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INTEGRAL NEUTRON THERMALIZATION

QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING MARCH 31, 1964

Contract AT(04-3)-167
Project Agreement No. 2
U.S. Atomic Energy Commission

April 17, 1964

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GENERAL ATOMIC
DIVISION OF
GENERAL DYNAMICS

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I. INTRODUCTION

The primary objectives of this program of research are to (1) develop scattering models for the common reactor moderators, (2) generate scattering kernels based on these models, and (3) compare the predictions of the models with experiments that are sensitive to specific model properties affecting reactor spectra or design.

During the first quarter of the program molecular or crystalline models for all the reactor moderators were studied. These models vary greatly in their refinements. Special attention was given to beryllium, D_2O , and CH_2 , and much of the effort went into the specification and coding of the new general scattering law code GASKET, which is designed to generate scattering kernels for any of the reactor moderators. It will, hopefully, be a fast production code and will replace the several codes that must now be used or adapted for use in calculations of the scattering matrices in reactor analysis.

In the experimental portion of the program the moderators H_2O , D_2O , and BeO were considered, and some of the problems of correlating experimental spectral results with theory were cleared up. Various auxiliary experiments were performed to improve experimental techniques and to try to shed some light on the new Garelis-Russell technique of measuring subcritical reactivity. Results for the angular scattering experiment are being compared with theory for both H_2O and vanadium, using a new rigorous treatment of the multiple scattering problem. This experiment is designed to test the adequacy of the angular properties of proposed scattering kernels in a manner useful for reactor analysis (essentially the transport properties).

Two papers originating from this program will be presented at the American Nuclear Society Meeting which will be held in Philadelphia in June, 1964. These papers are "Neutron Spectrum Measurements in a UF_4 + Paraffin Multiplying Assembly;" by J. C. Young, J. M. Neill, and J. R. Beyster, and "Pulsed Neutron Measurements of Reactivity in a UF_4 + Paraffin Multiplying Assembly," by J. L. Russell, Jr., J. R. Brown, and J. C. Young.

II. EXPERIMENTAL

2.1. Neutron Spectrum Measurements in D₂O

Measurements of quasi-infinite-medium neutron spectra in high-purity deuterium oxide poisoned with boron have been made at three positions within a rectangular assembly. These measurements were made to test realistic molecular models⁽¹⁾ generated to describe the integral neutron thermalization properties of the important reactor moderator D₂O, and to alleviate some of the discrepancies encountered in previous measurements⁽²⁾.

The improved geometrical arrangement used for these measurements is shown in Fig. 1. The dimensions of the assembly were 18 by 18 by 10 in. The poison was introduced as boron-aluminum foils (18 by 18 by 0.020 in. thick) accurately spaced every 1/8 in. The foils were placed so that the 18 by 18 in. face was perpendicular to the re-entrant tube. The three spectra were measured at 15.24, 22.86, and 30.48 cm from the source side of the assembly. Figure 1 shows the re-entrant tube in the 22.86-cm position.

The spectra were calculated using the bound deuterium kernel described in Ref. 1 and the computer code GAPLSN.⁽³⁾ The GAPLSN calculation used a P₀ and P₁ source. Three-dimensional cadmium-covered-indium flux plots were measured at each re-entrant tube position. The flux measurement axial to the source was made to supply the GAPLSN calculation with a spatial distribution for the input source. This measurement is shown in Fig. 2. The transverse spatial distribution measurements verified that the fluxes were cosines with a 3.5-cm extrapolation length, and were used to calculate the geometrical buckling which is utilized in GAPLSN to make leakage corrections necessary because of the finite dimensions of the assembly. Since the spatial source is put in GAPLSN at 2.38 ev, the measured distribution was modified in such a way that the calculated distribution matched the measured shape at indium resonance. The calculated spectra are compared with the experimental data in Fig. 3. The relative intensities of the three different positions are preserved for the calculated spectra. The experimental data are normalized at 1 ev to the 30.48-cm position, and the relative intensities of the other two positions were determined using sulphur and gold foil monitors.

As can be seen in Fig. 3 there is excellent agreement on relative intensity for the three experiments, as well as on the spectral shape. This indicates that the bound molecular model described in Ref. 1 is adequate for predictions of quasi-infinite-medium neutron spectra in D₂O.

Fig. 1--Geometrical arrangement used for measuring neutron spectra in borated D_2O .

Fig. 2--Spatial distribution of indium resonance neutrons axial to the source in the borated D_2O assembly.

Fig. 3--90° angular measurements in borated D₂O at various positions from the source.

The previous measurements⁽²⁾ made in D₂O were also in fair agreement, but this agreement may have been fortuitous, since a more accurate analysis of the boron concentration used in the experiments indicates an error in the analysis used in the previous calculations. This may indicate that the hydrogen contamination in the old geometrical arrangement may not have been handled adequately. The atom densities used in the present experiment are given in Table 1.

Table 1
ATOM DENSITIES FOR D₂O MEASUREMENTS
(In 10⁻²⁴ atoms/cm³)

N _{deuterium}	0.05672
N _{oxygen}	0.02849
N _{aluminum}	0.000915
N _{boron}	0.000143
N _{hydrogen}	0.000261

To check the transport properties of the D₂O scattering kernel more fully, measurements will be made in D₂O of the angular flux spectrum at the surface and in the presence of a strong flux gradient. Also, infinite-medium neutron spectra in D₂O heavily poisoned with a resonance absorber such as cadmium or samarium are being planned, first at room temperature and eventually in the pressure vessel.

2.2. Neutron Spectrum Measurements in BeO

It was decided to remeasure a quasi-infinite-medium spectrum in an assembly of BeO for the following reasons:

1. Experimental techniques have been improved since the last measurements were made.⁽⁴⁾
2. Sinclair's⁽⁵⁾ frequency spectrum has been used to generate a new scattering kernel for BeO, and it was considered desirable to test this kernel.
3. It was hoped that if good agreement could be obtained between the measured and the theoretical spectra, BeO could then be used in

studying spectral changes at absorption discontinuities. The slowing down power of BeO is small enough that spectral changes with position would not be too large and could easily be measured.

Earlier measurements⁽²⁾ had shown that it was not possible to excite a BeO assembly easily into fundamental modes in each direction by gamma excitation, utilizing the (γ, n) reaction on beryllium. It would be desirable to do this so that the leakage corrections, expected to be large, would be more amenable to accurate calculation. It was therefore decided to proceed first by performing measurements on BeO in the standard geometry as illustrated in Fig. 4.

The BeO assembly was poisoned with 0.010-in. sheets of 1 wt-% borated stainless steel to give about 1.0 barns/beryllium atom. Flux plots were taken in the axial and horizontal transverse directions along the central axes by means of cadmium-covered indium foils. The horizontal transverse plot gave an approximate cosine shape, and the axial plot is illustrated in Fig. 5. The central portion of this curve can be approximated well by an exponential function, so that the leakage correction along this axis at the center can be calculated fairly accurately. A neutron spectrum measurement was taken under the experimental arrangement of Fig. 4 and is shown in Fig. 6. The spectrum is very hard because of the high ratio of absorption to slowing down power and because of large leakage. This large leakage also manifested itself by a very high background-to-signal ratio in the measurement. This is the reason why the statistics below 0.01 ev are poor and are reflected by the scattering of the flux points. No calculation has yet been made to compare with this measurement. Further measurements as a function of position are planned. The comparison between theory and experiments will be covered in a subsequent report.

2.3. Neutron Spectrum Measurements in H₂O

The work on thermal-neutron spectra in poisoned water was continued by measuring a number of 90-degree angular spectra in the thin-tank geometrical arrangement shown in Fig. 7. The water poison concentration of about 5 barns/hydrogen atom (43.7 g/liter of boric acid) was the same as that used in experiments reported earlier.⁽²⁾

In order to eliminate the possibility of room return neutrons and to improve the boundary conditions over those of previous measurements, a 1-in. slab of boron carbide was placed on both sides of the 12 in. by 12 in. by 4 in. tank containing the borated water. In addition to the conventional BF₃ monitor counter, an aluminum monitor was placed over the water-cooled fansteel target and two gold foils were located close to the re-entrant tube on the 12 in. by 12 in. side of the tank, facing the precollimator.

Fig. 4--Geometrical arrangement used to measure infinite medium neutron spectra in borated BeO.

Fig. 5--Spatial distribution of Indium resonance neutron in borated BeO.

Fig. 6--Infinite medium neutron spectra measured in borated BeO.

Fig. 7--Geometrical arrangement used to measure 90° angular spectra in borated water.

Fairly good agreement was obtained between the three monitoring systems, and the results are used in the normalization of the measured neutron energy spectra.

The 90-degree angular measurements were made at the surface at 2.54, 5.08, and 7.62 cm from the precollimator side of the 4-in. slab. These energy spectra, along with the corresponding theoretical spectra calculated with the GAPLSN transport-theory code, will be presented in the next progress report. Preliminary indications are that improved agreement between the transport calculation and the experimental results has been obtained.

Cadmium-covered indium foils were used to measure the spatial dependence of the neutron flux in three mutually perpendicular directions at the position of the re-entrant tube. Of these the 4-in. transverse flux plot is used as the epithermal-source distribution in the GAPLSN calculations.

2.4. Sensitivity of the 16-M Detector Bank

Further effort has been made to determine more accurately the sensitivity of the bank of 32 BF_3 counters used on the 16-m flight path. This sensitivity, $S(E)$, must be known accurately because it enters directly into the determination of neutron spectra measured under this thermalization program.

The sensitivity is usually measured by comparing the bank's response for a given incident flux to the response from a comparison detector whose efficiency is known. At present the effort is being directed to resolving the differences in the $S(E)$ from two comparison detectors: a lithium glass scintillator and a bank of 10 low-pressure BF_3 counters with a $1/v$ response. The problem is discussed in detail in Ref. 6.

The post-collimator on the 16-m flight path has been surveyed across the horizontal axis with the lithium glass crystal, and no significant change in the shape or intensity of neutron spectra with position was observed. This eliminated one possible cause for the discrepancy.

The effect of neutron reflection from the material behind the 16-m detector system has also been investigated. The backing material used was successively 1/4 in. borated polyethylene, 0.020 in. cadmium, 1/4 in. borated polyethylene, and finally 0.020 in. cadmium plus 1/4 in. borated polyethylene and 1 in. of boron carbide. The leakage spectrum of pure polyethylene was measured under these four conditions by the $1/v$ detector bank and no change in the shape of the measured neutron spectra could be observed. The results were therefore combined to calculate a new $S(E)$ for the 32-detector bank which agreed with the previous accepted set of values over the reduced experimental energy range.

$S(E)$ was then remeasured over the usual energy range using the $1/v$ bank as a comparison detector. It was found to be perhaps slightly lower below 0.01 ev, but otherwise the same as the accepted values, and is shown in Fig. 8, together with a previously calculated $S(E)$.⁽⁶⁾ The $S(E)$ measured with the lithium glass detector and reported in Ref. 6 has been reanalyzed because monitor drift was found during its measurement, which accounted for the peculiar values of the sensitivity at low energies. The monitor drift was corrected for, and the resultant $S(E)$ is much more normal and is shown in Fig. 9, together with the calculated $S(E)$ normalized over the energy range 0.001 to 0.5 ev. The low energy values of $S(E)$ shown in Fig. 9 can be ignored because of their poor statistics. When $S(E)$ is normalized in this manner, it appears that the disagreement between the two measured sensitivities is at higher energies. It may be possible to resolve this disagreement by a more accurate analysis of multiple scattering in the lithium glass crystal. This analysis has already been started.

2.5. Neutron Monitor Studies

During any measurement of neutron spectra, the intensity of the pulsed neutron source must be monitored to provide normalization of the spectra at various positions in the assembly under measurement, and to allow for the correct subtraction of the background spectra.

In the past, monitoring has been done by a fission counter, but this method has not always proved satisfactory. It is desirable to locate this counter and its preamplifier and power supply at some distance from the source in order to reduce the effects of neutron and gamma radiation on the equipment, as these effects impair its long-term stability. However, it is necessary to attach the monitor to the assembly or an extension of it so as to maintain the same source-assembly-monitor configuration when the assembly and source are moved together past the precollimator of the fixed time-of-flight path. In practice it has been difficult to achieve this reliably; therefore, other forms of monitoring have been studied as alternatives.

