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INTEGRAL NEUTRON THERMALIZATION

QUARTERLY PROGRESS REPORT

FOR THE PERIOD ENDING JUNE 30, 1964

Contract AT (04-3)-167

Project Agreement No. 2

U. S. Atomic Energy Commission

July 24, 1964

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I. INTRODUCTION

This quarterly progress report describes the work performed under Project Agreement No. 2 of Contract AT (04-3)-167 with the U. S. Atomic Energy Commission for the quarter ending June 30, 1964. The research accomplished on the neutron thermalization program has included:

1. Further development of the universal scattering-law code (GASKET) for calculating thermal scattering cross-sections.
2. Space independent and dependent integral neutron spectral measurements to check the adequacy of the theoretical predictions from the reactor physics standpoint.
3. Further work on the differential scattering experiment.

In addition to the above tasks an extensive effort was made to standardize the format and complete more cases for the Spectrum Book which EACRP* has recently elected to sponsor. A study devoted to the theoretical analysis of pulsed neutron experiments has been completed by Noel Corngold and is attached as Appendix II to this quarterly report.

The main program emphasis was as follows: A new scattering kernel for D_2O has been calculated including intramolecular interference. It appears that this representation does not fit experiment as well as the scattering kernel based on the incoherent approximation. Further improvements may need to be made by including intermolecular and liquid effects, and a formulation of this problem is underway at present. Good progress was made on the new scattering code GASKET during this quarter and it should be put into final form during this contract period. Very encouraging agreement has been obtained between spectral measurements and theory for both D_2O and H_2O under a variety of experimental conditions. A complete

*European-American Committee on Reactor Physics

series of space dependent BeO spectra have been measured and the pronounced effect of Bragg scattering was observed in the energy region near the cutoff. A similarly large effect is expected in poisoned steady state beryllium spectra which will be measured in the near future. The angular cross-section measurements previously made in water were studied extensively to ascertain the importance of multiple scattering. A new flight path has been completed to eliminate many troublesome experimental difficulties as well as increase intensity by over an order of magnitude. Preliminary tests were completed and complete measurements will be conducted of the water scattering cross section at about 100 neutron energies from $< .01$ ev to 5 ev.

The following publications and scientific meeting papers were prepared during this reporting period:

1. "Neutron Thermalization in Zirconium Hydride"
Nuc. Sci. and Eng. 19, 230 (1964) by
J. C. Young, J. A. Young, G. K. Houghton,
G. D. Trimble and J. R. Beyster.
2. "Lattice Vibrational Spectra of Be, Mg and Zn"
Phys. Rev. 134A, 1476, (1964) by
J. A. Young and J. U. Koppel.
3. "Scattering Kernel for Beryllium"
Nuc. Sci. and Eng. Vol. 19 (1964) by
J. A. Young and J. U. Koppel.
4. "On the Validity of the Pulsed Neutron Measurement of $K\beta/\ell$ "
Trans. of the American Nuc. Soc.
June 1964 by J. L. Russell, Jr.,
J. R. Brown and J. C. Young.
5. "Neutron Spectrum Measurements in a (UF_4 + paraffin) Multiplying Assembly"
Trans. of the American Nuc. Soc.
June 1964 by J. C. Young, J. M. Neill and J. R. Beyster.

II. EXPERIMENTAL

2.1 NEUTRON SPECTRUM MEASUREMENTS IN D₂O

Studies of angular neutron spectra in a direction perpendicular to the source-assembly axis in high-purity D₂O poisoned with a 1/v absorber (boron) have been reported previously.⁽¹⁾ The studies were performed to test the bound deuterium models⁽²⁾ generated to describe the thermal neutron scattering properties of this moderator. Since these measurements were in good agreement with the calculated spectra, the study of the moderator was expanded to include more severe tests of the scattering kernel and numerical procedures used in reactor calculations. This series of experiments has included measurements of the neutron spectra in the presence of various flux gradients, including the leakage spectrum at the surface of the assembly with the same 1/v absorber used previously. These measurements were made to test the higher order angular dependence (P_1) of the scattering kernel. For a more severe test of the P_0 scattering matrix than was possible with a 1/v absorber, quasi-infinite-medium neutron spectra were measured in D₂O heavily poisoned with a resonance absorber (cadmium).

The geometrical arrangement used for these studies is shown in Figure 1. For the angular neutron spectrum measurements at various flux gradients and at the surface, the re-entrant tube was inserted in such a manner that "A" in Figure 1 was equal to 15.24 cm. The angular neutron spectra 90° to the source-assembly axis were measured at D (Figure 1) equal to 12.7, 17.8, 20.3, 22.9 and 25.4 cm. In addition to these spectrum measurements, indium resonance neutron spatial flux distributions were taken in all three dimensions of the assembly. These spatial distributions were used to determine the leakage corrections and input source for the calculated spectra. The spectra were calculated using the bound deuterium kernel described in Reference 2. The one-dimensional transport theory

Fig. 1--Geometrical arrangement used in D_2O spectrum measurements.

code GAPLSN⁽³⁾ was used to calculate the spectra along the "D" dimension of Figure 1. The experimental data and the calculated spectra are shown in Figures 2 through 6. From the center of the assembly (Figure 2) to its surface (Figure 6) there is a 50% hardening of the neutron spectra as indicated by the thermal flux differences with the spectra normalized to the same $E\phi(E)$ at 10 ev. This difference is sufficiently large to indicate a distinct sensitivity of the geometry studied to P_1 scattering as well as the P_0 . The agreement between the experimental and calculated spectra shown in these figures indicates that the scattering description used in these calculations is adequate for predicting the angular neutron spectra in D_2O poisoned with a $1/v$ absorber.

The neutron spectrum measurements in D_2O poisoned with cadmium to 2.4 barns/deuterium atom at 0.025 ev were taken at "A" (Figure 1) equal to 15.24, 22.86 and 30.48 cm. Cadmium covered indium flux distributions were also mapped for this solution and used for leakage corrections and the input source for the GAPLSN calculation. Two of the monitors shown in Figure 1 were activation detectors. The third monitor was a U^{235} fission counter which sampled a neutron flux proportional to the source neutrons. Each of these monitors tracked to within $\sim 2.5\%$. Figure 7 shows the measured spectra as compared to the calculated spectra. The calculated spectrum at the 30.48 cm position was normalized at 1 ev to the experimental data for that position. As can be seen in Figure 7, the shape, as well as the relative intensity, of the spectra at 30.48 cm and 22.86 cm are in good agreement. However, at 15.24 cm the relative intensity for the calculated spectra is about 10% lower than that predicted by the experiment. This difficulty is most likely due to the manner in which the spatial source distribution is put into the GAPLSN calculation. The measured distribution is taken at indium resonance (1.45 ev), but is entered into GAPLSN at 2.08 ev. The change in the shape of the spatial flux distribution calculated by GAPLSN is illustrated in Figure 8. If the calculated distribution at 2.08 ev is modified to give the measured distribution at indium

Fig. 2--Neutron spectrum in borated D_2O .

