish more firmly the adequacy of proposed theoretical scattering descriptions. From the single differential scattering cross sections, one can directly obtain the energy dependent transport cross section. Much of the work conducted under the General Atomic integral thermalization program is quite well known, so in the sections that follow it will be necessary only to summarize and reference the results. The main function of the following sections will be to present the status of the latest investigative techniques and the newest experimental results from our research.

Status of Spectral Measurements

Clean geometry measurements of thermal neutron spectra have been conducted routinely since 1953¹ at a number of laboratories. The measurements are usually performed in non-multiplying media, but check experiments in multiplying media have generally confirmed their adequacy.² These measurements serve a useful function in that they establish the accuracy² and adequacy of proposed thermal scattering descriptions for predicting reactor spectra. The reaction rates in various reactor materials (which determine the reactor performance) are given by the product of this reactor spectrum and the appropriate material cross sections. Thus one must have adequate methods of predicting the reactor spectra.

The most reliable spectral investigations conducted at General Atomic are being continuously compiled into a Spectrum Book (GA-5319).³ The total number of cases now exceeds 100. The primary purpose of this compilation is to allow other groups an opportunity to check their reactor design codes and data. The format of this compilation has been carefully standardized so that adequate information is available for the intercomparisons. Figures 1a and 1b show an example of this format in the EACRP version. The number of spectral investigations possible considering all the moderators, poisons, and geometries of interest is too prohibitive to study experimentally, so representative and

Date Submitted:

Status : Completed

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EACRP-L-62

NEUTRON SPECTRA

GA-Be
1ALISTING OF EXPERIMENTAL POINTS $\Phi(E)$ v. E

E(eV)	$\Phi(E)$	E(eV)	$\Phi(E)$	E(eV)	$\Phi(E)$
4.11396-02	4.399+05	6.08928-01	4.271+08	2.31429-02	1.043-10
1.74437-02	1.291+06	5.92276-01	4.810+08	2.07427-02	9.838+09
9.58784-01	2.619+06	5.28058-01	5.449+08	1.86319-02	8.939+09
6.05244+01	4.273+06	4.65734-01	6.234+08	1.67128-02	8.336+09
4.16496+01	6.309+06	4.06853-01	7.145+08	1.49783-02	7.982+09
3.04020+01	8.998+06	3.58448-01	8.352+08	1.34180-02	7.363+09
2.31639+01	1.197+07	3.18174-01	9.459+08	1.20194-02	7.052+09
1.82336+01	1.511+07	2.84308-01	1.097+09	1.07693-02	6.561+09
1.47249+01	1.915+07	2.52369-01	1.257+09	9.65427-03	6.274+09
1.21395+01	2.288+07	2.22700-01	1.514+09	8.66107-03	5.735+09
1.01797+01	2.748+07	1.97956-01	1.775+09	7.77725-03	5.062+09
8.65889-00	3.287+07	1.77106-01	2.138+09	6.99112-03	4.556+09
7.45504-00	3.784+07	1.57792-01	2.590+09	6.27916-03	4.217+09
6.48585-00	4.332+07	1.40062-01	3.184+09	5.63667-03	3.817+09
5.69407-00	4.833+07	1.25150-01	3.881+09	5.05958-03	3.340+09
5.03890+00	5.509+07	1.11548-01	4.744+09	4.54220-03	3.013+09
4.49063+00	6.118+07	9.92041-02	5.703+09	4.07976-03	3.007+09
3.82959+00	7.259+07	8.87926-02	6.670+09	3.66668-03	2.563+09
3.14509+00	8.969+07	7.93648-02	7.740+09	3.29838-03	2.348+09
2.62904+00	1.054+08	7.08504-02	8.920+09	2.96576-03	2.137+09
2.23035+00	1.231+08	6.32266-02	1.012+10	2.66642-03	1.893+09
1.91595+00	1.454+08	5.64045-02	1.111+10	2.39775-03	1.757+09
1.66365+00	1.682+08	5.03374-02	1.185+10	2.15711-03	1.684+09
1.46034+00	1.885+08	4.49377-02	1.235+10	1.93968-03	1.598+09
1.29632+00	2.142+08	4.01529-02	1.244+10	1.74383-03	1.524+09
1.15788+00	2.361+08	3.59059-02	1.225+10	1.56802-03	1.437+09
1.01472+00	2.720+08	3.21477-02	1.185+10		
8.74517-01	3.167+08	2.88146-02	1.161+10		
7.61443-01	3.662+08	2.58642-02	1.104+10		