The use of foils was immediately considered, and activation of gold foils has in fact been used before. The difficulty with the use of gold foils is that their activation is predominantly by the absorption of thermal neutrons, whereas it is desired to monitor the fast-neutron flux from the source. In using the gold foils, then, it has been necessary to attach them to the measured assembly so that they are shielded from room return neutrons, and to position them uniformly about the source axis so as to reflect any shift in the position of the electron beam on the target. In practice it has not always been possible to achieve these objectives.

Fig. 8--Sensitivity of 32 bank of Bf_3 's determined using a $1/v$ detector in comparison to the calculated value for the bank.

Fig. 9--Sensitivity of bank of ^{32}Bf 's determined using a Li^6 scintillation in comparison to the calculated value for the bank.

Sulphur foils have also been used, utilizing the $S^{32}(n,p)P^{32}$ reaction, which has a threshold of about 1 Mev. P^{32} decays by strong beta rays, so the sulphur foils should provide reasonable monitoring of the primary flux. The sulphur foils were used in groups of four, placed symmetrically about the source on an aluminum holder. The cross section for the reaction is low and required the close proximity of the foils to the source. Unfortunately, there was too much heat dissipated in this area, and the sulphur foils became flaky or pasty and did not prove satisfactory. Finally, monitoring by aluminum was tried using the $Al^{27}(n,\alpha)Na^{24}$ reaction, which has a threshold of some 6 Mev. Na^{24} decays by strong gamma rays and can readily be counted. The aluminum was machined into the shape of an end cap and was slipped over the rear of the target. It is recognized that this kind of positioning will be satisfactory only when the source is at the front, not at the side, of the assembly under measurement.

Table 2 compares the results of these monitors for a set of runs in the form of counts per pulse normalized by average values. How near the values are to each other in any one row of this table points to the adequacy of the monitoring devices. It would appear in this set of runs that the fission monitor was reliable. The agreement between the values along each row is reasonably good. It may be concluded that the use of the aluminum and gold foils will provide monitoring accuracy to within 2-1/2%. The fission counter serves as a basic monitor which gives more immediate values of the neutron source intensity than do the gold or aluminum foils, and should therefore be used as well.

Table 2
COMPARISON OF NEUTRON-MONITOR DATA

Run	Normalized Counts Per Pulse		
	Aluminum	Gold	Fission Counter
1	0.896	0.939	0.947
2	---	1.104	1.025
3	1.065	1.040	1.048
4	---	1.044	1.034
5	1.069	1.037	1.044
6	---	1.141	1.051
7	1.102	1.083	1.044
8	---	1.191	1.010
9	0.868	0.900	0.914
10	---	0.822	0.856

It has also been discovered that the shape of the low-energy end of a measured neutron spectrum is sensitive to drifts in the monitor, especially when the background-to-signal ratio is high. A systematic method for correcting for this drift has been developed, so that the fission counter can be used with confidence as the normalizing device for purposes of background subtraction.

2.6. Pulsed Neutron Measurements on UF₄-Paraffin

An assembly of blocks of a homogeneous mixture of UF₄ and paraffin which had been constructed for spectrum measurements⁽⁶⁾ was used for a short study of the effect of a reflector on the pulsed neutron determination of reactivity by the $\kappa\beta/\ell$ technique.⁽⁷⁾⁽⁸⁾

This UF₄-paraffin mixture has been extensively studied at Oak Ridge National Laboratory.⁽⁹⁾ The uranium is 2 wt-% enriched and the H:U²³⁵ atomic ratio is 195. Some of the parameters of the system as obtained by ORNL are listed in Table 3.

Table 3
UF₄-PARAFFIN DATA^a

Atom densities:	
U ²³⁸	7.79×10^{21} atoms/cm ³
U ²³⁵	1.61×10^{20}
C	1.52×10^{22}
F	3.18×10^{22}
H	3.15×10^{22}
Critical buckling	$(4344 \pm 65) \times 10^{-6}$ cm ⁻²
Neutron age to thermal	43.1 ± 3.4 cm ²
Bare extrapolation distance	2.7 ± 0.3 cm
H:U ²³⁵ atom ratio	195
Wt-% UF ₄	92.1
Mixture density	4.5 g/cm ³
U ²³⁵ enrichment	2.0 wt-%
K _∞ (from two experiments)	$\begin{cases} 1.216 \pm 0.013 \\ 1.197 \pm 0.015 \end{cases}$
Thermal diffusion area, L ²	1.8 cm ²
Reflector savings	5.5 cm

^aFrom Ref. 9.

The amount of material available for these experiments permitted the building of a bare assembly 16 in. by 16 in. by 17 in. high. As the assembly is unreflected, the reactivity is approximately \$39 subcritical.⁽⁶⁾

The Garelis-Russell method⁽⁷⁾ shows that for a one-group, homogeneous reactor the delayed neutron "background" in a pulsed neutron source measurement is related to the prompt component of the flux by

$$\bar{\phi}_d = R \int_0^{1/R} \phi_p \left[\exp \left(\frac{\kappa\beta}{\ell} t \right) - 1 \right] dt, \quad (1)$$

where $\bar{\phi}_d$ = average value of the delayed neutron flux,

ϕ_p = time-dependent prompt-neutron flux,

R = pulse repetition rate,

t = time,

$\kappa\beta/\ell$ = constant of the system.

In Ref. 8 it was shown that this analysis was valid for a bare system and that, in fact, the determination of $\kappa\beta/\ell$ did not depend on the position of measurement.

To investigate the effect of a reflector on this method of analyzing the experimental data, experiments were carried out on the UF₄-paraffin assembly with a reflector on one side. The reflector was either a 4-in.-thick paraffin slab or a 4-in.-thick water solution of 10.8 g/liter of H₃BO₃. The entire assembly was cadmium covered. Placing the reflector on one side only was expected to provide an extreme case for checking the method of Garelis and Russell.

A small 0.25-in.-diameter U²³⁵ fission counter was placed in various positions in the core and reflector to measure the time distribution of neutrons following repetitive pulsing of the assembly by the electron linear accelerator (LINAC). The source location was also changed for several measurements. In one measurement, a cadmium-covered U²³⁸ fission counter was used for comparison with the U²³⁵ counter and was found to give the same result.

In the first measurements in the core and in the reflector it was observed that at a pulse repetition rate of 60 pps, the fundamental mode decay had not been attained by the time the delayed neutron "background" had been reached. As a result it appeared that the α was different in the core from that in the reflector. Once the fundamental-mode decay is established, all parts of the assembly will have the same α .

To demonstrate that a fundamental-mode decay did exist, the assembly was pulsed at very low repetition rates of 0.84 and 0.43 pps. The full source intensity for the LINAC, approximately 10^{12} neutrons per pulse, was used, with a long delay ($\sim 800 \mu\text{sec}$) prior to the start of the multichannel time analyzer. Because of the large source strength it was possible to get reasonable counting rates at these long times after the pulse. Figures 10 and 11 show the combined results of the measurements at 60 pps, 0.84 pps, and 0.43 pps with the delayed neutron background subtracted. The positions of measurement and source location are shown in Fig. 12. The measurement positions are along a line through the core center and reflector center.

It can be seen in Figs. 10 and 11 that for this extreme case, time modes close in magnitude to the fundamental exist and that the fundamental is just beginning to be observed at very long times after the pulse. One effect which was observed from these measurements at long times after the pulse is that this can be carried only so far in time, since room return begins to be observed. From Figs. 10 and 11 the α determined from times beyond $1300 \mu\text{sec}$ is 163 sec^{-1} in the core and 183 sec^{-1} in the reflector, indicating that they have probably not yet quite reached the fundamental mode. Within the uncertainties in these measurements, a value of $170 \pm 10 \text{ sec}^{-1}$ probably represents the fundamental-mode α , since both core and reflector seem to be approaching this value from opposite directions.

Table 4 gives the results obtained for each of the positions measured. The $\kappa\beta/\ell$ is obtained from the integration of the data according to the Garelis-Russell expression as given in Eq. (1). All these measurements were made with a pulse repetition rate of 60 pps except for those of the U^{238} counter, which was run at 30 pps. The results of Table 4 are shown schematically in Fig. 13.

It can be seen that the $\kappa\beta/\ell$ determined for this core depends upon position and is thus not a constant for the system and as such is meaningless for this extreme case. This is a result of the interplay between several time modes, one associated with the core, one with the reflector, one with the combination of the two, and perhaps others. These different time modes do not differ greatly in the present situation, so the effect of each is observed at relatively long times.

More analysis along the lines of trying to separate these time modes and associate a $\kappa\beta/\ell$ with each is indicated. Some preliminary hand unfolding of some of these data for the paraffin reflector case indicates that a large part of the observed neutron flux versus time behavior may be made up of various magnitudes (positive and negative) of the following approximate time decay modes: 170, 130, 110 and $20 \mu\text{sec}$. If it were possible to fit the experimental data with a sum of exponentials (by a machine code), it would in principle be possible (using the Garelis-Russell integral) to assign a

Fig. 10--Die-away of neutrons at center of core.

Fig. 11--Die-away of neutrons at center of reflector.

Fig. 12--Pulsed source locations (A, B, C) and detector positions (1, 2, 3, 4) used in the measurements.

Table 4
RESULTS OF $\kappa\beta/\ell$ DETERMINATIONS

Core Condition	Configuration	$\kappa\beta/\ell$	α^{-1} (μsec)
Bare Core	A-2	226	110
Paraffin Reflector	A-2	179	---
	A-4	202	---
	B-1	218	---
	B-2	216 (208 ^a)	163
	B-4	102	183
	C-2	180	---
	C-3	47	---
Borated Reflector	B-2	209	117
	B-4	95	---

^a Using cadmium-covered U^{238} fission counter.

$\kappa\beta/\ell$ to each of the time modes and, if there were as many positions of measurement as there were individual time modes, to obtain the $\kappa\beta/\ell$ for each mode in the following way:

$$(\bar{N}_D)_x = R \sum_{i=1}^m \frac{Q_i}{\alpha_i} C_i,$$

where x = position of measurement,

i = time mode,

m = number of modes,

Q_i = magnitude of mode i at $t = 0$,

α_i = reciprocal of dieaway time of mode i (sec^{-1}),

C_i = unknown value of $\kappa\beta/\ell$ for mode i ,

R = repetition rate,

$\bar{N}_D$ = average delayed neutron flux.

This latter analysis has been attempted by hand without much success owing to the fact that when two decays are not greatly separated in time, the hand method of unfolding is not sufficiently accurate.

Fig. 13--Results of $k\beta/l$ determination for various combinations of source and detector.

In summary, the result is that the Garelis-Russell method does not work in this extreme case but is probably still useful for reflected reactors not so subcritical and for bare systems.

2.7. Reanalysis of Multiplying-assembly Spectrum Measurements

Spectrum measurements were made in 1962 on a sub-critical multiplying assembly composed of uranium-aluminum plates in water.⁽²⁾⁽¹⁰⁾ An improved analysis of the data was recently made using a more accurate mean-emission-time correction and systematically removing the background arising from delayed neutrons. The procedure for removal of this background is described in Ref. 6.

The agreement between theory and experiment has been improved for both the angular flux measurements illustrated in Fig. 14 and the scalar flux measurements illustrated in Fig. 15. The surface leakage measurement, which disagreed considerably with the calculations, is not shown, since the inaccuracy of the measurement is believed to arise from a positional error.

2.8. Angular Distribution Experiment for H₂O And Vanadium

A number of activities were carried out on the scattering angular distribution experiment during this reporting period. Further experiments were performed on light water and on vanadium as a check on the experimental method. The experimental results were analyzed in terms of absolute cross sections without normalization to the total-cross-section data, and a rigorous method of calculating multiple scattering corrections was developed and compared with our older procedures. In addition, a large number of checks on the experimental apparatus were performed to examine background sources, detector gamma sensitivity, collimation validity, etc. Finally the new flight path designed exclusively for this experiment was installed and much of the necessary shield structure was set up.

The newer angular distribution data for water will not be given here, since they essentially agree with the older data.⁽²⁾ It is difficult on the basis of these data to detect discrepancies with the predictions of the current bound hydrogen molecular model. It will be recalled that agreement to within 10% or 20% with theory at most angles had been obtained in the past except at the forward angles. At the forward angles, difficulties are being found in obtaining agreement on $\sigma(E, \mu)$ calculated with three different versions of the same bound-hydrogen molecular model, so it is not clear yet how good the comparison will be. It is still the objective, with the improved experimental arrangements of the new flight path, to determine this angular distribution for water, and the energy-dependent transport cross sections to which it leads, with high precision in this contract year.