Fig. 3--Neutron spectrum in borated D_2O

Fig. 4--Neutron spectrum in borated D_2O

Fig. 5--Neutron spectrum in borated D₂O

Fig. 6--Neutron spectrum in borated D_2O

Fig. 7--Angular spectra in D_2O poisoned by $Cd(NO_3)_2$

Fig. 8--Calculated flux distribution in $D_2O + Cd(NO_3)_2$ axial to the source at different neutron energies

resonance, the relative intensity of the energy spectrum at 15.24 cm would be raised by about 6%. This correction would put the measured and calculated relative intensities in agreement within the limits of the monitoring system. The apparent disagreement at the 15.24 cm position is in the relative intensity and not in the shape of the spectrum, the calculated and measured spectra at this position are compared in Figure 9. Here the calculated spectrum has been normalized at 1 ev to the measured data. This illustrates that equally good agreement in spectral shape is obtained at the 15.24 cm position, as in the other two positions. Modification of the spatial flux distribution at the 2.08 ev energy group, to give the measured distribution at indium resonance, has been performed on previous data. This modification does not change the spectral shape of the energy spectrum, therefore good agreement should be maintained.

To summarize, the scattering kernel described in Reference 2 for the important reactor moderator D_2O is adequate for predicting the neutron spectra measured in various absorbers and under varying conditions at room temperature. The calculational techniques utilized appear to be adequate. Future studies of this moderator should include tests at other temperatures as well as heterogeneous configurations. These could include spectra measured in lattices and near absorbing boundaries similar to control rods. In addition, a new theoretical model for D_2O is being developed under this program in which some of the approximations made in Reference 2 have been removed.

2.2 NEUTRON SPECTRUM MEASUREMENTS IN BeO

During this quarter, further measurements were made of the thermal neutron spectra in an assembly of poisoned beryllium oxide. A measurement, made earlier this year and reported in Reference 1, was obtained at only one position and was not a good measurement due to monitoring inadequacies and a very high background. This high background was subsequently found to originate from the then uncompleted modifications to the Linac experimental

Fig. 9--Neutron spectrum in D₂O poisoned with cadmium at D = 6 inches

room. In the new measurements, proper shielding has been used to obtain a low background and, in addition, the monitoring difficulties have been overcome.

Figure 10 illustrates the BeO geometry in which angular flux measurements were made at 90° to the source-assembly axis. The beryllium oxide was poisoned by placing two 0.005 in. thick sheets of 1% by weight borated stainless steel between 0.42 in. thick layers of BeO. This poisoning represented about 1.2 barns/Be atom. Figure 11 shows the neutron spectra measured at 6 in., 12 in. and 18 in. from the source side and indicates a softening of the spectrum that one would expect from an inspection of the measured epicalcium neutron distribution shown in Figure 12. From the slope of the curve in this latter figure, it can be seen that the local buckling at the 6 in. position is less negative, which gives rise to a neutron spectrum that is harder than those observed at the 12 in. and 18 in. positions.

The elastic coherent scattering manifests itself by a change in the neutron intensity below the Bragg cutoff to give a peak at about 0.0033 ev. Elastic coherent scattering effects would not normally be observed in a quasi-infinite medium experiment as the equation below for the flux in an infinite homogeneous medium indicates.

$$\phi(E) = \frac{S(E)}{\Sigma_a(E)} + \frac{1}{\Sigma_a(E)} \int_0^\infty dE' \Sigma_s(E' \rightarrow E) \phi(E') - \frac{\Sigma_s(E)}{\Sigma_a(E)} \phi(E)$$

Fig. 10--Geometrical arrangement used for BeO spectral measurements

Fig. 11--90° angular spectra in BeO

Fig. 12--Relative intensity of the episcadmium neutron flux measured in BeO

On the right hand side of the equation the scattering kernel $\sum_s (E' \rightarrow E)$ can be divided into an inelastic component and an elastic component, and on integration that elastic component becomes

$$\frac{\sum_s (E)}{\sum_a (E)} \phi(E)$$

which cancels with the last term on the right hand side of the equation.

However, the BeO assembly was characterized by a large negative total buckling B^2 , so that in the diffusion theory approximation $\sum_a (E)$ in the equation above is replaced by $\left[\sum_a (E) + D(E)B^2 \right]$. Since

$$D(E) \approx \frac{1}{3 \sum_s (E) (1 - \bar{\mu})}$$

it can be seen that when the neutron wavelength becomes larger than the Bragg cut-off, $\sum_s$ then decreases greatly so that $D(E)B^2$ becomes a larger negative term, which means that $\left[\sum_a (E) + D(E)B^2 \right]$ becomes smaller. This gives rise to a change of neutron intensity at the Bragg cut-off causing the peak observed in the measured spectra. This effect is a spatial one, therefore, the neutron intensity change is underestimated when evaluated numerically on a diffusion theory basis.

The theoretical curves for these measurements have not been calculated since elastic coherent scattering has not yet been added to the present BeO scattering model. In addition, it has not been found possible to calculate a neutron spectrum in the B_1 or P_1 approximation with the BeO kernel we have now. This scattering kernel has been calculated from the BeO frequency distribution of Sinclair⁽⁴⁾ by means of the code SUMMIT.⁽⁵⁾ The cause of the difficulty has not yet been discovered though it is believed to arise from some local coarseness of the energy mesh used in the calculation of the kernel or in the selection of some of the arbitrary constants used in the phonon expansion in SUMMIT. Consequently, these data have not been incorporated in the Spectrum Book.

2.3 NEUTRON SPECTRUM MEASUREMENTS IN H₂O

2.3.1 Angular Neutron Spectra in a 4-inch Slab

Measurements of thermal neutron spectra have been made in slab geometry in the presence of strong one-dimensional flux gradients. These measurements were performed to culminate studies^(1, 6) which have been pursued for some time. The spectra were measured in clean geometrical configurations which were designed to produce typical spectra encountered in actual reactor operations, such as spectra near control rods or core-reflector interfaces.

Angular neutron spectra perpendicular to the source-assembly axis were measured in water poisoned to about 5 barns per hydrogen atom with boron. The geometrical arrangement used for these measurements is shown in Figure 13. The one-inch thick B₄C boundaries, which were not included in previous measurements,⁽⁶⁾ were used to reduce epicadmium room return neutrons which may have been perturbing the surface leakage spectrum measurements performed previously. Cadmium covered indium foils were used in all three dimensions of the assembly to measure the epithermal spatial source distribution needed as an input parameter in the calculations and to determine the leakage corrections necessary for the finite dimensions of the slab. Measurements of the epicadmium neutron flux were also taken without the boundaries to determine their effect on room return neutrons. The spatial distributions axial to the source are shown in Figure 14. The two curves have been normalized at their peaks. The curve shown for the data without the boundaries indicate that room return is appreciable beyond 20 cm. This effect is reduced with the boundaries in place. However, it should be noted that the curve shown for the case with the boundaries has a somewhat smaller slope. This is most likely from fast neutron reflection from the boundaries.

Fig. 13--Geometrical arrangement used for spatially dependent neutron spectra measured in borated water

Fig. 14--Cadmium covered indium flux distributions in borated water axial to the source

Figure 15 shows the spatial distribution taken in the thin dimension of the slab with and without the boundaries. Here the shape did not change, which may indicate that the reflection from the boundaries is symmetrical in the thin dimension.

The neutron spectra measured in the geometry of Figure 13 were done with the re-entrant tube 15.24 cm from the source side with "D" being equal to 2.54, 5.08, 7.62 and 10.16 cm (surface). The corresponding spectra were calculated using the Nelkin Bound hydrogen scattering model in the Sn transport theory code GAPLSN. The spatial neutron flux distribution obtained in the 10 cm dimension of the assembly using cadmium covered indium foils was used to determine the input source distribution for these calculations. P_0 , P_1 source and P_0 , P_1 scattering matrices and 24 neutron energy groups were utilized in the calculations. The atom densities which were used are given in Table 1.