EXPERIMENTAL ARRANGEMENT

<p>COMMENTS</p> <p>90° Angular neutron spectrum in beryllium, poisoned to 0.253 barns/Be atom (0.0253 eV) with 1% borated stainless steel at room temperature.</p> <p>Method of measurement: Time-of-flight measurements, using a pulsed neutron source.</p> <p>The absorption cross sections were obtained from references 4 and 5.</p> <p>The common normalization factor, which was used for GA-Be-1A to C, was obtained from the best fit of theory to experiment for these three cases in the energy range from 0.8 to 1.3 eV.</p>	<p>LAPLACIANS AT POINT OF MEASUREMENT</p> <p>$B^2 = 0.00840 \text{ cm}^{-2}$ (total buckling transverse to the source-assembly axis).</p> <p>Laplacian deduced from flux plots, using cadmium covered indium foils.</p>	<p>CALCULATION PARAMETERS</p> <p>Scattering kernel: GASKET (300°K, Be) code (1) Transport theory: GAPLSN code (2) Temperature: 0.0253 eV Source: DSZ code (3) Self shielding corrections applied for absorber.</p>
	<p>CORRECTIONS APPLIED TO DATA</p> <p>Detector sensitivity: Count loss correction. Mean emission time. Background subtraction. Flight path transmission.</p>	<p>ATOM DENSITIES IN THE ASSEMBLY</p> <p>Moderator: $1.1605 \times 10^{23} / \text{cm}^3$ (Be) Absorber: $3.6003 \times 10^{19} / \text{cm}^3$ (B) $9.9001 \times 10^{19} / \text{cm}^3$ (Ni) $1.5190 \times 10^{20} / \text{cm}^3$ (Cr) $5.1614 \times 10^{20} / \text{cm}^3$ (Fe) $6.236 \times 10^{20} / \text{cm}^3$ (Al)</p>
	<p>REFERENCES</p> <ol style="list-style-type: none"> 1. Beyster, J. R., et al., "Integral Neutron Thermalization," Annual Summary Report, October 1, 1964 through September 30, 1965, USAEC Report GA-6824. 2. Alexander, J. H., G. W. Hinman and J. R. Triplett, "GAPLSN, A Modified DSN Program for the Solution of the One-Dimensional Anisotropic Transport Equation," General Atomic Report GA-4972, March 1964. 3. Beyster, J. R., et al., "Integral Neutron Thermalization," Annual Summary Report, October 1, 1961 through September 31, 1962, USAEC Report GA-3542, p. 148. 4. BNL 325 5. Young, James A. and J. U. Koppel, Nucl. Sci. Eng. <u>19</u> (1964) 367 	

Figure 1a. Example of the EACRP format (Part 1) for presentation of thermal spectral data.

Figure 1b. Example of the EACRP format (Part 2) for presentation of thermal spectral data.

even extreme test cases are usually investigated. Specifically the comparisons of measured and calculated thermal neutron spectra provide checks of the following:

1. The thermal neutron scattering kernel.
2. Cross section data.
3. Transport approximations in reactor codes.
4. Numerical problems in the codes, such as the selection of the spatial, angular, and energy meshes.
5. Source descriptions of the thermal problem.
6. Discontinuities at the joining energies between fast and thermal calculations.
7. Coherent scattering phenomena.

Infinite Medium Spectra

The first class of spectral investigation which one undertakes generally is that of infinite or quasi-infinite medium neutron spectra. One usually includes here spectral investigations in which the spatial dependence of the neutron flux is small so that the leakage can be handled by the inclusion of a localized buckling. In general, this implies hydrogenous systems due to the inherently low L^2/τ ratio. For other moderators it is difficult to reduce L^2 sufficiently by poisoning yet retain sufficient neutrons in energy regions where binding effects are important. It is not possible to summarize in a few words the status of these spectral studies. Thus in Table 1 an attempt is made to indicate the degree of predictability of the spectra for several hydrogenous moderators under various conditions. The scattering kernels used are also given. To determine the extent of the disagreement, the theoretical and experimental spectra are first normalized together in the slowing down region at about 2 eV. The maximum discrepancy between the two curves above the thermal peak is then noted. One can always find larger disagreements than those noted here by looking at the lowest neutron energies. However, due to the existence of detailed balance and neutron conservation restraints on the spectra, the discrepancy at low energy is usually associated with the same disagreement at the higher energies.

There are some important observations to be made from the information presented in this table:

1. The residual disagreements between theory and experiment shown in Table 1 do not appear to be large enough to seriously affect the prediction of reactor parameters which depend on a knowledge of these infinite medium spectra. Obviously all spectral examples of interest have not been studied, so no completely general statement is possible yet.
2. One finds that more spectral studies exist for H_2O than

Table 1. Disagreement Between Theory and Experiment for Quasi-Infinite Medium Neutron Spectra

	H ₂ O	ZrH _x	CH ₂	C ₆ H ₆
1/V absorber - ambient temperature	10%	25%	10%	10%
1/V absorber - elevated temperatures	15%	20%	---	---
1/V absorber - low temperatures	---	---	10%	---
Res. absorber - ambient temperature	Er-15% Gd-10% *Cd-18% *Sm-15%	---	---	---
Res. absorber - elevated temperatures	Er-15% Gd-5%	---	---	---
Scattering kernel	GASKET FLANGE	SUMMIT	GAKER SUMMIT	SUMMIT

*Nelkin model.

for any other moderator, which reflects its importance in reactor design. A comparable theoretical effort has been made to refine the molecular model for H₂O by including either the anisotropic vibrations of the hydrogen atoms⁴ or the rotational band structure for the molecule.⁵ The residual discrepancies have diminished considerably due to the physical refinements now incorporated in the models, but they still persist and are consistently in the same direction. In Figure 2 the ratios of the experimental to theoretical spectra in erbium poisoned water are given for two theoretical models. (The ratio of the two theoretical spectra is also given.) The experimental spectrum is generally harder than the theoretical. The explanation of this effect, regardless of the magnitude of its significance to reactor design, is certainly worth understanding. It is not impossible in the case of resonance absorbers that some of the discrepancies encountered are due to inaccuracies in the available absorption cross section data.

3. Spectra in zirconium hydride are quite interesting, since the presence of the Einstein harmonic oscillator levels at 0.13, at 0.26, and even at 0.39 eV can be seen. The calculation of the scattering kernel for this moderator presents large numerical problems, and these to some extent limit the comparisons. The disagreements, although rather large for these

Figure 2. Comparison of experimental and theoretical neutron spectra in erbium positioned H_2O at room temperature.

simple cases, occur near the oscillator levels and indicate clearly that our present model⁶ with the broadened level at 0.13 eV is not quite adequate.