Fig. 14--O⁰ angular neutron spectra in a water moderated multiplying assembly.

Fig. 15--Scalar neutron spectra in a water moderated multiplying assembly.

These data, coupled with the previous precision total-cross-section, infinite-medium spectrum measurements and space-dependent spectra measurements, will be adequate to establish the validity of the important features of our molecular models for water.

As a check on the experimental apparatus, the scattering angular distribution was measured from vanadium an incoherent scatterer. A spherically symmetric distribution was not observed, but the same distribution was observed for two sample thicknesses differing by a factor of three. The angular distribution is made up of two components: the elastic scattering from the crystalline lattice and the inelastic scattering from the lattice. To evaluate both, the lattice frequency spectra, $\rho(\omega)$, must be known. For our calculations we used the Eisenhauer-Pelah frequency distribution⁽¹¹⁾ shown in Fig. 16. The elastic scattering cross section, important mainly at forward angles and the lower neutron energies, is given by

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_{\text{incoh}}}{4\pi} \exp\left[-\kappa^2/2m \int \frac{\rho(\omega)}{\omega} \coth\left(\frac{\omega}{2T}\right) d\omega\right]$$

The neutron inelastic scattering cross section is calculated from the code SUMMIT II in a manner similar to that used for carbon, beryllium or any other crystalline moderator. The experimental and theoretical angular distributions at present do not agree in shape as well as expected, and various causes for this are being investigated.

An evaluation of the importance of multiple scattering in the samples was performed during the quarter. For any scattering data to be useful to the theorist, these nonnegligible corrections must be accurately made to obtain absolute (unnormalized) angular cross sections. The first procedure used in the data analysis was the Vinyard method⁽¹²⁾ of correcting for multiple scattering. Second and subsequent collisions in the foil are assumed to give rise to isotropic scattering angular distributions; energy change in collision is not taken into account. Absolute cross sections were obtained by normalization to the known total cross section. The first step has been to remove the last procedure from the analysis of the data so that absolute cross section values are obtained directly from the angular distribution experiment. The second procedure was a rigorous technique for calculating multiple scattering in the sample developed by H. Honeck of Brookhaven National Laboratory. (A General Atomic report is in preparation on the method.)

Extensive studies of both procedures for calculating sample corrections have been made. The results of two of these studies are shown in Fig. 17 and 18. In Fig. 17 a comparison is made of the correction to the data for multiple scattering for two incident neutron energies. C (the ordinate) is

Fig. 16--The Eichenhauer-Pelah frequency distribution for vanadium.

Fig. 17--Multiple scattering corrections calculated using two techniques as a function of scattering angle.

Fig. 18--Multiple scattering corrections calculated using two calculational techniques for $\theta_s = \theta_o$.

the factor by which uncorrected cross sections must be multiplied to compensate for the effects of multiple scattering (energy change in collision is included). The analysis is done for the case where the sample is oriented such that a normal to the sample will bisect the angle between incident and scattered beam. It is interesting that the Vinyard procedure does a relatively good job of correcting for multiple scattering except at back angles for high incident neutron energies. Even here the error made is not as large as might have been expected. A second study, in Fig. 18, shows the importance of sample thickness to the correction factor C . A beam normally incident on the sample is assumed for convenience. Three sample thicknesses are shown--10, 21.5, and 32 mils of water. The neutron energy is 1 ev. Surprisingly, unless one goes to ridiculously thin samples, the multiple scattering cannot be significantly reduced at back angles. Also, the forward angle scattering is very little perturbed by multiple scattering even for rather great thicknesses. Parenthetically, it is felt that it would be enlightening to actually try the angular distribution experiment for sample thicknesses covering a very large range--10 mils to 150 or 200 mils--since we now have an accurate method of calculating the neutron transport in the sample. This procedure will further check the analytical methods and also the properties of the theoretical scattering kernel, which of course must be used extensively in making these corrections.

A number of auxiliary studies have been performed to establish the techniques used in the $\sigma(E, \mu)$ experiment and to try to find out where improvements can be made in the setup. Each experiment will be discussed only briefly below.

1. To determine absolute cross sections, the size and shape of the neutron beam hitting the scattering sample must be known. This was studied with the Li^6 glass detector masked to admit only a 1/8-in. beam. The beam size was found to be 2 in. within the spatial resolution available (which is not as good as will be used in the future).

2. Detector gamma sensitivity was investigated by a number of techniques, since it was feared that the ~ 2 -Mev hydrogen capture gamma ray would not be subtracted by our normal background subtraction techniques. This appears not to be a problem.

3. The experiments were repeated with helium instead of air in the scattering chamber to lessen spurious scattering of the neutron beam. This step improves the background considerably (>50%).

4. Neutron wall scattering in the shield housing the samples and detectors was studied and was found to be nonnegligible. The new housing will as a result be larger and have more low-energy neutron shielding in the walls.

5. A strange phenomenon was found in the Li^6 detector when the detector was located at zero degrees. Under certain electron beam conditions a large gamma-flash-induced effect on the pulse height distribution can be obtained. Source shielding was designed to eliminate this as well as to maintain the high source intensity.

6. Total-cross-section and other studies were made of the sample materials used in the scattering experiment to establish that the samples, their placement, and methods of support were not giving spurious results.

The new scattering setup, shown in Fig. 19, is nearly complete. Many of the new features have been discussed above. One problem was getting the electron beam to the target position, which is located 8 ft off the beam axis. However, with a simple system of independent quadrupole lenses and steering magnets, 95% of the electron beam can be placed on the target in a 1/4-in. spot. This is a surprising (and welcome) result and was the only real uncertainty in the new experimental arrangement. Experiments are expected to begin about April 1 with the new setup.

Fig. 19--Geometrical arrangement used in the angular distribution measurements.

III. THEORETICAL

3. 1. Spectra in Heterogeneous Media

The desirability of obtaining measurements of the thermal neutron spectra under the ideal conditions of an infinite homogeneous medium which is excited by a spatially flat source of fast neutrons is obvious. This physical situation permits analysis within a simple mathematical framework.

In practice, however, the theoretically ideal conditions of measurement can only be approached. In the class of experiments we have been considering in the past, it has been our intention to reduce the dependence of the measurements on phenomena of a specifically transport and geometrical nature, and to emphasize instead the dependence of the spectrum on the scattering kernel $\Sigma_0(E', E)$.

The largest perturbations of the ideal infinite medium conditions have origins of a geometrical nature. The effects which must be considered are associated with (1) the possible heterogeneous composition of the moderating assembly, (2) the finite size of the assembly, and (3) the spatial variation of the neutron source. For a completely meaningful comparison of measured and calculated neutron spectra, it is advantageous that the experiment be designed in such a way that the spectral effects induced by the geometry are negligible. This condition pertains in most measurements of neutron spectra in homogeneously poisoned, bare, hydrogen-moderated assemblies, which are very large in comparison with a mean free path of a thermal neutron. In the case of moderation by heavy nuclei, such as carbon or beryllium, the spectral perturbations, though small, are significant even in quite heavily poisoned systems. Fortunately, the largest geometrical perturbations of the neutron spectrum are amenable to quantitative calculation by means of simple techniques.

In most experimental determinations of the neutron spectrum, a moderately high neutron absorption is desirable in order to emphasize the sensitivity of the spectrum to the effects of chemical binding, as well as to conform to the physical conditions encountered in practical problems of reactor design.

Ideally, in this class of experiment the absorbing material should be homogeneously distributed throughout the assembly. In systems moderated by light and heavy water, the ideal condition can be realized

by forming aqueous solutions with chemical compounds which contain some neutron-absorbing element such as boron or samarium. In solid moderators we approach the ideal condition by using thin sheets of absorbing material placed less than a mean free path apart. In general, it is not difficult to achieve a reasonable compromise between high absorption on the one hand and a low self-absorption and flux depression on the other.

Thus, the experiment is designed to minimize the effects of the heterogeneous distribution of poison. It still remains, however, to correct for the residual effects. Further techniques for handling heterogeneous absorbers are necessary for analysis of subsequent experiments planned under the thermalization program. In most experiments involving thermalization by solid moderators, the absorber is present in the form of thin foils of thickness $2a$ which are separated from one another by less than a mean free path of moderator. The macroscopic absorption cross section $\Sigma_a^F(E)$ of the foil material is usually much greater than the macroscopic scattering cross section, so to a good approximation the foil may be considered as a pure absorber.

To see how to estimate the effects of the heterogeneous composition of an assembly on a measured neutron spectrum, we consider a one-dimensional, infinite, periodic array of identical thin foils whose centers are a distance $2d \gg 2a$ apart. The space between the foils is uniformly filled with moderating material. Assume that the foils are purely absorbing and that the moderator atoms scatter neutrons isotropically. The isotropic slowing-down source which feeds the thermal energy range is spatially flat in the moderator and, since the material in the foils does not scatter, it vanishes in the absorber.

The angular flux spectrum $f(z, \mu, E)$ in the moderator satisfies the steady-state equivalent:

$$\left[\mu \frac{\partial}{\partial z} + \Sigma_a(E) + \Sigma_s(E) \right] f(z, E, \mu) = 1/2 \int \Sigma_0(E', E) \phi(z, E') dE' + S(E) \quad (2)$$

In the foil,

$$\mu \frac{\partial f}{\partial z}(z, E, \mu) + \Sigma_a^F(E) f(z, E, \mu) = 0 \quad (3)$$

Because of the spatially periodic distribution of foils and moderator, $f(z, E, \mu)$ satisfies the conditions

$$\begin{aligned} f(d, E, \mu) &= f(d, E, -\mu) & (a) \\ f(0, E, \mu) &= f(0, E, -\mu) & (b) \\ f(z, E, \mu) &= f(-z, E, -\mu) & (c) \end{aligned} \quad (4)$$

as well as the usual condition of continuity with respect to z .

To an extremely good approximation, for $a \ll d$ the quantity that is measured in the class of experiment being considered in the spatially averaged flux spectrum in the moderator, i. e.,

$$\begin{aligned}\phi_m(E) &= \frac{1}{d-a} \int_a^d dz \int_{-1}^1 f(z, E, \mu) d\mu \\ &\approx \frac{1}{d} \int_0^d dz \phi(z, E) .\end{aligned}\tag{5}$$

We can obtain an equation for $\phi(E)$ by integrating Eq. (2) over all μ and over all positions extending from a to d :

$$\begin{aligned}-\frac{1}{d-a} \int_{-1}^1 \mu [f(a, E, \mu) - f(d, E, \mu)] d\mu + [\Sigma_a(E) + \Sigma_s(E)] \phi_m(E) \\ = \int \Sigma(E', E) \phi_m(E') dE' + S(E) .\end{aligned}\tag{6}$$

Similarly, integrating Eq. (3) over all angles and over the interval from 0 to a gives

$$\frac{1}{a} \int_{-1}^1 \mu [f(a, E, \mu) - f(0, E, \mu)] d\mu = \Sigma_a^F(E) \phi_i(E) ,\tag{7}$$

where

$$\phi_i(E) = \frac{1}{a} \int_0^a dz \int_{-1}^1 f(z, E, \mu) d\mu\tag{7a}$$

is the average flux per unit energy in the foil. Combining Eqs. (7) and (6), taking into account Eq. (4), gives

$$\frac{a}{d-a} \Sigma_a^F(E) \phi_i(E) + [\Sigma_a(E) + \Sigma_s(E)] \phi_m(E) = \int \Sigma(E', E) \phi_m(E') dE' + S(E) ,\tag{8}$$

which is just the balance equation for neutrons with energy E , regardless of their position or direction of motion.

The theoretical problem of homogenizing the heterogeneously distributed absorber throughout the volume of the assembly consists in finding an approximate relation between $\phi_i(E)$ and $\phi_m(E)$. The simplest approximation consists in setting $\phi_i(E) = \phi_m(E)$. Since the foil tends to shield its interior from neutrons at its surface, this leads to an overestimate of the rate at which neutrons are absorbed.