Table 1

ATOM DENSITIES FOR 5.2 BARNS PER
HYDROGEN BORATED WATER
($\times 10^{-24}$ Atoms/cm³)

N boron	0.000424
N hydrogen	0.06605
N oxygen	0.03366

The results of the calculations compared to the experimental data are shown in Figures 16 through 19. The calculated spectra have been normalized to the experimental data at 1 ev. The inset on the figures gives the normalized ratio of the theory to the experiment. The calculated shape of the neutron spectrum for each position is improved over the measurements made previously, this is particularly true for the surface leakage whose agreement with theory is now as good as that of spectra measured

Fig. 15--Cadmium covered indium flux distributions in borated water transverse to the source

Fig. 16--Neutron spectrum measurement in borated water

Fig. 17--Neutron spectrum measurement in borated water

Fig. 18--Neutron spectrum measurement in borated water

Fig. 19--Neutron spectrum measurement in borated water

internally. This improvement is most likely due to improved boundary conditions under which the recent measurements have been made. It should be noted that the measurements made as near as 1 inch to the surface were not appreciably affected by the B_4C boundaries.

To check the relative intensity predicted for the neutron energy spectrum for each space point by the GAPLSN calculation, three monitors were utilized in taking the experimental data. Two of these monitors utilized foil activation to sample the source neutrons at two different energies. The positions of these monitors are shown in Figure 13. The aluminum cap, placed over the source, sampled the fast flux (~ 6 Mev) emitted from the source. The gold foil monitors were placed in the assembly to sample the thermal flux. The third monitor, a fission counter, was used to sample the room return neutrons, which should also be proportional to the source neutrons. Table 2 compares the results of these monitors in the form of monitor counts per pulse normalized by the average value of each column. How near the values are to each other in any one row of this table points to the adequacy of the monitoring devices. From this table it may be concluded that the fission monitor is reliable and that these monitoring techniques should provide monitoring accuracy to within $\pm 2 - 1/2\%$.

Table 2

COMPARISON OF NEUTRON MONITORING DATA

D cm	Normalized Counts Per Pulse		
	Aluminum	Gold	Fission
2.54	1.102	1.083	1.044
5.08	1.069	1.037	1.044
7.62	1.065	1.040	1.048
SURFACE	0.896	0.939	0.947

To compare the relative intensities, the theoretical and experimental neutron spectra $E\phi(E)$ were averaged from 1 ev to 2 ev neutron energy. The experimental average $E\phi(E)$ was then normalized to the theoretical average $E\phi(E)$ at the center position of the tank ($D = 5.08$ cm) and the percent difference was calculated for the other positions. The results are given in Table 3.

Table 3.

RELATIVE NEUTRON INTENSITIES OF THEORY AND
EXPERIMENT FOR EACH POSITION

Position cm	Theory	Data	% Difference
2.54	0.3326	0.3291	2.0
5.08	0.4788	0.4678	2.3
7.62	0.5109	0.5109	0.0
SURFACE	0.4366	0.5032	15.0

As can be seen from Table 3, the calculated relative intensities are in good agreement for each position except at the surface. This may indicate that either the P_1 scattering matrix is incorrect, which would manifest itself at the surface, or the reflection from the B_4C boundaries has boosted the source intensity at the surface.

Future studies of this moderator should include measurements in heterogeneous configurations such as those described in Section 2.3.2. Additional work should also include studies designed as severe tests of the P_1 and higher order components of the scattering matrix, and measurements should be made with this moderator at elevated temperatures with resonance absorbers.

2.3.2 Angular Spectra in a 4-inch Slab of Water at 0° , 30° and 45°

It was decided to measure angular flux spectra in a 12 in. by 12 in. by 4 in. tank of borated H_2O in a plane perpendicular to the source-assembly axis. The experimental geometry of the configuration is similar to that shown in Figure 20. The measurements were made at 1 in. from the central axis always in a direction perpendicular to the source flux gradient so that effects due to the anisotropy of the source were eliminated. It was believed that these spectra would exhibit a strong angular dependence as the tank was rotated reflecting the change in the transverse flux gradient from the 4 in. to the 12 in. dimension. Unfortunately the measurements preceded the calculations and though both agreed well, the angular dependence was small. This arose because the water was poisoned heavily enough that the spectra were not sensitive to the leakage. A successful test would have required the imposition of a strong angularly-dependent flux gradient. Consequently, no useful conclusions could be drawn, and the data are not presented here.

The apparatus developed for the above measurements was, however, utilized in a further experiment. The configuration is shown in Figure 20 and indicates how a strong gradient was obtained by placing concentrated cadmium sulphate solution outside a thin tank containing dilute boric acid. The inner tank was poisoned mildly with the boric acid in order to reduce its leakage probability. The diffusion corrections for axial and vertical leakage then became sufficiently small to allow an accurate calculation of the angular spectra by the one-dimensional transport code GAPLSN⁽³⁾ in the horizontal flight path direction.

Spectral measurements were made with the horizontal axis of the tank at 0° , 30° , and 45° to the flight path direction. Some difficulties were encountered during the measurements. Since the lead used to spread the neutrons was attached directly to the assembly, the heat generated in the lead was transmitted directly to the tanks so that the fluids inside the

Fig. 20--Geometrical configuration for water interface experiment

tanks became warm and expanded. The outer tank was able to leak the excess, but since the inner tank containing the boric acid solution was not vented some bowing occurred. This bowing was estimated to be about 0.010 in. at the position of measurement, and would not have been serious except that the neutron spectra were strongly spatially dependent. Monitoring difficulties were encountered when the fission counter normally used became inoperable during the experiment. Gold foils, used as a backup monitor, were therefore utilized for background subtraction and normalization purposes. They did not, however, prove satisfactory since the fluxes did not normalize correctly in the $1/E$ region.

In performing the calculations for this problem, a water kernel at 39.5°C was used instead of the 54°C which is more typical for this assembly. This was done because of the additional cost of generating a new kernel at this temperature, and because the difference of 15°C was considered to be too small to affect the spectra significantly.

The results of the measurements compared to theory are shown in Figures 21 to 23. Figure 24, showing the three calculated spectra only, illustrates the strong angular dependence. Each theoretical curve and measurement are normalized separately due to the experimental monitoring difficulties. The 0° measurement fits the calculated values to within 15%. The fit of experiment to theory at the other two angles is not as good, with errors of up to 30% being indicated. Some conclusions should be drawn as to whether these discrepancies are experimental in origin or reflect non-physical features of the scattering model for water.

The principal features of water that the Nelkin model⁽⁷⁾ neglects are:

1. That the translation motion of the H_2O molecules is hindered and not free.
2. The vibrational modes of motion are non-isotropic.
3. That a band of rotational frequencies rather than a discrete oscillator exists.

Fig. 21--Angular spectrum at $\theta = 0^\circ$

Fig. 22--Angular spectrum at $\theta = 30^\circ$

Fig. 23--Angular spectrum at $\theta = -45^\circ$

Fig. 24--Calculated spectra for interface experiment showing relative intensity and shape variation

Item 2 has been treated to a reasonable approximation by Koppel,⁽⁶⁾ and one could extrapolate from his results that the anisotropy could account for only 5% of the discrepancies of the present measurements. Item 1 should have little effect on the spectra at the higher energies, above 0.5 ev where the translational motions are nearly free anyway. At the lower energies below 0.1 ev, the hindered treatment should tend to make the calculated spectra harder which is in the wrong direction. Item 3 could affect the spectra either way. Because the spectra are very sensitive to position and angle, and because there are uncertainties in the measured positions and angles, it is impossible to be definite about a conclusion that the discrepancy does not arise from an inadequacy of the Nelkin water model. These angular spectra were also calculated with no P_1 scattering, but this did not change the shape or intensity of the flux spectra at the position of measurement. Obviously the experiment was not a good test for P_1 scattering, but it provided sufficiently reasonable agreement to have confidence in the P_0 scattering matrix. It would be desirable to repeat this experiment with the strong cadmium sulphate solution diluted by a miscible non-hydrogeneous, non-moderating material to reduce the position sensitivity and hence the experimental uncertainties. In addition, a better test of the P_1 scattering would be obtained by making measurements at 0° at different distances from the interface.