Spectra in Finite Media

Over a period of years a large number of space dependent spectral studies have been conducted. Generally these have been carried out in nearly homogeneous media where self-absorption in poisoned foil material (if any) could be easily taken into account. The space dependence often has been deliberately introduced by setting up essentially a vacuum interface, by change of medium or absorption, and by temperature discontinuities. In addition, a spatial dependence arises naturally because of source location for nonhydrogenous systems. In principle measurement of spectra under these conditions, if outright heterogeneity is avoided, should not be much more difficult or subject to error than measurement of infinite media spectra. In practice very interesting anomalies have occurred.

The calculation of space dependent neutron spectra is performed as follows at General Atomic. The scattering kernel is generated by means of one of the standard computer codes listed in Tables 1 and 2. (Ultimately these kernels will all be generated using the new GASKET-FLANGE⁷ system.) The average P_0 and P_1 Legendre moments are then calculated using the scattering kernels in the code GATHER-II⁸ for the particular energy group structure desired. For an accurate calculation of the thermal spectra, one should know accurately the spatial and angular distribution of the thermal source produced by the epithermal or fast neutrons. This source distribution is presently specified as input to the transport theory calculation with the code DSZ.⁹ This code determines the P_0 and P_1 source contributions, though not to the accuracy we would like. The spectrum of the angular flux is next calculated by GAPLSN,¹⁰ a one dimensional S_N transport theory code, using the group cross sections and the distributed source. $D(E)B^2$ terms are added to the group absorption cross sections to account for leakage in directions normal to the direction of the S_N calculation. The disadvantage factors for the cross section averaging are obtained from a cell calculation employing the GAPLSN code.

The status of theory-experiment comparison for some non-hydrogenous moderators has been summarized in Table 2. The finite medium conditions were introduced primarily by the neutron source location and the existence of a relatively large value of L^2/τ in the medium. Some observations on the status of the data in this table are:

1. The agreements between theoretical and experimental

Table 2. Accuracy Of Mildly Position-Dependent Neutron Spectra

	D ₂ O	Be	BeO	C
1/V absorber - ambient temperature	5%	5%	15%	5-30%
1/V absorber - elevated temperatures	---	---	---	5%
Res. absorber - low temperatures	---	---	---	10%
Res. absorber - ambient temperature	Cd-5%	---	---	Sm-10%
Res. absorber - elevated temperatures	---	---	---	Sm-10%
Scattering kernel	GAKER	GASKET	SUMMIT	SUMMIT
		FLANGE		

spectra for the moderator D₂O seem to be the best. This occurs even though the D₂O scattering kernel used is calculated in the incoherent approximation¹¹ and the rotational degrees of freedom are represented by a discrete oscillator rather than a band of frequencies. The residual disagreements noted here are less than the quoted experimental uncertainties.

2. Spectra measured in poisoned beryllium have been found to agree quite well with the calculated spectra.¹² Some examples are shown in Figure 3. The calculational model employed is based on the incoherent approximation,¹³ although beryllium is a coherent scatterer. In fact, for beryllium one can calculate spectra surprisingly well for heavily poisoned media using the free gas model with a moderator mass of 9.¹⁴ This observation does not appear to agree with that of Poole, Nelkin, and Stone, who reported theory-experiment comparisons in heterogeneous multiplying media.¹

3. The case of carbon recently has become complicated. It will be noted from the first column of Table 2 that both 5% and 30% discrepancies have been obtained. The older measurements¹⁵ at a poison concentration of 0.4 barns per carbon atom differed by less than 5% with theory, while newer measurements¹⁶ at over 1 barn per carbon atom disagree markedly. The reason for this situation is not presently understood.

Two cases will be discussed to show the status of theory-experiment comparison in situations where spatial dependence has been deliberately introduced.

Case I. Generally the objective here is to study scalar and

Figure 3. Axial spectral measurements in boron poisoned beryllium.

angular flux spectra under conditions imposed largely by the geometrical conditions. Thus the simple geometry shown in Figure 4 was selected for the study of position dependent spectra for water. The tank was 12 in. by 12 in. by 4 in. thick, and the water was poisoned to 5.15 barns per hydrogen atom. The spectra measured in the gradient direction are shown in Figure 5. P_0 only and P_0 plus P_1 calculations of the spectra are given using the Nelkin model.¹⁷ Spectra using the Radkowsky prescription¹⁸ are also shown. It is apparent that the P_1 scattering must be included. It is also apparent that the Nelkin model, although very adequate inside the medium, does not give good agreement for the leakage spectra. This is mainly a matter of an intensity rather than spectral shape discrepancy. It is, however, a discrepancy of some 30%, and attempts should be made to resolve it first by performing a higher order S_N calculation including up to P_3 scattering with a much finer spatial mesh near the surface.

Case II. In another series of one dimensional spectral measurements for some solid moderators, the general experimental arrangement in Figure 6 was used. The arrangement was such that both a temperature and poison discontinuity could be introduced. Several configurations were studied with the region next to the neutron source being either lightly poisoned beryllium or graphite (as shown). The region farthest from the source was usually graphite, quite heavily poisoned either homogeneously or heterogeneously. Typical results showing some of the difficulties of interpretation are given in Figure 7. It is apparent that the spectra in the hot, heavily poisoned graphite are not in good agreement with theory. Further attempts to resolve the difficulties have led to other, even more serious problems, especially in cases where a graphite core (poisoned region) and beryllium reflector (unpoisoned region) have been combined.¹⁶

Generally one finds in space dependent spectral studies that considerably more care must be exercised to obtain a meaningful experimental result than in studies of quasi-infinite media neutron spectra. Even so, sizeable discrepancies still remain to be resolved.