To estimate the effects of self-shielding, we start from Eq. (3). This equation admits the solution

$$f(z, E, \mu) = f(a, E, \mu) \exp[\Sigma_a^F(E)(a - z)/\mu], \quad |z| < a. \quad (9)$$

Equation (9) allows us to relate the absorption rate in the foil to the angular flux at its surface. As a first step in obtaining a simple connection between the $\phi_i(E)$ and $\phi_m(E)$ we assume an isotropic surface flux incident on the slab:

$$f(a, E, \mu) = \frac{\phi_-(E)}{2} \quad \text{for } \mu < 0. \quad (10)$$

Observing the symmetry implied by Eq. (4c), we find

$$\begin{aligned} a\phi_i(E) &= \int_{-a}^a dz \int_{-1}^0 f(z, E, \mu) d\mu = \frac{\phi_-(E)}{2\Sigma_a^F(E)} \int_{-1}^0 d\mu \mu [\exp(2\Sigma_a^F(E)a/\mu) - 1] \\ &= \frac{\phi_-(E)}{2\Sigma_a^F(E)} \{1/2 - E_3[2a\Sigma_a^F(E)]\}. \end{aligned} \quad (11)$$

It remains to relate the incident flux $\phi_-(E)$ to the average flux in the moderator. To do this we first note that

$$f(a, E, \mu) = \frac{\phi_-(E)}{2} \exp[-2a\Sigma_a^F(E)/\mu] \quad \text{for } \mu > 0. \quad (12)$$

The exponential factor just gives the attenuation of the flux $f(-a, E, \mu)$ ($\mu > 0$) after transmission through the slab. We now assume that the flux spectrum in the moderator is independent of position. It then follows from Eqs. (10) and (12) that

$$\phi_m(E) = \int_{-1}^1 f(a, E, \mu) d\mu = \frac{\phi_-(E)}{2} \{1 + E_2[2a\Sigma_a^F(E)]\}. \quad (13)$$

Using this result together with Eq. (11) in Eq. (8), we find

$$[\Sigma_a^{\text{eff}}(E) + \Sigma_s(E)] \phi_m(E) = \int \Sigma_o(E', E) \phi_m(E') dE' + S(E) , \quad (14)$$

where the cross section which takes into account the self-shielding effect is given by

$$\Sigma_a^{\text{eff}}(E) = \Sigma_a(E) + \frac{a}{d-a} \Sigma_a^F(E) K[2a\Sigma_a^F(E)] \quad (15)$$

with

$$K(x) = \left[\frac{1 - 2E_3(x)}{2x} \right] \frac{2}{1 + E_2(x)} . \quad (16)$$

Equation (16) is a result already obtained by Bengtson⁽¹³⁾ and other authors.⁽¹⁴⁾ The bracketed term in Eq. (16) is the self-shielding factor given by Case, Placzek, and de Hoffmann⁽¹⁵⁾ for the case of an absorbing foil in an isotropic neutron bath. The remaining factor is a correction for the effect of the anisotropy of the flux at the surface of the foil.

Equation (16) gives a reliable estimate of the self-shielding effect as long as the scale of the heterogeneity is small compared with a diffusion length in the poisoned system, and as long as the ratio $\phi_-(E)/\phi(E)$ is not very different from unity. This ratio is sensitive to the foil separation d only when d is considerably longer than a mean free path.

3.2. Universal Scattering-law Code (GASKET)

The desirability of having a universal scattering-law code has been discussed in previous reports. A new formulation for the logic of this code has been obtained and is presented below. Approximately 50% of the programming (FORTRAN statements) has been completed. The program which computes the scattering law from frequency spectra is named GASKET, while the program which computes scattering kernels from the GASKET results is named FLANGE.

3.2.1. Statement of the Problem

The GASKET program computes the thermal neutron scattering law

$$S\left(\frac{p^2}{2}, \epsilon\right) = e^{\epsilon/2T} \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\epsilon t} X\left(\frac{p^2}{2}, t\right) dt \quad (17a)$$

$$= \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\epsilon t} X\left(\frac{p^2}{2}, t + i/2T\right) dt \quad (17b)$$

$$= \frac{1}{\pi} \int_0^{\infty} X\left(\frac{p^2}{2}, t + i/2T\right) \cos \epsilon t \, dt, \quad (17c)$$

where the third form (17c) is valid because of property (22) below. $S[(p^2/2), \epsilon]$ is therefore a real even function of ϵ , and is related to the differential cross section by the well-known expression⁽¹³⁾

$$\frac{1}{\sigma_0} \sigma_s(E' \rightarrow E, \underline{\Omega}' \rightarrow \underline{\Omega}) = \left(\frac{E'}{E}\right)^{1/2} \left(\frac{M+1}{M}\right)^2 \frac{1}{4\pi} e^{-\epsilon/2T} S\left(\frac{p^2}{2}, \epsilon\right), \quad (18)$$

where

$$\epsilon = E - E' \quad (19)$$

is the energy transferred to the neutron, and

$$\frac{p^2}{2} = E + E' - 2\sqrt{EE'}\mu \quad (20)$$

is a measure of the momentum p transferred to the neutron; μ is the cosine of the scattering angle; E' and E are the initial and final neutron energies, respectively; M is the ratio of the mass of the scattering particle to the mass of the neutron; T is the effective temperature; and σ_0 is the free-atom scattering cross section.

The function $X(p^2/2, t)$ satisfies the following "moment conditions" (listed originally by Placzek⁽¹⁴⁾):

$$X\left(\frac{p^2}{2}, t\right) \text{ is analytic in } t \text{ near } t = 0, \quad (21)$$

$$X\left(\frac{p^2}{2}, t + i/2T\right) \text{ is real for real } \frac{p^2}{2}, \text{ and real } t$$

(detailed balance condition), (22)

$$\left. \frac{\partial}{\partial t} X\left(\frac{p^2}{2}, t\right) \right|_{t=0} = \frac{ip^2}{2M}, \quad (23)$$

$$\frac{\partial^2}{\partial t^2} X \left(\frac{p^2}{2}, t \right) \Big|_{t=0} = -\bar{T} \frac{p^2}{M} - \left(\frac{p^2}{2M} \right)^2 \quad (24)$$

($\bar{T} = 2/3$ of the mean energy of the scattering particles),

$$X \left(\frac{p^2}{2}, 0 \right) = 1. \quad (25)$$

The fundamental simplifying assumptions to be made are (1) that X has the Gaussian form⁽¹⁵⁾

$$X \left(\frac{p^2}{2}, t \right) = \exp \left[\frac{p^2}{2} G(t) \right] \quad (26)$$

or a direct generalization of this form (see Eq. (52)), and (2) that the motions of the scatterer which contribute to the self-scattering may be represented in terms of N independent mechanical modes:

$$G(t) = \sum_{n=1}^N G_n(t) \quad (27)$$

It follows that

$$X \left(\frac{p^2}{2}, t \right) = \prod_{n=1}^N X_n \left(\frac{p^2}{2}, t \right) = \prod_{n=1}^N \exp \left[\frac{p^2}{2} G_n(t) \right] \quad (28)$$

The functions $G_n(t)$ must, from Eq. (25), vanish for $t = 0$. They may therefore be expressed in the form

$$G_n(t) = \frac{w_n}{M} [\gamma_n(t) - \gamma_n(0)] \quad (29)$$

or

$$G_n(t + i/2T_n) = \frac{w_n}{M} [\gamma_n(t + i/2T_n) - \gamma_n(0)] \quad (30)$$

Form (30) involves real functions only and is therefore easier to work with. T_n is the temperature appropriate to the n^{th} mode; it will normally be the same for all modes but some provision for exceptional cases is desirable. The quantities w_n are relative weights for the N modes, such that

$$0 \leq w_n \leq 1 \quad \text{and} \quad \sum_{n=1}^N w_n = 1 \quad (31)$$

Provision is made in the current GASKET program for the following modes:

1. Translational (free gas).
2. Brownian (diffusive) motion.
3. Vibrational (isotropic, with distributed phonon frequency spectrum).
4. Vibrational (anisotropic, with orientation parallel to primary lattice plane, with distributed spectrum).
5. Vibrational (anisotropic, with orientation transverse to primary lattice plane, distributed spectrum).
6. Vibrational (isotropic, with discrete frequency spectrum).

Other modes may be added to this list.

3.2.2. Translational Mode

The free-gas scattering law is obtained from

$$G_1(t) = \frac{w_1}{M} (it - Tt^2) \quad (32)$$

or

$$G_1(t + i/2T) = - \frac{w_1}{M} \left(\frac{1}{4T} + Tt^2 \right) \quad (33)$$

The resulting contribution to the scattering law is easily evaluated as

$$S_1\left(\frac{p^2}{2}, \epsilon\right) = (2\pi p^2 w_1 T_1 / M)^{-1/2} \exp \left\{ - \frac{M}{2p^2 T_1 w_1} \left[\epsilon^2 + \left(\frac{p^2 w_1}{2M} \right)^2 \right] \right\} \quad (34)$$

However, if other modes are present ($w_1 < 1$), form (34) is not convenient. The reason is that form (34) must be combined by convolution with corresponding expressions for the other modes (see Eq. (49)), while form (33) may be combined with corresponding expressions by the simpler process of addition.

3.2.3. Brownian Motion

The following expression is obtained by R. J. Glauber⁽¹⁶⁾ from the classical Langevin theory of Brownian motion:

$$\frac{1}{w_2} G_2(t) = - \frac{2T_2}{M\eta} (\eta|t| - 1 + e^{-\eta|t|}) , \quad (35)$$

where the viscosity η is in turn defined by

$$\eta = \frac{T_2}{DM} ,$$

with D being a dimensionless diffusion coefficient. This expression, being strictly classical, is inconsistent with condition (23), which is a quantum-theory result. However, if $|t|$ is taken as the modulus of the complex variable t , the result is

$$G_2(t + i/2T) = - \frac{2T_2 w_2}{M\eta} [\eta \sqrt{t^2 + (2T)^{-2}} - 1 + \exp [-\eta \sqrt{t^2 + (2T)^{-2}}]] , \quad (36)$$

which is consistent with (23). To show this, it is sufficient to replace t with $t - i/2T$ in (36) and expand the exponentials,

$$\begin{aligned} \frac{1}{w_2} G_2(t) &= - \frac{2T_2}{M\eta} [\eta \sqrt{t^2 - it/T_2} - 1 + \exp (-\eta \sqrt{t^2 - it/T_2})] \quad (35a) \\ &\cong - \frac{2T_2}{M\eta} \left[\frac{1}{2} \eta^2 \left(t^2 - \frac{it}{T_2} \right) + O(t^{3/2}) \right] , \end{aligned}$$

so that $(1/w_2) G_2'(0) = i/M$ as required by (23).

3.2.4. Vibrational Modes

The vibrational modes are specified in terms of phonon frequency spectra $f(\omega)$ as follows:

$$\gamma(t) = \int_{-\infty}^{\infty} \frac{f(|\omega|) \exp(\omega/2T)}{2\omega \sinh(\omega/2T)} e^{i\omega t} d\omega$$

$$= \int_0^{\infty} \frac{f(\omega)}{\omega} \left(\coth \frac{\omega}{2T} \cos \omega t + i \sin \omega t \right) d\omega, \quad (37)$$

$$\gamma(0) = \int_0^{\infty} \frac{f(\omega)}{\omega} \coth \frac{\omega}{2T} d\omega, \quad (38)$$

$$\begin{aligned} \gamma(t + i/2T) &= \int_{-\infty}^0 \frac{f(|\omega|)}{2\omega \sinh(\omega/2T)} e^{i\omega t} d\omega \\ &= \int_0^{\infty} \frac{f(\omega)}{\omega} \operatorname{csch} \frac{\omega}{2T} \cos \omega t d\omega. \end{aligned} \quad (39)$$

It can be shown using (26), (29), and (31) that (37) is consistent with (23) provided that

$$\int_0^{\infty} f(\omega) d\omega = 1. \quad (40)$$

Furthermore, $f(\omega)$ must vanish at the origin at least as strongly as ω^2 in order that the integrals (37) through (39) should exist. This restriction is not an essential one, however, since in the expression (30) for $G_n(t + i/2T)$, singularities associated with zero frequency will cancel.

The upper limit of the integrals (37) through (40) is in reality a cutoff frequency $\omega_{\max}$ rather than infinity. The physical significance of this cutoff is, of course, that a medium composed of discrete molecules cannot support vibrational modes with wavelengths much smaller than the mean spacing of the molecules. In a dilute gas this spacing approaches infinity, the cutoff frequency tends to zero, and the spectrum approaches

$$f(\omega) = \delta(\omega), \quad (41)$$

a delta-function at zero frequency. The gas-law formulae (32) and (33) are easily derived from (30), (37) through (39), and (41). This result offers one approach to any difficulties associated with zero frequency as described in the preceding paragraph; however, still simpler methods are also available for that problem.