2.4 SENSITIVITY OF THE 16-M AND 50-M DETECTOR BANKS

2.4.1 16M Sensitivity

The importance of a well defined energy dependent neutron sensitivity, $S(E)$, of the $B^{10}F_3$ detectors used in these thermalization experiments has long been recognized, since this $S(E)$ is used directly in reducing the measured data to energy spectra. The sensitivity of the bank of 32 $B^{10}F_3$ counters normally used on the 16-meter flight path has been checked on several occasions. This was done by comparing a standard spectrum obtained by using this bank with the same spectrum made by another detector, whose sensitivity was easily calculable. In addition, the $S(E)$ of the $B^{10}F_3$ bank was also calculated⁽⁸⁾ on the basis of known geometry and neutron cross sections. Both of these methods should yield the same $S(E)$ for the $B^{10}F_3$ bank, but a substantial discrepancy between the calculated and experimental $S(E)$ was observed between 1 and 100 ev when a lithium glass scintillator was used as a comparison detector.⁽¹⁾ When a bank of 10 low-pressure BF_3 counters having a $1/v$ response was used as a comparison detector, reasonably good agreement with the calculated $S(E)$ was obtained. It became evident that a better analysis of the lithium detector was required to describe its sensitivity adequately.

The count rate of the detector is directly related to the neutron capture probability in the Li^6 glass due to the reaction $Li^6(n, \alpha)T + 4.8$ Mev. The sensitivity, $S_1(E)$ of the 2.5 in. diameter lithium glass detector was calculated according to the following expression:

$$S_1(E) = T_{A1}(1-T_g) \left(\frac{\Sigma_a Li}{\Sigma_{tg}} + \frac{\Sigma_{sg}}{\Sigma_{tg}} [1 - T_g'] \frac{\Sigma_a Li}{\Sigma_{tg}} \right)$$

The macroscopic cross sections, Σ , with their subscripts, have their usual meaning. The lithium glass transmission is given by $T_g = e^{-\Sigma_t X}$. The thickness, X , of the glass is 1/2 inch. The transmission of the 0.002 in. aluminum light shield is given by:

$$T_{Al} = e^{-(\Sigma_a + 1/2 \Sigma_s)_{Al} t}$$

In this calculation only primary and secondary absorption were taken into account. This procedure was considered accurate enough because the secondary absorption contributes only 20 per cent to the total absorption at a neutron energy of 100 ev. The secondary scattering length X' used in T_g is assumed to be 3/8 in. since it should be between 1/4 in. and 1/2 in., and is probably close to the lower limit. The biggest uncertainty in this $S_1(E)$ calculation is introduced by the fact that the chemical composition of this glass is not very well known. According to the supplier of this detector, the Li^6 content may vary from 6.5 to 7.5 per cent by weight. The $S_1(E)$ has been calculated for both of these Li^6 concentrations, and the results are shown in Figure 25. For the other materials of the glass, the following weight percentages were used in these calculations: 65% SiO_2 , 10% Al_2O_3 , 3.3% MgO , and 4% Ce_2O_3 . Assuming a lithium content of 6.5% (15.2% Li_2O), the $S(E)$ of the big BF_3 bank was obtained using the lithium glass as a comparison detector. This $S(E)$, along with earlier reported⁽¹⁾ values of $S(E)$ is represented in Figure 26. Fairly good agreement is now obtained between the experimental determinations of $S(E)$. Inconsistencies at the lower energies can be ignored because of poor statistics.

Fig. 25--Calculated efficiencies for Li^6 detector

Fig. 26--16 meter B¹⁰_{F₃} detector bank sensitivity

It still remains to be shown that the Li^6 content of this glass is indeed approximately 6.5%. A chemical analysis of part of the glass may not be easy, and this would not yield the Li^6 enrichment which is quoted as a nominal 96%. An alternative method would be to measure the thermal neutron transmission of the lithium glass. Calculations indicate that a 2.4% difference in transmission at 100 ev can be expected between glass containing 6.5 and 7.5% Li^6 . This difference may be large enough to determine the lithium content experimentally. Another way to determine $S(E)$ more accurately would be to remeasure the sensitivity of the B^{10}F_3 with a thicker Li^6 glass of which the composition was well known, or possibly with a B^{10} glass. Due to the high thermal neutron cross section of B^{10} , the boron glass sensitivity has a factor of about four in its favor as compared to lithium glass. However, it has been pointed out by Bollinger et al. (9) that boron glasses are more sensitive to low energy γ rays than lithium glass, and this may limit the usefulness of boron glass for this application.

2.4.2 50 Meter Sensitivity

Since the old helium-filled 50-meter flight path was replaced recently by a new evacuated tube, it was felt that the relative sensitivity of the big B^{10}F_3 detector, normally used on this flight path, should be remeasured. The 50-meter path is an extension of the 16-meter flight path, and therefore the sensitivity of this detector is usually compared to that of the B^{10}F_3 bank at the 16-meter station. Surface leakage spectra from pure polyethylene, as observed by the 50-meter bank, were compared with the same spectra measured by the 16-meter B^{10}F_3 detector. Using the 16-meter bank $S(E)$, (6) it was found that there was no significant difference between the measured 50-meter bank sensitivity and the $S(E)$ for the same detector and the old helium filled flight path, as reported in Reference 8.

2.5 NEUTRON MONITOR STUDIES

During the last quarter, further attention has been given to measurements of relative neutron intensities. It was pointed out in Ref. 1 that these measurements are needed to provide normalization of neutron spectra at various positions in an assembly, and also to allow proper subtraction of background spectra.

The relative intensity of the pulsed neutron source is measured by a fission counter, which is located several meters away from the target. The large distance between this counter and the assembly makes it rather insensitive to changes in the position of the assembly. The thermal neutron flux in the assembly under investigation is monitored by the $\text{Au}^{197}(n, \gamma)\text{Au}^{198}$ reaction in gold foils which are located on the outside surface of the assembly and are shielded against room-return neutrons by a cadmium sheet. Finally, the fast neutrons are monitored by aluminum caps which are slipped over the target or by aluminum slugs which are attached to the source side of the assembly. A 15-hour gamma ray activity of Na^{24} is produced in the aluminum monitors by the $\text{Al}^{27}(n, \alpha)\text{Na}^{24}$ reaction, which has a threshold of about 6 Mev. Since exposure times are usually an appreciable fraction of this 15-hour half life, the following correction factor has been used to allow for decay during irradiation,

$$F = \frac{\lambda t}{1 - e^{-\lambda t}}$$

where t is the exposure time and λ is the decay constant. This correction amounts to about 10% for the aluminum monitors in a four hour long experiment.

A complete survey has been made of the monitor data obtained from these three independent monitors over the past few months. The fission counter, which gives the most immediate results, is usually the most reliable monitor. Comparisons between the normalized counts per neutron pulse for each monitor show that our present method of monitoring is quite

adequate for background subtraction. The signal-to-background ratio can normally be monitored to within a few percent, but even a 5% error in this ratio will only introduce an error of 1% in the measured spectra, if the background is 20%. Two typical sets of monitor data are shown in Table 4. The corresponding experiments are described in Sections 2.3.2 and 2.1 of this report. There is good agreement between the normalized fission monitor counts of the signal runs and of the corresponding background runs, but the aluminum and gold monitors show some sizeable differences. Each monitor has been normalized to its average value.