Spectral Studies in Heterogeneous Media

The problem most often facing the reactor designer is that of specifying spectral dependences in heterogeneous configurations of fuel and moderator. Until rather recently this problem has not appeared to be amenable to precise study by time of flight spectral techniques. However it appears now that techniques for neutron beam extraction without much perturbation of the cell spectrum are within our grasp and that many problems of

Figure 4. Experimental arrangement for spectral measurements in H_2O .

Figure 5. Position dependent neutron spectra in borated water (5.15 barns/H).

Figure 6. Experimental arrangement for spectral measurements across a poison and temperature discontinuity in graphite.

Figure 7. Neutron spectra at an interface dividing cold, mildly poisoned graphite from hot graphite heavily poisoned with boron and samarium.

practical significance can thus be studied. In our program we have achieved some success with both cylindrical and slab geometry configurations of moderator and poison.

The program of measurements in cylindrical geometry has been conducted in the setup shown in Figure 8. The environment of a cylindrical fuel element can be viewed by one of two probe tubes. One tube penetrates the element while the other tube looks at the spectra as a function of distance from the fuel element. Various reentrant hole sizes have been used in both situations, and we find little dependence of the spectral shape on the size of the reentrant tube as long as it is small compared with the neutron mean free path. Typical spectra measured in and around a $\text{ZrH}_{1.67} - \text{U}^{235}$ fuel rod immersed in a cylindrical tank of cadmium poisoned D_2O are shown in Figures 9 and 10. As can be seen, the zirconium hydride and uranium are still influencing the neutron spectra even at 5.58 cm from the tank center since previous measurements in this heavily poisoned D_2O show no peaking of the spectra below 0.2 eV. The effect of the 6.3 eV resonance in U^{238} can clearly be observed. It would be very valuable if the energy range and resolution of the measurements could be extended to cover the resonance region up to 300 eV.

A typical example of the theory-experiment comparison obtained can be seen from the geometry of Figure 11, where spectra in a one dimensional slab configuration of alternate 1-in. layers of pure and 1.3 wt-% borated polyethylene have been studied. Again, a wide variety of reentrant tube sizes were investigated. The final data were taken with a 0.25-in.-diameter reentrant tube.

The spatial distribution of the indium resonance flux across the assembly was measured by activating cadmium covered indium foils. These plots allowed the local buckling, B^2 , to be determined in the vicinity of the spectral measurement region. The calculations of the neutron spectra were performed in the usual way. The Koppel scattering kernel for polyethylene¹⁹ was used to generate P_0 and P_1 group cross sections by means of the GATHER⁸ code. An infinite slab cell problem with reflective boundary conditions was then performed by means of the GAPLSN¹⁰ code in an S_{16} approximation. A spatially flat source was used across the cell with an energy dependence calculated by the DSZ code.⁹

The vector flux from the first or sixteenth angular interval is compared with the experimental values at the corresponding position in Figure 12. The comparison of measured and theoretical spectra indicates a very satisfactory agreement. The whole structure of the spectral variation across the cell can be calculated and measured without resort to any renormalization

Figure 8. Geometry for fuel and moderator spectral measurements.

Figure 9. Measured neutron spectra across a $\text{ZrH}_{1.67}$ -U fuel element surrounded by cadmium poisoned D_2O .

Figure 10. Measured neutron spectra in cadmium poisoned D_2O surrounding a $ZrH_{1.67}$ -U fuel element.

Figure 11. Experimental arrangement for spectral measurements in a polyethylene cell.

procedure. The results shown here are useful not only because they confirm the adequacy of the polyethylene kernel of Koppel,¹⁹ but also because they indicate that the analytical and experimental methods are in good shape for this type of problem.

Two further GAPLSN calculations were performed in this polyethylene cell. In one, the P_1 scattering cross sections were set to zero, which changed the fluxes as shown in Figure 12. Differences are observed which appear small on the logarithmic scale of the figure, but the ratio of the neutron absorption rate in the borated polyethylene to that in the pure polyethylene shows significant differences. With $P_0 + P_1$ scattering, the ratio is 8.053, and with P_0 scattering only, the ratio is 7.345, which is a difference of almost 10%. In the second GAPLSN calculation, a P_0 transport approximation was used. In this calculation the total cross section Σ_T is replaced by the transport cross section Σ_{tr} , and the ingroup scattering cross section is reduced by the difference between Σ_T and Σ_{tr} . This approximation shows flux differences that are at most only 2% different from the calculation with full $P_0 + P_1$ scattering. This result is consistent with the observations of Cohen,²⁰ who also demonstrated that the transport approximation was quite accurate. It is to be noted in passing that the experiment is sufficiently sensitive to the inclusion of P_1 scattering.

Figure 12. Neutron spectra across a cell comprising 1 in. pure and 1 in. 1.3% wt. borated polyethylene.

From these heterogeneous studies, it would appear that we possess the techniques for making direct spectral measurements in heavily absorbing fuel rods of about 1/2-in. diameter located in assemblies of low multiplication. It would be desirable to pursue these studies in order to clear up remaining discrepancies in the determination of cell spectra. (See Section 9 of Reference 7.)