The quantity $G_1(t)$ for the translational mode behaves as $-t^2$ for large t , while $G_2(t)$ for the Brownian motion behaves as $-t$. The corresponding X functions therefore vanish strongly for large t . In contrast, the integral (39) will at best behave as t^{-1} , so that $G_n(t)$ and $X_n(t)$ approach non-zero constants. Specifically, in this case,

$$\lim_{t \rightarrow \infty} X_n(t + i/2T) = \exp \left[- \frac{p^2 w_n}{2M} \gamma_n(0) \right] \quad (42)$$

is the contribution to the Debye-Waller factor. If the total X function for the problem contains no contributions from translational or diffusive motion modes, the Fourier transform (17c) will diverge. A similar situation arises if $X(t)$ is oscillatory as $t \rightarrow \infty$. The program must therefore separate the divergent contributions to (17c) from the other terms, and represent these by the appropriate delta functions.

The oscillatory behavior of $X(t)$ arises for mode 6, the discrete vibrational spectrum, for which the spectrum is specified in the form

$$f_6(\omega) = \sum_k a_k \delta(\omega - \omega_k) , \quad (43)$$

where

$$\sum_k a_k = 1 , \quad (44)$$

in order to conform to (40). One obtains at once

$$X_6 \left(\frac{p^2}{2}, t + i/2T_6 \right) = \prod_k \exp \left[- \frac{p^2 w_6 a_k}{2M\omega_k} \coth \frac{\omega_k}{2T_6} \right] \exp \left[\frac{p^2 w_6 a_k}{2M\omega_k} \operatorname{csch} \frac{\omega_k}{2T_6} \cos \omega_k t \right] . \quad (45)$$

Since

$$e^{z \cos \theta} = \sum_{n=-\infty}^{\infty} I_n(z) e^{-in\theta} = I_0(z) + 2 \sum_{n=1}^{\infty} I_n(z) \cos n\theta , \quad (46)$$

each term in (43) generates a phonon series.

$$X_{6,k} \left(\frac{p^2}{2}, t + i/2T_6 \right) = \exp \left[- \frac{p^2 w_6 a_k}{2M\omega_k} \coth \frac{\omega_k}{2T_6} \right] \cdot \left[I_0 \left(\frac{p^2 w_6 a_k}{2M\omega_k} \operatorname{csch} \frac{\omega_k}{2T_6} \right) + 2 \sum_{n=1}^{\infty} I_n \left(\frac{p^2 w_6 a_k}{2M\omega_k} \operatorname{csch} \frac{\omega_k}{2T_6} \right) \cos (n\omega_k t) \right] . \quad (47)$$

The Fourier transform (17b) or (17c) of this quantity is

$$S_{6,k} \left(\frac{p^2}{2}, \epsilon \right) = \exp \left[- \frac{p^2 w_6 a_k}{2M\omega_k} \coth \frac{\omega_k}{2T_6} \right] \cdot \left\{ I_0 \left(\frac{p^2 w_6 a_k}{2M\omega_k} \operatorname{csch} \frac{\omega_k}{2T_6} \right) \delta(\epsilon) + \sum_{n=1}^{\infty} I_n \left(\frac{p^2 w_6 a_k}{2M\omega_k} \operatorname{csch} \frac{\omega_k}{2T_6} \right) [\delta(\epsilon - n\omega_k) + \delta(\epsilon + n\omega_k)] \right\}. \quad (48)$$

It is appropriate to account for such contributions after the S-function for other modes, if any, has been computed. Let $S^{(K-1)}(p^2/2, \epsilon)$ be the scattering law computed for the other modes; or if there are none, $\delta(\epsilon)$ may be used. The incorporation of (48) is accomplished by means of Fourier convolution:

$$S^{(K)} \left(\frac{p^2}{2}, \epsilon \right) = \int_{-\infty}^{\infty} S^{(K-1)} \left(\frac{p^2}{2}, \epsilon - \epsilon' \right) S_K \left(\frac{p^2}{2}, \epsilon' \right) d\epsilon'. \quad (49)$$

For S_K of form (48), this convolution involves no numerical integration. The result of incorporating the first term in (43) is therefore

$$S^{(K)} \left(\frac{p^2}{2}, \epsilon \right) = \left[\exp \left(- \frac{p^2 w_6 a_1}{2M\omega_1} \coth \frac{\omega_1}{2T_6} \right) \right] \sum_{n=-\infty}^{\infty} I_n \left(\frac{p^2 w_6 a_1}{2M\omega_1} \operatorname{csch} \frac{\omega_1}{2T_6} \right) S^{(K-1)} \left(\frac{p^2}{2}, \epsilon - n\omega_1 \right). \quad (50)$$

If $S^{(K-1)}$ is continuous in ϵ , the evaluation of (50) only requires interpolation. If $S^{(K-1)}$ is, on the other hand, itself a delta-function distribution, then each term $\delta(\epsilon - \epsilon_k)$ will generate a series of the form

$$\sum_n A_n \delta(\epsilon - \epsilon_k - n\omega_1), \quad (51)$$

with coefficients defined by (50).

This process is then repeated for the remaining terms in (43).

Anisotropic vibrational modes (4) and (5) require special treatment also. Given the frequency spectra f_4 (or $f_{||}$) and f_5 (or $f_{\perp}$), the quantities $G_4(t + i/2T)$ and $G_5(t + i/2T)$ are formed in the usual way using (38), (39), and (30). The composite X-function is then defined as⁽¹⁷⁻¹⁹⁾

$$\begin{aligned}
X_{4,5} \left(\frac{p^2}{2}, t + i/2T \right) &= \int_0^1 d\ell \exp \left[\frac{p^2}{2} \ell^2 G_5(t + i/2T) \right. \\
&\quad \left. + \frac{p^2}{2} (1 - \ell^2) G_4(t + i/2T) \right] \\
&= e^{(p^2/2) G_4(t + i/2T)} \int_0^1 d\ell \exp \left[- \frac{p^2}{2} (G_4 - G_5) \ell^2 \right] .
\end{aligned} \tag{52}$$

The integral in (52) yields functions which are well-known⁽²⁰⁾:

$$\begin{aligned}
\int_0^1 d\ell e^{-x\ell^2} &= \frac{1}{\sqrt{x}} \int_0^{\sqrt{x}} e^{-\mu^2} du = \frac{1}{\sqrt{x}} \operatorname{Erf}(\sqrt{x}), \quad x > 0 \\
&= \Phi \left(\frac{1}{2}, \frac{3}{2}, -x \right) = \frac{1}{\sqrt{-x}} \operatorname{Erfi}(\sqrt{-x}), \quad x < 0,
\end{aligned} \tag{53}$$

where Φ denotes a confluent hypergeometric function. The discrete vibrational modes may also be anisotropic. These may be provisionally treated as distributed spectra, with the delta functions replaced by high, narrow peaks, and with the low-frequency portion of the spectrum ($0 \leq \omega \leq \omega_1$) replaced by a translational mode in order to avoid numerical difficulties at large t values.

3.2.5. Problem Input

The six mode designations are listed following Eq. (31). The input contains 21 records, or blocks, as follows:

Block 1

NTP	Number of previously used values of t (0 if none)
NT	Number of new values of t required (0 if none)
NP	Number of values of $p^2/2$ (non-zero)
NE	Number of values of ϵ (non-zero)

Block 2

RWT (I), $I = 1 \dots 6$	Relative weight of mode I, unnormalized (ignored for mode 5)

Block 3

TMP (I), $I = 1 \dots 6$	Temperature for mode I, degrees K.

<u>Block 4</u>	
JS(I), I = 1... 4	Number of frequencies and spectrum values, modes 3-6
<u>Blocks 5, 7, 9, 11</u>	(present for each mode with JS $\neq$ 0)
X(J), J = JS(I), I = 1... 4	Frequencies in 10^{-12} sec $^{-1}$
<u>Blocks 6, 8, 10, 12</u>	(present for each mode with JS $\neq$ 0)
Q(J), J = JS(I), I = 1... 4	Spectrum values, unnormalized
<u>Block 13</u>	(present for NT $\neq$ 0)
T(I), I = 1... NT	New values of t, ev $^{-1}$
<u>Block 14</u>	(present for NTP $\neq$ 0)
TP(N), N = 1... NTP	Previously used values of t, ev $^{-1}$
<u>Block 15</u>	(present for RWT(3) $\neq$ 0)
GP3(N), N = 1... NTP	Previously computed values of G, mode 3
<u>Block 16</u>	(present for RWT(4) $\neq$ 0)
GP4(N), N = 1... NTP	Previously computed values of G, mode 4
<u>Block 17</u>	(present for RWT(4) $\neq$ 0)
GP5(N), N = 1... NTP	Previously computed values of G, mode 5
<u>Block 18</u>	
D	Dimensionless diffusion coefficient for mode 2
M	Mass of scattering nucleus, unified amu
<u>Block 19</u>	
PP(I), I = 1... NP	Values of $p^2/2$ required, ev
<u>Block 20</u>	
EE(I), I = 1... NE	Values of ϵ required, ev
<u>Block 21</u>	
TY(N), N = 1... 401	Table of confluent hypergeometric functions

3.2.6. Main Program

The main program reads the input, checks for input errors, and converts units as follows:

1. Temperatures in $^{\circ}\text{K}$ are converted to ev by dividing by 11,604. 9.
2. Frequencies in 10^{-12} sec $^{-1}$ are converted to ev by dividing by 1519. 30.

3. Mass in unified amu is converted to neutron mass units by dividing by 1.0086654.
4. Relative weights are normalized to unit sum (Eq. (31)).

The main program then calls the subroutines which compute the values of $G_n(t + i/2T)$ and $X(t + i/2T)$ as required, merge these with any previously computed values, and generate the scattering law for the required arguments. The printed and punched output data are then written.

3.2.7. Subroutines

Most of the formulae are evaluated by subroutines, all of which utilize parameters placed in a common storage block. The subroutines are described below.

INTG and CØINT. These are trapezoidal integration routines in which the independent variable is specified as an arbitrary nondecreasing sequence. INTG computes the integral

$$\text{INT} = \int_{x_1}^{x_{NS}} Q(x) dx, \quad NS \geq 2, \quad (54)$$

where $Q(x)$ is assumed to be piecewise linear within each of the $NS-1$ intervals. The contribution of the interval (x_{s-1}, x_s) is then

$$\frac{1}{2} (x_s - x_{s-1}) (Q_s + Q_{s-1}), \quad x_s \geq x_{s-1}.$$

CØINT computes the integrals

$$P(y_r) = \int_{x_1}^{x_{NS}} Q(x) \cos(xy_r) dx, \quad NS \geq 2, \quad x_s \geq x_{s-1}, \quad NR \geq 1, \quad y_r \neq 0, \\ r = 1, \dots, NR, \quad (55)$$

where $Q(x)$ is again assumed to be piecewise linear within each of the $NS-1$ intervals. The contribution of the interval (x_{s-1}, x_s) is

$$P_{sr} = \frac{Q_s}{y_r} \left[\left(1 - \frac{\sin \theta_{sr}}{\theta_{sr}} \right) \sin x_s y_r + \frac{1 - \cos \theta_{sr}}{\theta_{sr}} \cos x_s y_r \right]$$

$$+ \frac{Q_{s-1}}{y_r} \left[-1 - \left(\frac{\sin \theta_{sr}}{\theta_{sr}} \right) \sin x_{s-1} y_r + \frac{1 - \cos \theta_{sr}}{\theta_{sr}} \cos x_{s-1} y_r \right], \quad (56)$$

where $\theta_{sr} = (x_s - x_{s-1}) y_r$. $C\emptyset INT$ is used in the evaluation of formulae (17c) and (39).

GAS. This routine evaluates formula (33) for $G_1(t + i/2T)$.

BRØWN. This routine evaluates (36) for $G_2(t + i/2T)$.