When the neutron monitor counts are used to normalize the measured spectra at various points, it becomes more important that the monitoring system produces reliable results, since the measured neutron flux, $\phi(E)$, is directly proportional to the number of monitor counts. It has been found that the fission counter is fairly suitable for this purpose, but the gold and aluminum monitors show considerable differences between successive runs. This is particularly noticeable when the position of the assembly is changed with respect to the room and the target. This effect can be seen in the H_2O monitor data listed in Table 4. In this case both the gold and the aluminum monitors were attached to the assembly which was rotated with respect to the fixed target. Consequently, both the aluminum and the gold monitor activities reflect this position dependence. This difficulty could conceivably be minimized if a cadmium covered gold foil were located inside the assembly at, or near, the maximum of the epicadmium neutron flux. Here the position of the gold would not be too critical and possible differences due to room return neutrons would be minimized. Also, a more realistic representation of the neutron flux inside the medium would be obtained in this fashion.

Table 4

TYPICAL NEUTRON MONITOR DATA

Material Date	Run No.	Position	Counts per Pulse/Average Value		
			Fission	Aluminum	Gold
H ₂ O 5-9-64	4-S*	0°	1.045	1.067	1.053
	5-B**	0°	1.027	1.019	1.050
	9-S	30°	.979	1.086	1.031
	10-B	30°	.985	1.220	1.082
	11-S	-45°	.976	.890	.895
	12-B	-45°	.988	.799	.890
D ₂ O 4-1-64	1-S	Center	1.004	1.063	1.039
	2-B	Center	.984		.987
	3-S	Surface	.995	.968	1.026
	4-B	Surface	.980		.984
	5-B	3 in.	.956	.985	.935
	6-B	3 in.	.953		.916
	7-S	4 in.	1.039	.916	.935
	8-B	4 in.	1.027		1.100
	9-S	2 in.	1.020	1.068	.935
	10-B	2 in.	1.040		1.142

*Signal
**Background

2.6 ANGULAR DISTRIBUTION MEASUREMENTS

The work performed on the angular scattering distributions during this reporting period was aimed at completion and test of the new experimental apparatus. Many difficulties of the system which could affect the experimental data were studied. A description of the experiments, their results, and steps taken to improve our techniques are given below.

(1) The energy sensitivity function of the Li^6 glass detectors must be well known at all neutron energies involved in the experiment, since the energy of the scattered neutrons is not equal to that of the neutrons incident upon the sample. Sensitivity calculations in the Li^6 glass have been made and verified experimentally (Section 2.4). It was revealed that the sensitivity function actually changes insignificantly, at most 5%, over the entire experimental energy region of our measurements (up to 1 ev).

(2) The flux incident upon the scattering sample must be known in both shape and magnitude. The flux map, measured with Li^6 glass detector having a 1/8 in. entrance collimator, showed some asymmetry in the source distribution. A thick water moderator which is concave on the source side is being built, and will be located near the fast neutron source; this arrangement should yield a more uniform leakage density. A flat distribution of flux incident upon the sample is highly desirable for the determination of absolute cross sections and to permit a reduction of the physical source size.

(3) The results of the neutron monitoring tests performed in the flight path are not clear as yet. The correlation between the source monitor and the scattered neutron detector in the 0° position is not linear at low intensities. The difficulty, we know, does not lie in detector dead time corrections.

(4) An investigation of flight path collimator was undertaken. In an effort to explain the flight path monitor difficulties, leakage of neutrons into the flight path was checked. The collimation proved to be more than sufficient. Only the source was shielded for this study. The leakage could not be shown to be different from ambient background with no electron beam present.

(5) The effective size of the neutron beam incident upon the sample was studied. The spot was larger than desired, therefore further collimation in the form of a large B_4C filled snout was installed. Its addition had a twofold effect: reduction of room background and cancellation of the penumbra effects upon the sample.

(6) A large gain in beam intensity occurred as a result of the new flight path arrangement. Data can now be accumulated at over a factor of ten times the rate previously obtained when this work was set up behind the capture cross-section experiment.

Construction of multiple detector equipment for the improvement of experimental techniques, providing more rapid data acquisitions, is still underway. The area of greatest concern at present is the development of uniform reliable thin water samples. At present various ways of bonding of aluminum are being investigated. It is expected that differential scattering measurements with multiple detectors will begin to be taken again in July and August, and special effort will be made to obtain improved absolute scattering cross sections for H_2O . Meanwhile, a summary of the experimental results taken to date are given in Appendix I, as presented at the Washington APS meeting.

2.7 NEUTRON SPECTRUM BOOK

Further progress has been made in the compilation of data for the neutron spectrum book. This book gives measured and theoretical neutron spectra for a variety of poisons and moderators at different temperatures, and tested in several geometries. The book is designed to be a guide to help reactor design groups to make comparative calculations to check the physics, mathematics, and numerical procedures which are used in their analyses.

Twenty nine spectra have now been completed and compiled by this laboratory, and additional cases which have been contributed by M. J. Poole from Harwell are now being evaluated.

The major difficulty encountered in the compilation of the spectra lies in the problem of normalizing the theoretical calculation with the measured data. It is believed that normalized to a high energy source would be the most appropriate method. This would certainly be achieved to first order if the normalization of the two fluxes was made in the $1/E$

portion of the spectra. However, this requires that the energy range of the calculations be extended upwards from the present limit of 2.38 ev which is determined by the fixed energy boundaries of the code GATHER,⁽¹⁰⁾ used to calculate group thermal cross sections. These cross sections are used by the Sn transport theory code GAPLSN.⁽³⁾ To extend the energy range of the calculations, two procedures are possible. An asymptotic expansion for the epithermal flux by Corngold can be calculated and tacked on in some consistent fashion to the calculated thermal spectrum or epithermal cross sections can be calculated from the code GAM⁽¹¹⁾ and these can be used in conjunction with the thermal group cross sections in the GAPLSN⁽³⁾ code to obtain the extended spectrum.

The procedures described above pose certain problems. The Corngold expansion is applicable to infinite medium cases in which any poisoning has a $1/v$ cross section. If this method were used, it could not be applied to all of our cases which would present an inconsistency in the theoretical approach. In addition, it is quite difficult to determine what is a consistent fashion in joining the thermal and epithermal calculated spectra. The other method, considered the best, requires considerably more computer time and has presently given some anomalous results. There is a jump in the spectrum at about 2.5 ev in H_2O which apparently is related to small differences between scattering cross sections calculated by the GAM code and by the GATHER code. This is serious enough, that correct normalization cannot be achieved until the error is resolved.

There is a third method, that has been used in which a space dependent spectrum is calculated using cross sections from the GAM code only. This spectrum is then tacked on to the thermal spectrum in the same way as the Corngold expansion with the same procedural limitations. No conclusion can be reached at this stage on our normalization procedure, and consequently, many of the spectra in the book are presented with calculations only up to 2.38 ev and fit in the $1/E$ region.