Status of Single Differential Scattering Investigations

It has been apparent for some time that the interpretation of measured spatially dependent thermal flux spectra is often rather ambiguous. This fact nonetheless does not decrease the significance of predicting the vector flux accurately in typical reactor calculations. It has therefore become desirable to develop procedures for checking various important properties of the neutron scattering kernels, especially the angular properties. One would, for example, like to be assured from auxiliary experiments that the transport cross sections $[\sigma_{tr}(E)]$ are given properly by the theoretical scattering models. If the differential scattering cross sections can be correctly predicted by theory, one has at least narrowed the reason for theory-experiment spectral discrepancies down to one of the other six problem areas listed in the previous section plus, of course, the possibility of experimental error.

From the standpoint of a thorough check of the scattering description, it is desirable to have the complete double differential scattering cross section measurement. From the pragmatic point of view, however, these measurements are far from complete and the number of really reliable measurements is not plentiful. At this stage it appears that the sum total of all double differential measurements available for any moderator will not be of much help in settling the problem of the predictability of angular flux spectra. Unfortunately, certain gross liberties are routinely taken in the analysis of double differential scattering measurements which make suspect the accuracy of the reported data. The most serious of these liberties is the lack of a multiple scattering correction. It has thus been decided both at General Atomic and at Munich²¹ that a simpler experiment, which nonetheless is sensitive to the desired angular properties of the scattering kernel, would aid in the solution of certain problems in reactor technology. Therefore, a number of measurements of $(d\sigma/d\Omega)$, the single differential scattering cross section, have been completed and reported.^{22,23} In the remainder of this section the current status of such theory-experiment comparisons will be discussed.

$d\sigma/d\Omega$ for H_2O

The experiments which have been completed are measurements of the single differential scattering cross section ($d\sigma/d\Omega$) of H_2O for some 55 neutron energies from about 0.005 to 10 eV for thin water samples. Supplemental experimental studies of the multiple scattering from the water samples have also been conducted. The experimental data have all been corrected for the multiple scattering in the samples except for energies above 2.2 eV. The need for this correction has been carefully established, and the calculations of the multiple scattering correction have been made using two differential scattering models for H_2O (i. e., the Nelkin water model¹⁷ and the Koppel⁵ version of the Haywood model).²⁴ Additional studies of the importance of the multiple scattering correction have been made using our general purpose multiple scattering computer code MUSE.²⁵ The corrected experimental data have been compared with the theoretical predictions of the Nelkin model and the Haywood-Koppel model as computed by the GASKET-FLANGE⁷ codes. Improved agreement has been obtained with the new scattering description.

Some examples of the current status of theory-experiment comparisons for water are shown at selected energies in Figures 13, 14, and 15. At the lowest energies shown, about two-

Figure 13. Angular scattering by H_2O of 0.0205-eV neutrons.

Figure 14. Angular scattering by H_2O of 0.0652-eV neutrons.

Figure 15. Angular scattering by H_2O of 0.850-eV neutrons.

thirds of the theory-experiment discrepancy is removed by representing the H_2O rotational motion as a distributed frequency band instead of a discrete oscillator.⁶ The X's are calculated scattering cross sections including anisotropic vibrations, retaining the discrete torsional oscillator. The influence of combining the two refinements into one model is not yet known but will be investigated in the future. One can see in Figure 15 that as the neutron energy increases, all the disagreement between theory and experiment disappears. The disagreement around 0.06 eV has been the predominant disagreement evidenced by theory until the present time. In fact if one includes anisotropic vibrations, it is possible that a disagreement in the other direction may occur. This is by no means certain, however, since the two improvements are somewhat equivalent. The experimental errors shown are the statistical errors only and have been obtained directly from the raw data without regard to the fact that some smoothing, due to the profusion of measured energy points, has occurred in establishing the value plotted. Thus the errors shown are probably too large by a factor of 1.5 to 2 if only counting statistics are considered. On the other hand it has not been clear until recently how well the systematic corrections could be made to the experimental data.

The measurements above 1 eV agree well with the water model predictions up to 2.2 eV. Above 2.2 eV disagreements still exist, but these data are not yet properly corrected for multiple scattering. At energies from 0.85 to 2.2 eV the calculated single differential scattering cross sections have been determined in two ways: (1) with the Nelkin model, and (2) by a free gas model of mass 1 at an effective temperature $\bar{T}$ of 0.117 eV. Both results agree.

$d\sigma/d\Omega$ for D_2O

Initial measurements of the single differential thermal neutron scattering cross section for heavy water have been completed and compiled for 55 neutron energies ranging from 0.006 to 10 eV for two sample configurations having a transmission of about 90% and 80%. These new experimental data have been compared with the experimental measurements of Springer²¹ and the theoretical calculations of $d\sigma/d\Omega$ performed by Koppel²⁶ using essentially Butler's formulation²⁷ of the double differential scattering cross section for D_2O . Generally the results of this experiment do not agree with those of Springer as well as in the case of scattering from H_2O . The angular dependence shows the pronounced effect of coherent scattering. The cross sections have been calculated including coherent effects in both intramolecular scattering and intermolecular scattering.

The theoretical predictions of cross sections at angles less than 15 to 20 degrees are at present still subject to some error. Measured interference peaks are generally somewhat less pronounced than those observed in Springer's experimental work²¹ but have the same characteristic deviation from the theoretical peaks, namely the experimental peaks are narrower.