GTF. This routine evaluates formula (30) for $G_n(t + i/2T)$, $n = 3, 4, \text{ or } 5$, using formulae (38) and (39), and normalizing input data with (40). The first step is the evaluation of the normalizing factor R , where

$$\frac{1}{R} = \int_0^{\omega_{NS}} f(\omega) d\omega = \frac{1}{3} f_1 \omega_1 + \frac{1}{2} \sum_{s=2}^{NS} (\omega_s - \omega_{s-1}) (f_s + f_{s+1}). \quad (57)$$

Here, $f(\omega)$ is assumed to be proportional to ω^2 in the interval $0 \leq \omega \leq \omega_1$. It is assumed that all of the ω_s , $s = 1 \dots NS$, and the t_r , $r = 2 \dots NR$ are non-zero. However, $\omega_0 = 0$ is understood and t_1 may be zero or non-zero. GTF computes, in turn, the quantities

$$\beta = \frac{1}{2T}, \quad (58)$$

$$g_0 = \frac{f_1}{\beta \omega_1^2}, \quad (59)$$

$$g_s = \frac{f_s}{\omega_s} \operatorname{csch}(\beta \omega_s), \quad s = 1 \dots NS, \quad (60)$$

$$\gamma_n(t_r + i\beta) = \int_0^{\omega_{NS}} g(\omega) \cos(\omega t_r) d\omega, \quad r = 1 \dots NR, \quad (61)$$

$$h_s = g_s \cosh(\beta \omega_s), \quad s = 0 \dots NS, \quad (62)$$

$$\gamma_n(0) = \int_0^{\omega_{NS}} h(\omega) d\omega, \quad (63)$$

$$G(t_r + i\beta) = \frac{w_n}{M} [\gamma_n(t_r + i\beta) - \gamma_n(0)] , \quad r = 1 \dots NR . \quad (64)$$

XTG. This routine evaluates the function $X(p^2/2, t_r + i\beta)$ according to formula (26), using a G-function containing contributions from modes 1, 2, and 3 at most.

XTGA. This routine evaluates formula (52) for the X-function containing the contributions of modes 4 and 5.

EERF. This routine evaluates the confluent hypergeometric function (formula (53)) used in the XTGA routine.

The available formulae are:

$$\text{Eerf}(x) = \int_0^1 e^{-x\mu^2} d\mu = \sum_{n=0}^{\infty} \frac{(-x)^n}{n! (2n+1)} \quad (65)$$

$$= e^{-x} \sum_{n=0}^{\infty} x^n / \left(\frac{3}{2}\right)_n , \quad (66)$$

where

$$\left(\frac{3}{2}\right)_n = \frac{3}{2} \cdot \frac{5}{2} \dots \left(n + \frac{1}{2}\right) = \Gamma\left(\frac{3}{2} + n\right) / \Gamma\left(\frac{3}{2}\right) ,$$

$$\begin{aligned} \text{Eerf}(x) = \frac{1}{2} \left(\frac{\pi}{x}\right)^{1/2} & - \frac{\frac{1}{2} e^{-x}}{x+1} \\ & \frac{2+2}{x+3} \\ & \frac{2+4}{x+5} \\ & \frac{2+6}{x+7} \dots \quad (x > 0) \end{aligned} \quad (67)$$

$$\cong \frac{1}{2} \left(\frac{\pi}{x}\right)^{1/2} - \frac{1}{2x} e^{-x} \left\{ \sum_{m=0}^{M-1} \left(\frac{1}{2}\right)_m (-x)^{-m} + O(|x|^{-M}) \right\} . \quad (68)$$

The power series (65) and (66) are useful for $|x| \ll 1$, and the continued fractions expansion (67) for $x \gg 1$.⁽²¹⁾ The asymptotic series (68) may be used for $|x| \gg 1$ only; for negative x the first term is ignored. For $x \sim 1$, a Hastings approximation may be used:⁽²²⁾

$$\text{Eerf}(x) \cong \frac{1}{2} \left(\frac{\pi}{x} \right)^{1/2} - (a_1 \eta + a_2 \eta^2 + a_3 \eta^3 + a_4 \eta^4 + a_5 \eta^5) \frac{e^{-x}}{\sqrt{x}}, \quad x > 0, \quad (69)$$

$$\eta = \frac{1}{1 + p\sqrt{x}},$$

$$p = 0.3275911,$$

$$a_1 = 0.225836846,$$

$$a_2 = -0.252128668,$$

$$a_3 = 1.259695130,$$

$$a_4 = -1.287822453,$$

$$a_5 = 0.940646070.$$

For negative x there appears to be no convenient recipe for the region $x \sim -1$. A related function has, however, been tabulated, (23)

$$I(x) = \int_0^x e^{(t^2 - x^2)} dt,$$

so that for $x < 0$

$$\text{Eerf}(x) = \frac{1}{\sqrt{-x}} e^{-x} I(\sqrt{-x}) = \frac{1}{|x|^{1/2}} e^{|x|} I(\sqrt{|x|}). \quad (70)$$

(Since the utility of these alternative methods has not been fully evaluated, provision is made in the initial version of the program for all of them.)

MERGE. This routine merges two lists, both of which are in non-decreasing order. It is used to combine previously calculated and new values of $G(t + i\beta)$.

INX. This routine computes the Bessel functions $I_n(x)$ from the backward recurrence relation

$$I_n(x) = \frac{2(n+1)}{x} I_{n+1}(x) + I_{n+2}(x).$$

3.2.8. FLANGE

This program utilizes the scattering law data generated by the GASKET code, and generates from these data both point kernels and group kernels.

Point kernels are calculated from the formula

$$\frac{\sigma_s^\ell(E' \rightarrow E)}{\sigma_o} = \frac{4\pi[\ell + (1/2)]}{\sigma_o} \int_{-1}^1 \sigma_s(E' \rightarrow E, \underline{\Omega}' \rightarrow \underline{\Omega}) P_\ell(\mu) d\mu, \quad (71)$$

where $\mu = \underline{\Omega}' \cdot \underline{\Omega}$ and

$$\frac{1}{\sigma_o} \sigma_s(E' \rightarrow E, \underline{\Omega}' \rightarrow \underline{\Omega}) = \left(\frac{E}{E'}\right)^{1/2} \left(\frac{M+1}{M}\right)^2 \frac{1}{4\pi} e^{-\epsilon/2T} S\left(\frac{p^2}{2}, \epsilon\right). \quad (72)$$

Legendre moments of orders $\ell = 0, 1, 2, 3$ may be generated. For the given values of E and E' , and the table of values of $S(p^2/2, E)$ provided by GASKET, linear interpolation in ϵ is used to define values of S corresponding to $\epsilon = E' - E$. The scattering angles corresponding to E, E' and $p^2/2$ are then computed from the relation

$$\mu = \frac{E + E' - p^2/2}{2\sqrt{EE'}}. \quad (73)$$

These angles are not the ones needed for the integration over μ in (71); these latter angles are specified in advance, and the appropriate values of S are obtained by quadratic interpolation.

Group kernels are calculated from the formula

$$G_{g' \rightarrow g}^\ell = \frac{\left(\ell + \frac{1}{2}\right) \left(\frac{M+1}{M}\right)^2}{\int_{E_{g'}}^{E_{g'+1}} \phi(E') dE'} \int_{E_{g'}}^{E_{g'+1}} dE' \frac{\phi(E')}{\sqrt{E'}} e^{E'/2T} \int_{E_g}^{E_{g+1}} dE \sqrt{E} e^{-E/2T} \int_{-1}^1 d\mu P_\ell(\mu) S\left(\frac{p^2}{2}, \epsilon\right), \quad (74)$$

where $\phi(E)$ is taken to be identically 1 unless better information is available. Delta functions included in the specification of $S(p^2/2, \epsilon)$ will in most cases be given special treatment, since they make possible the evaluation of (74) as a single quadrature.

The entire group kernel will be written on tape (or punched) in a form suitable for inclusion in the GGC-II data library.

3.3. Coherent Effects in Beryllium

As was reported in the annual report⁽²⁾ for 1963, a scattering kernel has been calculated for beryllium using the machine program SUMMIT. This kernel makes use of the so-called incoherent approximation, i. e., the interference of neutron waves scattering from different nuclei is neglected. As was shown in Ref. 25, this approximation leads to curves of the scattering law $S(\alpha, \beta)$ which are smooth and agree reasonably well with the measurements of Sinclair.⁽⁵⁾ Recent very accurate data obtained at Phillips⁽²⁴⁾ show bumps in the curves for $S(\alpha, \beta)$ versus α for fixed β values. These bumps would seem to be due to inelastic coherent scattering, which up to now has not been included in the theory for any solid moderator. Although the cross section for coherent (interference) scattering is in general very complicated and difficult to work with, the experiments might well be explained by a consideration of only one- or two-phonon terms in a multiphonon expansion of the coherent scattering cross section. This argument is fortified by the fact that experiments indicate that the incoherent approximation is quite adequate at large energy and momentum transfers.

The cross section for one-phonon coherent scattering is given by

$$\frac{d^2\sigma_{\text{coh}}}{d\Omega dE} = \frac{\sigma_{\text{coh}}}{4\pi} \sqrt{\frac{kE}{EO}} \frac{(2\pi)^3}{V} e^{-W} \sum_{\mathbf{s}\epsilon} \frac{|\vec{K} - \vec{C}_s|^2}{2NM\omega_s} \frac{\coth\left(\frac{\omega_s}{T}\right) + \epsilon\delta}{2} \sum_{\vec{\tau}} \delta(\vec{K} - \vec{\tau} + \vec{q}_s) \delta(E - E_0 + \epsilon\omega_s), \quad (76)$$

where σ_{coh} is the bound-atom cross section; E_0 and E are the initial and final neutron energies; V is the volume of a unit cell; $\vec{K}$ is the momentum transfer; M is the atomic mass; ω_s is the phonon energy; T is the temperature in energy units; $\vec{\tau}$ is a reciprocal lattice vector; N is the number of particles in a unit cell; $\vec{q}_s$ is the wave-vector of the phonon; and $\vec{C}_s$ is the phonon amplitude vector. ϵ is +1 for phonon emission and -1 for phonon absorption. The factor W is the Debye-Waller factor and is given by

$$W = \frac{1}{2M} \int_0^{\omega_{\text{max}}} d\omega \frac{f(\omega)}{\omega} \coth \frac{\omega}{2T},$$

where $f(\omega)$ is the lattice vibrational spectrum. The amplitudes $\vec{C}_s$ can be found by using the machine program used in Ref. 2 to calculate the frequencies ω_s necessary to generate $f(\omega)$. It is hoped that the inclusion of this one-phonon coherent scattering will make the theory agree with the experimental curves of Schmunk.

3. 4. Neutron Scattering by Polyethylene

3. 4. 1. Frequency Spectrum

Crystalline polyethylene is known to be formed by very long, kinked chains of CH_2 radicals, only weakly interacting with each other. (26)(27) Therefore, its frequency spectrum is radically different from the one corresponding to a three-dimensional crystal. In fact, analyzing its normal modes of vibration in terms of plane waves $e^{ik \cdot r}$, the corresponding frequencies can only depend on the projection of the wave vector k in a direction $\vec{e}$ parallel to the chains (at least to the extent that interaction between neighboring chains can be neglected). This means that except for the scale of the k -axis, the dependence of the frequency W on the wave number k is the same for any direction of k ; since the only parameter involved is the phase difference θ between the vibration of corresponding elements of two neighboring radicals of the same chain. This dependence, as calculated by Lin and Koenig, (26) is shown in Fig. 20, where the nine different branches (corresponding to the three atoms per radical) can be assigned to the following vibrational modes:

T	c-c-c-c	torsion
ϕ	c-c-c	bending
R	c-c	stretching
γ	CH_2	rocking
ζ	CH_2	twisting
μ	CH_2	wagging
ψ	H-C-H	bending
u	C-H	symmetric stretching
v	C-H	antisymmetric stretching

Only two of these branches go to $\omega = 0$ for $\theta = 0$ and can thus be considered as acoustical modes. The remaining ones are optical branches. In the modes T, ϕ , and R, one can assume that the CH_2 radicals move essentially as a whole unit, whereas in the other modes the C atoms are nearly at rest. This assumption leads to a relative weighting factor of 2/15 for the acoustical and 13/15 for the optical contribution to the effective frequency spectrum relevant to neutron scattering by the H atoms contained in polyethylene. This point was discussed in more detail in the previous annual report.

Fig. 20--Acoustical and optical branches for polyethylene according to Lin and Koeing.

The exact frequency spectrum derived from Fig. 20 would become singular at all those frequencies at which $dW/d\theta$ vanishes for some one of the nine branches. For our purposes, however, it will be sufficient to replace the exact spectrum by a coarse histogram in which the contribution of each branch is uniformly distributed over the range limited by its upper and lower frequency, as was done by Wunderlich⁽²⁸⁾ for specific heat calculations. The optical part of this equivalent spectrum is shown in Fig. 21.