The format of the spectrum book is still not completely standardized. At present, the British group under Poole has presented its data as $E \phi(E)$ versus E as we have done at General Atomic. Poole has suggested, however, that the data would be presented better by a differencing procedure in which a large constant part of the spectrum is removed. The method has the advantage that differences between theory and measurement would be accentuated, but it has the disadvantages of a less visual presentation and that the difference procedure might not be available to every reactor design group. The details of the method are not clear, and, therefore, no decision has yet been reached. It is anticipated that the normalization procedure, the format, and the method of data presentation will be standardized soon. Meanwhile more cases will be compiled in the present format.

III. THEORETICAL

3.1 SELF AND INTRAMOLECULAR INTERFERENCE SCATTERING BY D₂O

Because of the great similarity between the D₂O and H₂O molecules it is straightforward to extend Nelkin's⁽⁷⁾ selfscattering kernel to D₂O. This was done by H. Honeck⁽¹²⁾ some time ago. However, whereas for hydrogen the ratio of incoherent to coherent cross sections is 39:1, it is only 1:2.7 for deuterium. Also the oxygen cross section is no longer small compared to the total molecular scattering cross section as it is for H₂O. Hence in the case of the D₂O molecule it is not obvious that the incoherent approximation, as used by Nelkin, is good enough for predicting neutron spectra at thermal energies. In order to find out about the importance of the interference scattering we have adapted the code GAKER,⁽¹²⁾ originally written for Nelkin's water kernel, to compute and add up the 2 self and the 2 interference terms, as given by Butler,⁽¹⁴⁾ according to the formula

$$\sigma_{D_2O} = 2\sigma^{DD} + 2\sigma^{D_1D_2} + 4\sigma^{OD} + \sigma^{OO}$$

Butler's calculation uses Zemach and Glauber's⁽¹⁵⁾ formalism and postulates in the same way as Nelkin did, the following degrees of freedom:

1. Free translation of the D₂O molecule.
2. Harmonic torsional vibrations of small amplitudes about the three principal axes of inertia, all with the same frequency ω_1 .
3. One-bending vibrational mode with frequency ω_2 .
4. Two-stretching (symmetric and antisymmetric) vibrational modes with the same frequency ω_3 .

The exact scattering kernel corresponding to these assumptions is given by the following expressions.

$$\sigma(E_0 \rightarrow E, \Omega) = \sum_{\nu\nu'} \sigma^{\nu\nu'}(E_0 \rightarrow E, \Omega) \quad (1)$$

$$\sigma^{\nu\nu'}(E_0 \rightarrow E, \Omega) = \frac{A_\nu A_{\nu'} + \delta_{\nu\nu'} C_\nu^2}{T} \frac{\sin b_{\nu\nu'} \kappa}{b_{\nu\nu'} \kappa} \sqrt{\frac{E}{E_0}} e^{-\frac{\beta}{2}} S^{\nu\nu'}(\alpha, \beta)$$

$$S^{\nu\nu'}(\alpha, \beta) = \sqrt{\frac{2MT}{4\pi\kappa}} \exp \left[- \left(\lambda_{\nu\nu'} + \frac{1}{4} \right) \frac{m}{M} \alpha \right] \quad (2)$$

$$\sum_{n_1=-\infty}^{\infty} \sum_{n_2=-\infty}^{\infty} \sum_{n_3=-\infty}^{\infty} \exp \left[\frac{-\beta - \sum_{\mu} n_{\mu} \frac{\omega_{\mu}}{T} 2M}{4m\alpha} \right] \prod_{\mu} \frac{1}{n_{\mu}} \left(\frac{m\alpha}{MB_{\mu}^{\nu\nu'}} \right) \quad (3)$$

Here the meaning of the different constants is:

A_ν = coherent scattering length of nucleus ν

C_ν = incoherent scattering length

$b_{\nu\nu'}$ = distance between nuclei ν and ν'

M = D_2O molecule mass

m = neutron mass

$$\frac{1}{B_{\mu}^{\nu\nu'}} = \frac{MQ_{\mu}^{\nu\nu'} T}{m\omega_{\mu} \sinh \frac{\omega_{\mu}}{2T}} \quad (4)$$

$$\lambda^{\nu\nu'} = \frac{M}{m} \sum_{\mu=1}^3 P_{\mu}^{\nu\nu'} \frac{\coth \omega_{\mu}/2T}{\omega_{\mu}/T} \quad (5)$$

$$\alpha = \frac{\kappa}{2MT}; \quad \beta = \frac{\epsilon}{T} = \frac{E-E_0}{T}$$

The coefficients $P_{\mu}^{\nu\nu'}$ and $Q_{\mu}^{\nu\nu'}$ are vibrational and torsional amplitudes which are tabulated in Butler's paper. The diagonal coefficients are equal: $P_{\mu}^{\nu\nu} = Q_{\mu}^{\nu\nu}$. The $\lambda^{\nu\nu'}$ and $B_{\mu}^{\nu\nu'}$ are also tabulated by Butler but

in our calculations they were recomputed by the input routine to GAKER. Actually the coefficient λ^{OD} listed by Butler is too small by a factor of 10 and this error invalidates his results for the OD term. κ and ϵ denotes as usual, the momentum and energy exchange.

The computing time for the exact evaluation of Eq (3) in the energy range 0.001 - 1.0 ev. is prohibitive, but fortunately it can be considerably reduced by making suitable approximations. The usual procedure for evaluating Nelkin's kernel is to split the energy range in two regions. In the first region one treats the first oscillator rigorously, the second and third oscillators in the low temperature limit (only the ground state is populated). Furthermore one only retains the elastic (0 phonon) term of the third oscillator.

At an energy such that the third oscillator can also be excited one then switches to another approximation in which the first oscillator is treated in the short collision approximation, the second oscillator is treated exactly (this actually is unnecessary: the low temperature approximation should be good enough for $w_2 = 0.205 \omega$), and the third oscillator is treated in the low temperature approximation. It is clear that this procedure used for the D self scattering can be extended immediately to the oxygen. However, for the interference scattering ($D_1 D_2$ and OD terms) the situation is different, as shown below.

In order to analyze the different approximations it is best to go back to Zermach and Glauber's intermediate scattering function:

$$\chi^{\nu\nu'} = \int e^{i\epsilon t} e^{-\frac{\beta}{2}} S^{\nu\nu'}(\alpha, \beta) \quad (6)$$

For a harmonic system $\chi^{\nu\nu'}$ is a product of the χ functions corresponding to each mode.

$$\chi^{\nu\nu'} = \tilde{a}_{\nu\nu'} \prod_{\mu} \chi_{\mu}^{\nu\nu'} \quad (7)$$

with

$$\tilde{a}_{\nu\nu'} = (A_{\nu} A_{\nu'} + C_{\nu}^2 \delta_{\nu\nu'}) \frac{\sin b_{\nu\nu', \kappa}}{b_{\kappa\kappa', \kappa}}$$

The expression for the factors $\chi_{\mu}^{\nu\nu'}$ leading to the result (3) is

$$\chi_{\mu}^{\nu\nu'} = \exp \left[-\alpha T P_{\mu}^{\nu\nu'} \frac{\coth \frac{\omega_{\mu}}{2T}}{\omega_{\mu}} \right] \cdot \exp \left\{ \frac{\alpha T Q_{\mu}^{\nu\nu'}}{2\omega_{\mu} \sinh \frac{\omega_{\mu}}{2T}} \left[\exp \left(\frac{\omega_{\mu}}{2T} - i\omega_{\mu} t \right) + \exp \left(-\frac{\omega_{\mu}}{2T} + i\omega_{\mu} t \right) \right] \right\} \quad (8)$$