The nature of the discrepancy between the present data and those of Springer is shown in Figure 16. The peak cross section at the first diffraction maximum is plotted versus neutron energy. The cross section from coherent scattering theory is also given. One might expect to observe a cross section which is "relatively" energy independent, because under certain circumstances the magnitude of the cross section depends on the momentum transfer and this is the same for all energies at the first diffraction maximum. A possible source of trouble with our first experiments was thought to be the 5° angular resolution employed. The differences shown in Figure 16, however, would be hard to explain on this basis, since the diffraction peaks are very broad at low energies and yet these peak values are in better agreement with the higher energy data than with those of Springer. An angular resolution correction factor can also be developed to correct the data approximately, and this indicates only a small difficulty with resolution.

To clear up the problem of the peak cross sections, a high angular resolution experiment has been performed on D₂O. Solar slits made of highly absorbing material have been prepared to limit the angular aperture to less than 1°. Data on the scattering have been taken in 5° intervals over the first diffraction maximum for D₂O. Low and high angular resolution data were simultaneously taken for the scattering sample over the entire energy range 0.006 to 10 eV. The point by point agreement is better than 10% everywhere, so angular resolution would not seem to be the cause of the disagreement with Springer. Much further analysis of our data is necessary, but it generally appears that the Springer²¹ data are in disagreement with both theory and our experiments on the magnitude of the first diffraction maximum.

A nice feature of the present D₂O experiments is the fact that the diffraction pattern at small angles is sensitive to the pair correlation function between D₂O molecules. In Figure 17 the angular distribution for low energy scattered neutrons is shown for both theory and experiment. The coherent scattering model used employs a step pair correlation function which is not very realistic. It will be extremely interesting to see what shape of the pair correlation function is required to fit the measured data.

Figure 16. Peak cross section of D_2O at the first diffraction maximum.

Figure 17. Angular scattering by D_2O of 0.00632-eV neutrons.

$d\sigma/d\Omega$ for $\text{ZrH}_{1.85}$

The presence of a single well defined optical mode of vibration of the hydrogen in the zirconium lattice is well known. Acoustical modes have low dynamical weight for scattering and a low cutoff energy; hence one would expect that they could be included without difficulty. However, more experimental anomalies exist in our understanding of neutron thermalization for zirconium hydride than for the more complex moderators. Integral experiments on reactor temperature coefficients also tend to support divergent viewpoints. Our previous spectral results,⁶ however, could be explained by a rather simple phenomenological model for zirconium hydride, basically that of the Einstein oscillator. We have therefore tried to obtain further clean experimental results to try to resolve the anomalies. It is, for example, possible that the simple natural width model⁶ could predict the P_0 behavior of the scattering kernel properly and give spectral agreement but predict the transport behavior incorrectly. Thus we have concentrated on experiments to investigate the reliability of the model in determinations of angularly dependent or transport quantities.

The single differential scattering cross sections have been measured using a thin, pressed powder sample of $\text{ZrH}_{1.85}$ for 55 neutron energies from 0.006 to 10 eV. The multiple scattering correction factor, C_M , was significant as in the case of H_2O (see Figure 18). The differential elastic and inelastic scattering cross sections used for $\text{ZrH}_{1.85}$ in the MUSE²⁵ code are also used to calculate the theoretical single scattering cross section ($d\sigma/d\Omega$). Six selected angular distributions are given in Figure 19. Statistical counting errors are less than 5% at all points shown. The solid curves are the theoretical calculations, and the three dashed curves represent the experimental work of Springer, *et al.*²¹ It will be observed that both sets of experimental points tend to show fine structure. This is due to the coherent scattering from zirconium and cannot be expected to be the same in both experiments unless the experimental angular resolutions are the same. Further, Springer's data consist of angular distribution shape measurements for $\text{ZrH}_{1.91}$, not $\text{ZrH}_{1.85}$, and have not been adjusted for the slight hydrogen difference in the comparison shown. It is apparent from Figure 19 that the experimental $\bar{\mu}$ will be less than that given by the theoretical calculation. This is consistent with Springer's findings. In addition, the decrease (calculated by Memmert²⁸) in $\bar{\mu}$ as the first level in zirconium hydride becomes excited by inelastic neutron scattering is clearly observable. One can see this from a comparison of the more forward peaked 0.105-eV curve with the 0.153-eV curve in Figure 19.

Figure 18. Multiple scattering correction factors for a 39-mil-thick sample of $ZrH_{1.85}$ placed at 45° to the incident neutron beam.

Figure 19. Calculated and measured single differential scattering cross section of $\text{ZrH}_{1.85}$ for various incident neutron energies.

It will be recalled that our most recent model⁶ for zirconium hydride uses a band of harmonic oscillator frequencies with a mean energy of 0.14 eV and a 0.025-eV width. A Debye-type acoustical frequency spectrum cutting off at 0.02 eV with a weight of 1/90 is also used. Figure 19 is different from that reported at Karlsruhe,²² since the calculational results have been improved. Generally the agreement (between theory and experiment) seems rather good, again indicating that the basic harmonic oscillator model for zirconium hydride is not greatly suspect. Experiments reported by Leonard²⁹ on the angular distribution for double differential scattering at neutron energies to 0.4 eV indicate the same fact. In Figure 20, $\bar{\mu}$ has been plotted for the present experiments, the Springer experiments,²¹ the Memmert calculations,²⁸ and the natural width model calculations. Here we begin to see problems with the natural width model in that the predicted variation of $\bar{\mu}$ with energy is much more than that observed experimentally. The Memmert calculation was performed for the very idealized case of an unbroadened Einstein oscillator model for zirconium hydride.