3. 4. 2. Scattering Kernel for Polyethylene

In the previous quarterly report, an attempt was made to derive a scattering kernel for polyethylene by lumping the optical part of the frequency spectrum just described into four discrete δ -shaped lines, treating the acoustic part in the short collision approximation with $T_{\text{eff}} = T$. The physical picture corresponding to this model is that of a single CH_2 radical undergoing hindered rotations and free translations.

As was shown, calculated total cross section and infinite medium spectra were in reasonable agreement with experiment. However, for very strong $1/v$ poisoning, a bump in the calculated spectrum would build up around 0.15 ev, not corresponding to anything found experimentally. In order to avoid this effect, different lumpings of the frequency spectrum were tried out and the corresponding total cross section and spectra were then compared. From all these calculations it was concluded that the different lumping by itself had practically no effect on the results. But because of the short collision approximation for the lower frequency necessarily made by the code GAKER, the lumping in only three discrete lines, as seen in Fig. 21, gave better spectra than those for four discrete oscillators. Even with three oscillators it is necessary to switch to the short collision treatment of the lowest frequency at an energy E_s of about 0.28 ev, slightly below the highest vibrational frequency of 0.36 ev. This switching energy is only twice the lowest level and thus is not large enough to ensure a smooth transition between the two regions. In fact, the total cross section obtained in this case shows a large discontinuity at $E = E_s$. But it is possible to get around this difficulty by using the energy transfer switching criteria option lately introduced into GAKER instead of switching at a fixed initial energy E_s . This will certainly smooth out the sharp discontinuity of E_s and will also be much closer to the exact result corresponding to the assumption of discrete oscillators. The reason this can be done in the case of polyethylene as opposed to the case of water, where it was avoided, is that for polyethylene the lowest lumped frequency is about 0.14 ev as compared with 0.06 ev for water. This very low energy level leads to a strong oscillatory behavior of the scattering kernel at high initial energy and small energy exchange, with sharp peaks at a distance of the order of 0.06 ev from each other, thus requiring a very fine energy mesh

Fig. 21--Scattering cross section for polyethylene.

for accurate spectra and cross-section calculations. It is clear that in the case of polyethylene, the use of the better switching criteria will not cause any difficulty at all, at least not for energies up to 1 or 2 ev. The preceding considerations are illustrated in Figs. 22 and 23, which show the total scattering cross section and the 2% borated polyethylene infinite medium spectra obtained with the described different lumping of the polyethylene frequency spectrum.

3.5. Anharmonic Oscillator

In the present section, we discuss the quantum dynamics of an anharmonic oscillator and the associated problem of neutron scattering by such an oscillator. The dynamical problem is treated in Section 3.5.1. Here, we consider only the problem of anharmonic oscillations in one dimension, although the generalization to more than one dimension can be made. For the problem of neutron scattering by an anharmonically bound particle, we make the unnecessary but simplifying assumption that the Hamiltonian describing the particle motion in three dimensions can be written as a sum of Hamiltonians describing independent, one-dimensional, anharmonic oscillations in three mutually perpendicular directions.

3.5.1. Dynamics of a One-dimensional Anharmonic Oscillator

We consider the one-dimensional motion of a particle of mass M , whose Hamiltonian can be expanded in the form

$$H = \frac{1}{2} \left(\frac{p_x^2}{m} + m \omega^2 x^2 \right) + \lambda H_3(x) + \lambda^2 H_4(x) + \dots, \quad (77)$$

where λ is a smallness parameter, ω is the frequency that would pertain for purely harmonic oscillations, x is the displacement of the particle from its equilibrium position, p_x is the momentum conjugate to x , and the

$$H_n(x) = A_n x^n \quad (n \geq 3) \quad (78)$$

are the anharmonic contributions to the potential energy of the particle. The displacement x and its conjugate momentum p_x satisfy the commutation relation

$$[x, p_x] = xp_x - p_x x = i\hbar \quad (79)$$

and the equations of motion

$$\left. \begin{aligned} \dot{p}_x &= \frac{i}{\hbar} [H, p_x] \\ \dot{x} &= \frac{i}{\hbar} [H, x] \end{aligned} \right\}, \quad (80)$$

Fig. 22--Frequency distribution for polyethylene.

Fig. 23--Infinite medium neutron spectrum in 2% borated polyethylene.

where $\hbar$ is Planck's constant.

Before trying to construct solutions to the equations of motion, it is convenient to introduce a new coordinate q and conjugate momentum p ,

$$\left. \begin{aligned} q &= \left(\frac{m\omega}{2\hbar}\right)^{1/2} x - i \left(\frac{1}{2m\hbar\omega}\right)^{1/2} p_x \\ p &= \left(\frac{m\omega}{2\hbar}\right)^{1/2} x + i \left(\frac{1}{2m\hbar\omega}\right)^{1/2} p_x \end{aligned} \right\}, \quad (81)$$

so that

$$x = \left(\frac{\hbar}{2m\omega}\right)^{1/2} (q + p), \quad (82)$$

$$ip_x = \left(\frac{m\hbar\omega}{2}\right)^{1/2} (p - q) \quad (83)$$

with

$$[q, p] = -1, \quad (84)$$

$$H = H(q, p) = H_2(p, q) + \lambda H_3(p, q) + \lambda^2 H_4(p, q) + \dots, \quad (85)$$

where

$$H_n(p, q) = a_n \left(\frac{\hbar}{2m\omega}\right)^{n/2} (q + p)^n, \quad n \geq 3 \quad (86)$$

and

$$H_2(p, q) = \frac{\hbar\omega}{2} (qp + pq) = \hbar\omega \left(pq + \frac{1}{2}\right). \quad (87)$$

We note the identity between p, q and a, a^+ , the harmonic oscillator annihilation and creation operators, respectively:

$$\left. \begin{aligned} p &= a^+ \\ q &= a \end{aligned} \right\}. \quad (88)$$

We now turn our attention to the problem of solving the equations of motion (80). We want to show that by means of a sequence of contact transformations, it is possible to transform from p, q to new conjugate coordinates $\bar{p}, \bar{q}$ and from the Hamiltonian H to a new Hamiltonian $\bar{H} = \bar{H}(\bar{p}, \bar{q})$ in such a way that

$$[\bar{q}, \bar{p}] = -1, \quad (89)$$

$$\left. \begin{aligned} \dot{\bar{q}} &= \frac{i}{\hbar} [\bar{H}, \bar{q}] \\ \dot{\bar{p}} &= \frac{i}{\hbar} [\bar{H}, \bar{p}] \end{aligned} \right\}, \quad (90)$$

with $\bar{H}(\bar{p}, \bar{q})$ of the form

$$\bar{H}(\bar{p}, \bar{q}) = \bar{H}(\bar{p}\bar{q} + \bar{q}\bar{p}) \quad (91)$$

or, more explicitly, with $\bar{H}(\bar{p}, \bar{q})$ a series in powers of $\bar{p}\bar{q} + \bar{q}\bar{p}$. The useful consequence of this is that the quantity $\bar{p}\bar{q} + \bar{q}\bar{p}$ is a constant of the motion from which one can determine explicit periodic solutions for $\bar{p}, \bar{q}$, which in turn lead to series solutions for the original coordinates p, q . We also note that form (91) for $\bar{H}(\bar{p}, \bar{q})$ is attained only by means of a particular contact transformation. Equation (90), on the other hand, holds by definition for any contact transformation.

The contact transformation is a well-known concept both in classical and quantum mechanics. Here, we wish to briefly review the contact transformation in its quantum mechanical context, following the treatment given in an early edition of Dirac's book. (30)

Consider a unitary transformation from the state vector ψ to the state vector

$$|\bar{\psi}\rangle = U |\psi\rangle. \quad (92)$$

Since the transformation is unitary, U satisfies by definition $U U^\dagger = 1$, or

$$U^\dagger = U^{-1}, \quad (93)$$

where $U^\dagger$ is the adjoint of U . For any nonsingular U satisfying (93), the transformation (92) preserves the normalization of the state vectors; i. e.,

$$\langle \psi | \psi \rangle = \langle \bar{\psi} | \bar{\psi} \rangle.$$

Another important property of U is that if α is any observable, then the eigenvalues of

$$\bar{\alpha} = U \alpha U^{-1} \quad (94)$$

are the same as those of α . Further, if an algebraic relation holds between a set of operators α_j , the same relation holds between the transformed operators

$$\bar{\alpha}_j = U \alpha_j U^{-1}, \quad (95)$$

and if one operator is a function of another, the same functional relation holds between the transformed operators. One can also show that the result of two successive unitary transformations is unitary. The proof of these well-known properties of unitary transformations is easily accomplished, and may be found in Ref. 30.

A useful and direct consequence of the properties considered above is the preservation of the commutation relation between pairs of conjugate observables. If

$$\alpha_i \alpha_j - \alpha_j \alpha_i = \delta_{ij} , \quad (96)$$

then

$$\overline{\alpha}_i \overline{\alpha}_j - \overline{\alpha}_j \overline{\alpha}_i = \delta_{ij} . \quad (97)$$

Finally, we demonstrate that the canonical form of the equations of motion is preserved. That is, if

$$\dot{\alpha} = \frac{i}{\hbar} [H(p, q), \alpha] , \quad (98)$$

where α is an observable not depending explicitly on time, then

$$\frac{d}{dt} \overline{\alpha} = \frac{i}{\hbar} [\overline{H}(\overline{p}, \overline{q}), \overline{\alpha}] , \quad (99)$$

where

$$\overline{H}(\overline{p}, \overline{q}) = U^{-1} H(p, q) U . \quad (100)$$

Here, $H(\overline{p}, \overline{q})$ is the same function of

$$\left. \begin{aligned} \overline{p} &= U p U^{-1} \\ \overline{q} &= U q U^{-1} \end{aligned} \right\} \quad (101)$$

that $H(p, q)$ is of p and q , respectively. The proof of Eq. (99) is simple and direct:

$$\begin{aligned} \frac{d}{dt} \overline{\alpha} &= \frac{i}{\hbar} [H(p, q), \overline{\alpha}] \\ &= \frac{i}{\hbar} [H(p, q) \overline{\alpha} - \overline{\alpha} H(p, q)] \\ &= \frac{i}{\hbar} [U^{-1} U H(p, q) U^{-1} U \overline{\alpha} - \overline{\alpha} U^{-1} U H(p, q) U^{-1} U] \end{aligned}$$

$$\begin{aligned}
&= \frac{i}{\hbar} [U^{-1} H(\bar{p}, \bar{q}) U \bar{\alpha} - \bar{\alpha} U^{-1} H(\bar{p}, \bar{q}) U] \\
&= \frac{i}{\hbar} [\bar{H}(\bar{p}, \bar{q}) \bar{\alpha} - \bar{\alpha} \bar{H}(\bar{p}, \bar{q})] \quad \text{Q. E. D.}
\end{aligned}$$

We now apply these results to the system whose motion is described by the Hamiltonian (q). The objective is to show that up to any given order in λ , (q) may be transformed into the form of (91) by means of a sequence of contact transformations. To prove that this can be done, we use the method of proof by induction. We assume that $H(p, q)$ has the form

$$\begin{aligned}
H(p, q) = & \frac{\hbar\omega}{2} J + \lambda^2 H_4(J) + \dots + \lambda^{N-2} H_N(J) + \lambda^{N-1} H_{N+1}(p, q) \\
& + \lambda^N H_{N+e}(p, q) + \dots, \quad (102)
\end{aligned}$$

where $J = pq + qp$, q and p being a conjugate coordinate and momentum which satisfy Eq. (84). The term $H_n(J)$ ($n \leq N$) is a polynomial of degree $n/2$ in J for even values of n , and vanishes identically for odd values of n .