The exact phonon expansion (3) is obtained using the formula

$$\exp \left[\frac{x}{2} \left(y + \frac{1}{y} \right) \right] = \sum_{n=-\infty}^{\infty} y^n I_n(x) \quad (9)$$

which gives:

$$\chi_{\mu}^{\nu\nu'} = \exp \left[-\frac{2T}{\omega_{\mu}} P_{\mu}^{\nu\nu'} \coth \frac{\omega_{\mu}}{2T} \right] \cdot \sum_{h=-\infty}^{\infty} I_{\mu} \left(\frac{m\alpha}{MB_{\mu}^{\nu\nu'}} \right) e^{-n \frac{\omega_{\mu}}{2T} + i n \omega_{\mu} t} \quad (10)$$

The low temperature limit is obtained by writing:

$$\frac{1}{B_{\mu}^{\nu\nu'}} = \frac{M Q_{\mu}^{\nu\nu'} T}{m \omega_{\mu} \sinh \frac{\omega_{\mu}}{2T}} \rightarrow 2 \frac{M Q_{\mu}^{\nu\nu'} T}{m \omega_{\mu}} e^{-\frac{\omega_{\mu}}{2T}} \quad (11)$$

and

$$I_n \frac{m\alpha}{MB_{\mu}^{\nu\nu'}} \rightarrow \frac{Q_{\mu}^{\nu\nu'} T \alpha}{\omega_{\mu}} \frac{|n|}{|n|!} e^{-\frac{n\omega_{\mu}}{2T}} \quad (12)$$

The terms with $n > 0$ can then be neglected and we get

$$\chi_{\mu}^{\nu\nu'} \text{LT} = \exp \left[-\frac{\alpha T}{\omega_{\mu}} P_{\mu}^{\nu\nu'} \coth \frac{\omega_{\mu}}{2T} \right] \sum_{n=0}^{\infty} \left(\frac{Q_{\mu}^{\nu\nu'} T \alpha}{\omega_{\mu}} \right)^n \frac{e^{-in\omega_{\mu} t}}{n!} \quad (13)$$

This approximate result applies both to self and interference terms. A different situation arises in the case of the short collision approximation which is obtained from (8) by expanding the exponent in powers of t and retaining only terms up to 2nd order.

$$\exp \left(\frac{\omega}{2T} - i\omega t \right) + \exp \left(-\frac{\omega}{2T} + i\omega t \right) = 2 \cosh \frac{\omega}{2T} - 2 i\omega t \sinh \frac{\omega}{2T} - \omega^2 t^2 \cosh \frac{\omega}{2T} + O(t^3) \quad (14)$$

Within this approximation

$$\chi_{\mu}^{\nu\nu'} \text{sc} = \exp \left\{ -\alpha T \left[\frac{P_{\mu}^{\nu\nu'} - Q_{\mu}^{\nu\nu'}}{\omega_{\mu}^2} 2 \bar{E}_{\mu} + Q_{\mu}^{\nu\nu'} (it + \bar{E}_{\mu} t^2) \right] \right\} \quad (15)$$

where

$$\bar{E}_{\mu} = \frac{\omega_{\mu}}{2} \coth \frac{\omega_{\mu}}{2T}$$

is the average energy corresponding to the oscillator of frequency ω_{μ} . Now it is clearly seen that a Fourier transform of $\chi_{\mu}^{\nu\nu'} \text{sc}$ exists only if the coefficient $Q_{\mu}^{\nu\nu'}$ is positive. This is always the case for the self terms $\nu = \nu'$, for which also $P_{\mu}^{\nu\nu} = Q_{\mu}^{\nu\nu}$ thus yielding the familiar result

$$\chi_{\mu}^{\nu\nu} \text{sc} = \exp \left[-\alpha T P_{\mu}^{\nu\nu} (it + \bar{E}_{\mu} t^2) \right]$$

For the interference terms, however, it turns out that $Q_1^{\nu\nu'}$ is negative. In fact

$$Q_1^{\text{OD}} = -0.051968$$

$$Q_1^{D_1 D_2} = 0.022003$$

Thus, in this case the coefficient of t^2 is positive and expression (15) becomes meaningless unless it is multiplied by another χ -factor with a larger negative coefficient of the t^2 term in its exponent. This would be the case if

$$-Q^{\nu\nu'} \bar{E}_1 < \frac{m}{M} T$$

since then the negative coefficient in χ_{Tr} , the χ -factor corresponding to the free translation of the D_2O molecules, would predominate.

For the self terms, where $Q^{\nu\nu'} > 0$, the effect of the short collision approximation for the lowest frequency is to increase the translational Doppler broadening of the resonances corresponding to the remaining frequencies. Schematically the exact and approximate P_0 kernel compare, as shown in Fig. 27, so that the result of the approximation is like taking an average over the fine structure due to the lowest frequency. When $Q_1^{\nu\nu'} < 0$, consecutive peaks of the kernel have opposite sign and the average tends to 0 as the number of peaks between $E=0$ and $E=E_0$ increases. Hence it seems reasonable to drop the interference terms completely in the energy region in which the self terms are calculated in the short collision approximation. With this criteria and a switching energy $E_s = 0.25$ ev we have computed the P_0 and P_1 kernels to be used in neutron spectrum calculations.

3.1.1 Results and Conclusions

A typical result of these calculations is shown in Fig. 28 where we compare the $D_\nu D_\nu$ and OD_ν contributions to the P_0 kernel.

The scattering law $S(\alpha, \beta)$ defined here by

$$S(\alpha, \beta) = \frac{1}{A_D^2 + C_D^2} \sum_{\nu\nu'} (A_\nu A_{\nu'} + \delta_{\nu\nu'} C_\nu^2) \frac{\sin b_{\nu\nu'} \kappa}{b_{\nu\nu'} \kappa} S^{\nu\nu'}(\alpha, \beta)$$

has also been calculated for several sets of α and β . Two examples are given in Figs. 29 and 30, again showing the relative contributions of all terms.

Fig. 27--Schematic representation of the short collision approximation

Fig. 28-- D_1D_1 and OD contributions to the P_0 kernel

Fig. 29--Scattering law $S(\alpha, \beta)$ for D_2O at $\beta = -0.5$

Fig. 30--Scattering law $S(\alpha, \beta)$ for D_2O at $\beta = -1.0$

Finally the total scattering cross section obtained from the present calculation is shown in Fig. 31. Surprisingly it is seen from this plot that the interference terms, negligible beyond about 0.03 ev, actually worsen the agreement between theory and experiment. In fact, whereas the calculated non interference cross section fits the BNL 325 data quite well down to about 0.0025 ev, the complete calculated cross section starts deviating around 0.03 ev.

In consequence it appears that the intra molecular interference contribution to the scattering cross section is probably compensated by intermolecular interference scattering due to liquid effects and not included in the present calculation. Thus, it must be concluded that it is unjustified to consider the intramolecular interference alone and that it is preferable to retain only the self terms, as it was done by Honeck. However, Butler's oscillator weights and frequencies are physically better founded and will be kept in the future, although the difference between Honecks' and Butler's parameters may have very little influence on integral quantities like total cross section or neutron spectra.

3.2 UNIVERSAL SCATTERING LAW CODE (GASKET-FLANGE)

One of the major difficulties in calculating scattering kernels is that the necessary machine codes are very limited in scope. For example, the codes GAKER⁽¹²⁾ and SUMMIT⁽⁵⁾ will handle only one type of atomic motion, and, in addition, these codes have numerical problems and usually require an expert to use and interpret the results. It would be highly desirable to have a computer code which would handle many different types of atomic motion, and be free of numerical difficulties. Also one of the major points of emphasis for this code would be simplicity of use so that reactor engineers could use the code in design calculations. These provide motivation for the development of the codes GASKET and FLANGE.