Figure 20. Calculated and measured $\bar{\mu}$ for $\text{ZrH}_{1.85}$

Single differential scattering data from $\text{ZrH}_{1.85}$ at elevated temperatures have been measured for comparison with the predictions of the broadened Einstein oscillator model⁶ at temperatures more appropriate to reactor operating conditions. Pressed powder samples of zirconium hydride approximately 40 mils. thick and canned in 5-mil aluminum were used. Single differential scattering measurements were made at temperatures of 260°C and 483°C for comparison with data taken previously²² at room temperature.

Reduction of the 260°C data has been performed using the methods outlined in Reference 2. Multiple scattering corrections at elevated temperatures have not yet been computed, so room temperature multiple scattering correction factors have been employed for the data presented here. Figures 21 and 22 show comparisons of angular scattering cross sections at two energies at both 260°C and room temperature. The data at 483°C are presently being analyzed. It can be seen that there is little change from room temperature to 260°C . However, the multiple scattering corrections for 260°C may modify these results when they are applied.

Status of Total Cross Section Measurements

Measurements of total cross sections for the reactor moderators antedate all other types of integral measurements. These measurements, when performed precisely, serve as an excellent check on both the predictions of the theoretical scattering kernels and the more differential experiments, such as the single and double differential scattering experiments. The effect of chemical binding is still quite evident in total cross sections. At relatively high thermal energies the asymptotic expansion of the total cross section actually behaves as $\sigma_T = \sigma_{fa} [1 + (\bar{T}/2mE)]$, where $\bar{T}$ is the well known average kinetic energy per molecule and is very model sensitive. In addition the effect of elastic and inelastic coherent scattering can often be observed in appropriate energy regions.

Relatively few total cross section measurements have been performed in the General Atomic thermalization program, but increased attention in this area now seems warranted because of the general quality of the data in existence. Until the present time it has perhaps not been desirable to attempt a precision comparison between theory and experiment for the total cross section, but with the development of accurate computational methods such as GASKET-FLANGE,⁷ the comparisons seem to be justified. In this section we will briefly review the status of the existing theory-experiment comparisons of total cross sections for common moderators.

Figure 21. Angular scattering cross sections of $\text{ZrH}_{1.85}$ for 0.044-eV neutrons.

Figure 22. Angular scattering cross sections of $\text{ZrH}_{1.85}$ for 0.105-eV neutrons

$\sigma_T(\text{H}_2\text{O})$

The most recent total cross section measurements for H_2O are those of J. Russell.³⁰ These measurements appear to be quite precise with an experimental error of perhaps 0.3% to 0.5% per point. The most realistic calculation of the water total cross section is that using GASKET-FLANGE⁷ and the Koppel-Haywood⁵ frequency spectra. The comparison is shown in Figure 23. A real discrepancy of 5% still appears to exist near 0.03 eV. The asymptotic behavior of the measured cross sections, however, is quite consistent with the $\bar{T} = 0.117$ eV of the Nelkin or Koppel-Haywood determination.

$\sigma_T(\text{D}_2\text{O})$

In the discussion of spectra in finite media, it was concluded that most measured thermal spectra in D_2O could be accurately calculated by considering only incoherent scattering. The subtleties of the coherent scattering process for D_2O are clearly evident in the single differential scattering, and distinct discrepancies are observed. In Figure 24, the total cross sections calculated by Koppel,²⁶ who used essentially the formulation of Butler,²⁷ are compared with measured data from BNL-325. No real problems appear to exist above 0.002 eV. The two determinations agree to about 5%, which may still be somewhat outside of experimental errors.

$\sigma_T(\text{ZrH}_x)$

The total cross section for ZrH_x has been measured over a very limited energy range only. A comparison of the existing experimental data within the predictions of the theoretical models is shown in Figure 25. The experimental data are simply not adequate for precise comparison with the theoretical calculations. It is not clear, for example, that the discrepancies which are apparent here bear any resemblance to those evident in $\bar{\mu}$ in Figure 21, since the later experimental results would seem to indicate a need for further broadening of the oscillator level.

$\sigma_T(\text{Graphite})$

The most recent measurements of the total cross section of carbon were made by Walton.¹⁶ These measurements and the calculated total cross sections from GASKET-FLANGE are shown in Figure 26. In view of the obvious dense diffraction pattern, the comparison of theory and experiment is relatively

Figure 23. Calculated and measured total cross section for H_2O .

Figure 24. Total scattering cross section of D_2O .

Figure 25. Total cross section of zirconium hydride.

Figure 26. Measured and calculated scattering cross sections of graphite.

good. A problem exists, however, in making such a comparison, because the experimental total cross section samples are not necessarily completely polycrystalline (as the theory assumes). Thus some orientation due to fabrication can occur, and one must weight properly the cross sections measured with different sample orientation. Further work to resolve these remaining problems would seem to be warranted.

$\sigma_T(\text{Be})$

The measured and calculated total cross section of beryllium based on Young's lattice frequency spectrum¹³ are compared in Figure 27. The three curves shown were calculated either with the SUMMIT³¹ or GASKET⁷ code and using different energy group structures. It became evident that the energy mesh should be limited to differences of less than $2\beta_{\text{max}}$, or about 0.015 eV, since the kernel generating codes would not rigorously handle larger energy transfers. The lowest curve of Figure 27 does not satisfy this criterion and displays nonphysical oscillations that are not observed in the measured values or in the upper two curves, which do match this criterion.