The term $H_n(p, q)$ ($n \geq N+1$) consists of terms with m powers of p and k powers of q , such that $m+k \leq n$ (for example, the term $p^{m-1} q p q^{k-1}$). All terms which have the same m and the same k can be collected together and by means of the commutation relations expressed as a sum of terms, each of which is in normal form (i. e., with creation operators standing to the left of annihilation operators) and each of which are of degree n or less. If we let H_n^{mk} denote the collection of terms in H_n which simultaneously contain m powers of p and k powers of q , we can write

$$\begin{aligned}
H_n^{mk}(p, q) &= a_n^{(0)} p^m q^k + a_n^{(1)} p^{m-1} q^{k-1} + \dots + a_n^{(m)} q^{k-m} \quad k \geq m \\
&= a_n^{(0)} p^m q^k + a_n^{(1)} p^{m-1} q^{k-1} + \dots + a_n^{(k)} p^{m-k} \quad m \geq k.
\end{aligned} \quad (103)$$

We shall now show that all such terms with $m \neq k$ can be eliminated from H_{N+1} by means of a contact transformation

$$U_{N+1} = e^{\frac{i}{\hbar} \lambda^{N-1} S_{N+1}(\bar{p}, \bar{q})}, \quad (104)$$

where $S_{N+1}(\bar{p}, \bar{q})$, expressed as a sum of terms each in the normal form, contains no terms of order greater than $N + 1$.

Setting $S = \lambda^{N-1} S_{N+1}$, we have

$$\bar{H}(\bar{p}, \bar{q}) = e^{-iS/\hbar} H(\bar{p}, \bar{q}) e^{iS/\hbar} \quad (105)$$

$$\begin{aligned} &= \sum_{n=0}^{\infty} \frac{1}{n!} \left(\frac{i}{\hbar} \right)^n [H(\bar{p}, \bar{q}), S(\bar{p}, \bar{q})]_n \\ &= \sum_{n=0}^{\infty} \frac{1}{n!} \left(\frac{i}{\hbar} \right)^n \lambda^{(N-1)n} [H, S_{N+1}]_n, \end{aligned}$$

where $[H, S]_n$ is defined recursively by

$$\begin{aligned} [H, S]_0 &= 1, \\ [H, S]_1 &= [H, S], \\ [H, S]_2 &= \{[H, S]_1, S\} = \{[H, S], S\}, \\ [H, S]_n &= \{[H, S]_{n-1}, S\}. \end{aligned} \quad (106)$$

Collecting like powers of λ gives

$$\begin{aligned} \bar{H}(\bar{p}, \bar{q}) &= H_2(\bar{J}) + \lambda^2 H_y(\bar{J}) + \dots + \lambda^{N-2} H_N(\bar{J}) + \lambda^{N-1} H_{N+1}(\bar{p}, \bar{q}) \\ &\quad + \frac{i}{\hbar} \lambda^{N-1} [H_2, S_{N+1}] + \lambda^N H_{N+2}(\bar{p}, \bar{q}) + o(\lambda^{N+1}) \end{aligned} \quad (107)$$

Since S_{N+1} is at our disposal, the terms of order λ^{N-1} can be made to vanish provided that

$$\bar{H}_{N\hbar}(\bar{p}, \bar{q}) = \frac{i}{\hbar} [H_2, S_{N+1}] + H_{N+1} = 0, \quad (108)$$

which is equivalent to

$$\frac{i\omega}{2} [\bar{J}, S_{N+1}] + H_{N+1} = 0 \quad (109)$$

S_{N+1}^{mk} denote the collection of terms which contain p m times and q k times. By expressing S_{N+1}^{mk} as a sum of terms each of which is in normal form, we obtain

$$S_{N+1}^{mk}(p, q) = S_{N+1}^0 \bar{p}^{-m} \bar{q}^{-k} + S_{N+1}^1 \bar{p}^{-m-1} \bar{q}^{-k-1} + \dots + S_{N+1}^m \bar{p}^{-m-k} \quad m \geq k, \quad (110)$$

$$S_{N+1}^{mk}(\bar{p}, \bar{q}) = S_{N+1}^0 \bar{p}^{-m} \bar{q}^{-k} + S_{N+1}^1 \bar{p}^{-m-1} \bar{q}^{-k-1} + \dots + S_{N+1}^m \bar{q}^{-k-m} \quad m \geq k.$$

Now we can easily prove by induction that

$$[\bar{J}, \bar{p}^{-m} \bar{q}^{-k}] = -2(m-k) \bar{p}^{-m} \bar{q}^{-k} \quad (111)$$

Thus,

$$[\bar{J}, S_{N+1}^{mk}] = -2(m-k) S_{N+1}^{mk}, \quad (112)$$

and by choosing

$$i\omega(m-k) S_{N+1}^{mk} = +H_{N+1}^{(mk)}, \quad (113)$$

$\bar{H}_{N+1}^{mk}$ vanishes. This latter circumstance is not possible, however, for $m = k$, for then

$$\bar{H}_{N+1}^{mk} = H_{N+1}^{mk} \quad (m = k) \quad (114)$$

and $\bar{H}_{N+1}^{mk}$ vanishes only when H_{N+1}^{mk} vanishes. In summary, we can choose $S_{N+1}(\bar{p}, \bar{q})$ in such a way that $\bar{H}_{N+1}$ contains only those terms which contain $\bar{p}$ and $\bar{q}$ an equal number of times. Clearly, $\bar{H}_{N+1}$ vanishes identically if $N+1$ is odd. Finally, we note that the commutation relations can be used to express $\bar{p}^n \bar{q}^n$ as a power series of degree n in $\bar{J}$. If H_{N+1} is a polynomial of degree $N+1$ in $\bar{p}$ and $\bar{q}$, $\bar{H}_{N+1}$ is a polynomial of degree $(N+1/2)$ in $\bar{J}$, provided, of course, that $N+1$ is even.

One further point to note is that if $H_N(\bar{p}, \bar{q})$ is a polynomial of degree n in p and q , then $\bar{H}_N(\bar{p}, \bar{q})$ is also a polynomial of degree n in $\bar{p}$ and $\bar{q}$, assuming, of course, that $S_{N+1}(\bar{p}, \bar{q})$ is a polynomial of degree $N+1$. This follows on ordering the terms in $\bar{H}(\bar{p}, \bar{q})$ in powers of λ :

$$\left. \begin{aligned} \overline{H}(\overline{p}, \overline{q}) &= \sum_{k=v}^{\infty} \lambda^k \sum_{n=v}^{[k/N+1]} \frac{1}{n!} \left(\frac{\ell}{\hbar}\right)^n [H_{k+2-n(N-1)}, S_{N+1}]_n \\ \text{or} \\ \overline{H}_{k+2}(\overline{p}, \overline{q}) &= \sum_{n=v}^{[k/N-1]} \frac{1}{n!} \left(\frac{\ell}{\hbar}\right)^n [H_{k+2-n(N-1)}, S_{N+1}]_n \end{aligned} \right\} \quad (115)$$

where $[k/N - 1]$ is the integral part of $k/N - 1$. When expressed as a sum of terms each of which is in normal form, it is easy to see that the commutator of two polynomials is a polynomial whose degree is less by two than the sums of the degrees of the two members of the commutator. Using this fact and noting that $[\overline{J}, S_{N+1}^{mk}]_n$ involves n commutations of S_{N+1} , one finds that the degree of $[H_{k+2-n(N-1)}, S_{N+1}]_n$ is $k + 2$. Thus, the degree of terms of order λ^n in the Hamiltonian is not affected by the contact transformation.

The final result which we have been seeking is that to terms of order λ^N (N arbitrary), the Hamiltonian may be transformed to the form

$$H(p, q) = \frac{\hbar\omega}{2} J + \lambda^2 H_N(J) + \dots + \lambda^N H_N(J) \quad (116)$$

where $H_N(J)$ ($n \leq N$) is a polynomial of degree $n/2$ in J if n is even, and vanishes identically if n is odd.

Now let us consider the consequences of having the Hamiltonian in the form of Eq. (114). First of all, it is clear that

$$J = \frac{i}{\hbar} [H, J] = 0 \quad (117)$$

so that $J = pq + qp$ is a constant of the motion. Using

$$[J^n, p] = J^{n-1} [J, p] + [J^{n-1}, p] J \quad (118)$$

$$[J, p] = -2p \quad ,$$

we find that

$$p = \frac{i}{\hbar} [H, p] = i \Omega(J) p \quad ,$$

$$p(t) = e^{i\Omega t} p(0) \quad , \quad (120)$$

where $\Omega(J)$ is a polynomial in J . Further, since

$$J = pq + qp = 2pq + 1 = \text{const}, \quad (121)$$

we must have

$$q(t) = q(0) e^{i\Omega t}. \quad (122)$$

Thus, the creation and annihilation operators p and q , respectively (p and q remain creation and annihilation operators under a unitary transformation), have simple periodic solutions in terms of the operator quantities Ω and J .

Having obtained new p and q in terms of which the Hamiltonian takes form (114), it is a simple though perhaps tedious matter to obtain the old p and q by means of the transformation

$$\left. \begin{aligned} p \text{ old} &= U^{-1} p \text{ new} U \\ q \text{ old} &= U^{-1} q \text{ new} U \end{aligned} \right\}, \quad (123)$$

which is the inverse of the transformation (101). The result for p old and q old is a power series in λ with coefficients which are second- and higher-degree polynomials in p new and q new.

The only difference between p and q and the harmonic oscillator creation and annihilation operators lies in the nature of the time dependence of the solutions. For the harmonic oscillator the quantity Ω in (120) and (122) is a constant which is independent of the state of the dynamical system. For the anharmonic oscillator Ω depends on J and, hence, on the energy of the system.

It is also useful to note that harmonic oscillator states are simultaneously eigenstates of J , of the H in Eq. (114), and of $\Omega(J)$. From this and the fact that p and q connect only those pairs of harmonic oscillator states whose quantum numbers differ by 0 or ± 1 , it is easy to show that

$$\Omega_m = \langle m | \Omega | m \rangle = \frac{(E_{m+1} - E_m)}{\hbar},$$

where E_m is the energy. Thus, Ω is diagonal in the harmonic oscillator representation, and the diagonal elements Ω_m of Ω are equal to the energy spacings of the successive levels of the anharmonic oscillator.

One summarizes the foregoing perturbation formalism by stating that the transformed operators p new and q new are creation and annihilation operators, respectively, that the anharmonic oscillator eigenstates are simultaneously eigenstates of the Hamiltonian (116) and that in effect the

only difference between the anharmonic problem and the harmonic one lies in the values of the characteristic energies.

3.5.2. Neutron Scattering by a Single Anharmonically Vibrating Particle

As we shall demonstrate, the perturbation formalism discussed in Section 3.5.1 is ideally suited for discussing neutron scattering by an anharmonically vibrating particle. For the neutron scattering problem, we need to calculate

$$S(\mathcal{N}, \epsilon) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\epsilon t} \langle \exp[-i\mathcal{N} \cdot u(t)] \exp[i\mathcal{N} \cdot u(0)] \rangle dt ,$$

where ϵ and $\mathcal{N}$ are the energy and momentum, respectively, transferred from the neutron to the crystal (we use units where $\hbar = 1$, $\kappa_{\beta} = \text{Boltzmann's constant} = 1$) and

$$u(t) = ix(t) + jy(t) + kz(t)$$

is the operator representing the physical displacement of the particle from its position of equilibrium. For convenience, we shall assume that the Hamiltonian separates into a sum of three one-dimensional Hamiltonians; i. e.,

$$H = H_x + H_y + H_z ,$$

where H_x depends only on x and its conjugate momentum, etc. With this assumption we must evaluate

$$\chi_x^{(\kappa, t)} = \langle \exp[-i\kappa_x x(t)] \exp[i\kappa_x x(0)] \rangle$$

and similar quantities involving $y(t)$ and $z(t)$.

Maradudin and Fine have shown that, for any operators x and y

$$\begin{aligned} \langle e^{x} e^{y} \rangle = & \exp \left\{ \frac{1}{2} \langle x^2 + y^2 \rangle + \frac{1}{24} [\langle x^4 \rangle - 3 \langle x^2 \rangle^2 + \langle y^4 \rangle - 3 \langle y^2 \rangle^2 + \dots] \right\} \\ & \times \exp \left\{ \langle xy \rangle + \frac{1}{2} \langle x^2 y + xy^2 \rangle + \frac{1}{24} [\langle 4x^3 y + 6x^2 y^2 + 4xy^3 \rangle] - \frac{1}{8} [4 \langle xy \rangle^2 \right. \\ & \left. + 4 \langle x^2 \rangle \langle xy \rangle + 4 \langle xy \rangle \langle y^2 \rangle + 2 \langle x^2 \rangle \langle y^2 \rangle] + \dots \right\} , \end{aligned}$$

where ... indicates terms of order greater than four. In the present context, the operators x and y have the following significance:

$$x = -i \gamma_x x(t) ,$$

$$y = i \gamma_x x(0) .$$

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