About 75% of the coding for FLANGE, which calculates point and group kernels from scattering law data, has been checked. Debugging will

Fig. 31--Total cross section of D_2O as a function of neutron energy

not be completed until the GASKET code, which computes the scattering law data, is nearly completed.

All coding for GASKET has been completed for a first form of the program. The debugging of the code will take place in portions, testing each of the routines listed in the previous quarterly report⁽¹⁾ separately.

For each value of $p^2/2$, the GASKET program reads a list of values of $S(p^2/2, \epsilon)$ and an associated list of delta functions (see Eq. 51 of Ref. 1). Point kernels are then calculated for the 0th up to the 3rd Legendre moment. A special integration routine has been written and tested for this integration. It integrates a vector function with up to four components, starting with an initial number (> 3 , odd) of integration points and halving the integration intervals until either the integrals converge with decreasing interval or a maximum number of integration points (> 5 , odd) is reached. A Richardson extrapolation (see Hildebrand, Introduction to Numerical Analysis) is performed using the last two values of the integral.

Group kernels are calculated according to Eq. 74 of Ref. 1 using the same basic integration and interpolation routines.

The routine to interpolate in the $S(p^2/2, \epsilon)$ array currently uses the Lagrangian interpolation routine TERP⁽¹³⁾. Another routine has been coded to perform the iterative interpolation described in Hildebrand. The iterative method offers the advantage of performing only as much calculation as is needed to achieve a specified accuracy, whereas the Lagrangian routine requires a preset order of interpolation, adequate for the worst cases to be encountered. The iterative method as it has been coded has the additional advantage of working with an unordered list of values. Present checks indicate that there is not a great saving in over-all time in using either of the methods.

3.3 COHERENT NEUTRON SCATTERING FROM LIQUIDS

Two of the most important reactor moderators are liquids, namely H_2O and D_2O . Other liquids such as hydrogen, deuterium and sodium also

play an important role in neutron physics. Up to the present time, however, all scattering kernels consider the molecular translations to be free, i. e., intermolecular correlations are assumed absent in the liquid state. While this approximation has proved to be generally adequate for integral parameters, especially when the neutron energies are high, for differential measurements purely liquid effects come into play. At present, there is very little even in the way of formalism that would be of help for the inclusion of liquid effects in current scattering kernels. As a start toward improving this situation, an investigation has begun toward calculating the coherent neutron scattering from liquids.

The scattering function $S(q, \omega)$, aside from trivial factors is proportional to the following thermal average:

$$\begin{aligned} & \langle\langle \sum_{i,j} e^{-i \vec{q} \cdot \vec{r}_i(0)} e^{i \vec{q} \cdot \vec{r}_j(t)} \rangle\rangle_T = \langle\langle \rho_{-q}(0) \rho_q(t) \rangle\rangle_T \\ & = \Omega^{-2} \langle\langle \sum_{kk'} C_{k-q}^+(0) C_k(0) C_{k'+q}^+(t) C_{k'}(t) \rangle\rangle_T \end{aligned}$$

where $\vec{r}_j(t)$ is the position operator of the j -th particle, $\rho_q(t)$ the Fourier component of the density operator, Ω the volume of the system and $C_k(t)$ is a Boson or Fermion annihilation operator. In the random-phase approximation (RPA) the equation of motion for a component of the density operator is

$$\left[i\hbar \frac{d}{dt} + \epsilon_{k+q} - \epsilon_k \right] C_{k+q}^+(t) C_k(t) = (n_{k+q} - n_k) V(q) \sum_{k'} C_{k+q}^+(t) C_{k'}(t) \quad (1)$$

Here ϵ is the particle energy, n_p the number density ($n_p = C_p^+ C_p$), and $V(q)$ the Fourier transform of the two-body interaction potential.

Define the retarded Green's function

$$g_r(k+q, k; k'-q, k') \equiv \frac{\theta(t)}{i\hbar} \ll [C_{k+q}^+(t) C_k(t), C_{k'}(0) C_{k'-q}(0)]_{\pm} \gg_T \quad (2)$$

where $\theta(t) = 1$ for $t > 0$, $\theta(t) = 0$, $t \leq 0$, $d\theta/dt = \delta(t)$.

Eq. (1) can now be written as

$$\begin{aligned} \left[i\hbar \frac{d}{dt} + \epsilon_{k+q} - \epsilon_k \right] g_r = \delta(t) \ll [C_{k+q}^+(0) C_k(0), C_{k'-q}(0) C_{k'}(0)]_{\pm} \gg_T \\ + (n_{k+q} - n_k) V(q) \sum_{k''} g_r(k''+q, k''; k'-q, k'), \end{aligned}$$

or, upon taking the Fourier transform in time

$$\begin{aligned} (\hbar\omega + \epsilon_{k+q} - \epsilon_k) \bar{g}_r(k, k') = \frac{1}{2\pi} (n_{k+q} - n_k) \delta_{k', k+q} \\ + (n_{k+q} - n_k) V(q) \sum_{k''} \bar{g}_r(k'', k'), \end{aligned}$$

The algebraic equation for $\bar{g}$ is then

$$\bar{g}_r(k, k') = \frac{n_{k+q} - n_k}{\hbar\omega + \epsilon_{k+q} - \epsilon_k} \frac{\delta_{k', k+q}}{2\pi} + V(q) \sum_{k''} \bar{g}_r(k'', k'),$$

and thus

$$\bar{g}_r(k, k') = \frac{1}{2\pi} \frac{n_{k+q} - n_k}{\hbar\omega + \epsilon_{k+q} - \epsilon_k} \left[\delta_{k', k+q} + \frac{V(q)}{1 - V(q)} \sum_{k''} \frac{n_{k''+q} - n_{k''}}{\hbar\omega + \epsilon_{k''+q} - \epsilon_{k''}} \sum_{k'''} \frac{n_{k''' + q} - n_{k'''}}{\hbar\omega + \epsilon_{k''' + q} - \epsilon_{k'''}} \right]$$

If we define an advanced Green's function by letting

$\theta(t)/i\hbar \rightarrow -1/i\hbar \theta(-t)$ in Eq. (2), we find that $\bar{g}_a(k, k')$ satisfies an exactly similar equation.

It can readily be shown then that $F(\omega)$ the Fourier transform of the correlation function

$$\frac{i}{\hbar} \ll \sum_k C_{k-q}^+(0) C_k(0) C_{k'+q}^+(t) C_{k'}(t) \gg \text{ is}$$

$$\Gamma(\omega) = \lim_{\epsilon' \rightarrow 0} \frac{\hbar}{i} \left[\Gamma_r(\omega + i\epsilon') - \Gamma_a(\omega - i\epsilon') \right] \frac{1}{\epsilon + e}$$

where $\Gamma' = \sum \bar{g}(k, k')$, and $\epsilon = \pm 1$ depending upon the choice of commutators or anti-commutators. $F(\vec{q}, \omega)$ is then identified with $S(\vec{q}, \omega)$ the scattering function.

While this work has just begun it is hoped that, as a first step, an adequate test of the Random Phase Approximation for neutron scattering can be made on liquid argon where the potential (Lennard-Jones) is known to rather good accuracy. If the test is successful, it will be possible to proceed to the important liquid moderators.

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Editor's Note: Copies of Appendix I, "Angular Distribution of Thermal Neutron Scattering from H_2O ," and Appendix II, "On the Analysis of Pulsed, Multiplying Systems," are included into only those copies of the report submitted to the agency.