The agreement of theory with experiment for the upper two curves is fairly satisfactory except below the Bragg cutoff. It is to be noted that the theoretical curve is greater than the measurement. This, in turn, is related to the complete failure of the incoherent approximation at these neutron energies.

$\sigma_T(\text{BeO})$

The calculated total cross section for BeO is compared with the experimental results of BNL-325 in Figure 28. The calculation was done as follows: A scattering kernel based on the frequency spectrum of Sinclair³² was generated in the inelastic incoherent approximation by means of the SUMMIT³¹ code. To the new scattering kernel were added the coherent elastic scattering cross sections as computed by the HEXSCAT³³ code. The comparison in Figure 28 is reasonably satisfactory except at low neutron energies, where the energy mesh is too coarse and the incoherent approximation is not expected to be satisfactory. Despite the reasonably satisfactory agreement of σ_T in Figure 28, the scattering model is very approximate and does not give good results in spectral predictions.² A more realistic formulation of the neutron scattering problem for BeO is now under way.³⁴ It is also desirable to remeasure the total cross section for this moderator, especially below the Bragg cutoff.

Figure 27. Total cross section of beryllium.

Figure 28. Measured and calculated total cross section of BeO at room temperature.

Conclusions

In the previous sections the status of the integral neutron thermalization studies at General Atomic has been reviewed, particularly the measurements of neutron spectra, total cross sections, and angular scattering cross sections. It is evident from this review that changes of emphasis are occurring in this field.

One currently important area is that of dissemination of spectral data in a usable standardized format. The General Atomic Spectrum Book and the compilation of the European American Committee on Reactor Physics, which contains data from other institutions, are indicative of this trend. These data should prove useful in checking out many reactor design codes at the various reactor design centers.

Another current consideration is simply the question of accuracy. The continued refinement of the theoretical scattering models and the improved numerical procedures for generating scattering kernels demand improved data for comparison. These factors have increased the importance of the angular

scattering measurements due to their ability to detect nuances in the scattering model. In addition considerations of interference scattering, as in D_2O and Be, have necessitated a reappraisal of total cross sections, particularly below the Bragg cutoff energy. It is evident that some high precision measurements of σ_T for Be, BeO, and ZrH_n are necessary.

A further area is also related to the improved accuracy of the calculations. It appears that infinite medium spectra can be predicted with reasonable accuracy for many reactor calculations. Consequently, emphasis has changed from spectral measurements in homogeneous media to measurements in heterogeneous assemblies in which a strong spatial dependence is observed. Such studies are consistent with the remaining uncertainties in thermal reactors, such as flux distribution at control rods and core reflector interfaces, and also the spatial flux dependence across the cells of tightly packed lattices. The recent measurements of this nature are encouraging and should be very useful. This type of measurement should be extended if possible to cover the principal resonances of thorium and U^{238} .

The final area representing change in this field is that H_2O is beginning to receive less emphasis than the other moderators. This reflects the past effort expended to improve the H_2O scattering model. Remaining problems of interest for water systems concern mainly the study of spectra in very heterogeneous arrays. One would expect more emphasis to be placed on the various organics, beryllium and beryllium oxide, and finally graphite under high loading conditions. The uncertainties in neutron spectra in the various moderators can be expected in the future to be at very low energies, where coherent effects are dominant, or in the energy regions where the resonance absorption is not presently predicted accurately.

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APPENDIX C

TIME-OF-FLIGHT MEASUREMENT OF THE NEUTRON SPECTRUM
FROM A SPHERICAL U²³⁵ FAST REACTOR*

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The time-of-flight method was used to measure the neutron spectrum from the Accelerator-Pulsed Fast Assembly (APFA), which is a seven-inch diameter sphere of 93%-enriched uranium similar to the GODIVA reactor. The reactor was subcritical with a prompt multiplication of 18, giving a prompt neutron dieaway time constant of about 112 nanoseconds. The reactor is controlled by separation into three segments; for this experiment a 0.5-inch gap was introduced between the upper third and the remainder of the core. Neutrons were produced near the center of the sphere by (γ, n) reactions resulting from 30 MeV electrons striking the uranium at the bottom of a 1-inch diameter by 3-inch deep reentrant hole. The electron beam, from the General Atomic Linear Accelerator, was pulsed at 20 ns and 360 pps. The detectors used were a 2-in. by 2-in. liquid scintillator for energies above 1 MeV and a B_4C -CH₂ capture slab and gamma-ray sensor. A one-inch diameter collimated beam from the surface was allowed to drift down a 50-meter evacuated flight path. The flight distance and dieaway time results in a 15% energy resolution at 10 MeV, and somewhat better resolution at lower energies. The spectrum was obtained from 30 keV to 10 MeV with an estimated error of $\pm 10\%$.

Measurements were made at different positions and angles on the surface. It was found that the angular distribution of the leakage flux was cosine, and the spectrum was essentially independent of angle. The spectrum is plotted in Fig. 1. For comparison, a GAPLSN S_n transport theory calculation, normalized at a few MeV, is shown. The calculation was carried out for a spherical critical reactor. The calculated spectrum has fewer low energy neutrons than the measured one. Also shown for comparison is the photoplate measurements of Frye, et al., ⁽¹⁾ on the critical GODIVA reactor. The photoplate spectrum has an even greater excess of low energy neutrons.

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Fig. 1--Neutron spectrum of a spherical U²³⁵ fast reactor