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INTEGRAL NEUTRON THERMALIZATION

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1. INTRODUCTION AND SUMMARY

This annual summary report describes the work performed on the Integral Neutron Thermalization Program from October 1, 1967 through September 30, 1968, under Contract AT(04-3)-167, Project Agreement No. 2 with the U. S. Atomic Energy Commission. A general review of the experimental, analytical, and theoretical aspects of the work performed under this program is contained in the seven previous annual reports. (1-7)

During this contract year the first principles calculation of beryllium phonon dispersion curves was completed. Results are summarized in Section 4.2. The magnitude of the theoretical and numerical difficulties that had to be overcome in the performance of this work probably precludes the direct use of this type of calculation in a description of moderator properties for practical purposes. However, during the investigation it was discovered that the relatively simple free-electron model probably has a range of applicability significantly wider than formerly expected. This fact may well have practical importance and, together with an improved understanding of the complex relationship between the physics and mathematics in the first principles calculation, is an important product of this study.

Much other theoretical work has also been completed. For BeO a very elaborate lattice dynamical treatment has obtained greatly improved agreement, compared to rigid ion model results, with measured Raman frequencies and elastic constants. A scattering law based on this model has been prepared and comparisons have been made between the calculated and measured total cross section. This work is summarized in

Section 4. 4. New models for diphenyl, polyethylene, and UO_2 have also been developed and used in neutron scattering calculations. Further checks on the model for zirconium hydride, developed previously, have been carried out.

Some further development and refinement of theoretical techniques has been accomplished. The contribution from diffusive modes to the scattering law for water has been calculated, as described in Section 2. 6. Also, the validity of the short collision time approximation, which is of great value in reducing the cost of computing scattering laws, has been studied by making comparisons to exact results for water. The results, discussed in Section 2. 3, confirm the adequacy of the approximation in the range of energy- and momentum transfers for which it has been used in calculations at Gulf General Atomic. Progress has also been made in improving our capability of calculating one-phonon inelastic scattering, as discussed in Section 4. 3. This work is an important advance beyond the incoherent approximation, which has been a serious limitation on the adequacy of theoretical inelastic scattering calculations for non-hydrogenous moderators, both at Gulf General Atomic and elsewhere. An improved computation of the slowing down sources is discussed in Section 6. 2. Additional work on multiple scattering in double differential water measurements, summarized in Section 2. 5, has turned out to have a timely application to recently reported measurements taken with an unusual sample design. Finally, some preliminary work concerning the temperature dependence of phonon frequency spectra is described in Section 6. 1. This subject will be studied at greater length in the coming year.

Experimental work during the contract year includes a measurement of the total neutron cross section of uranium dicarbide (Section 5. 3) and further analysis of previously measured neutron spectra in ^{235}U and ^{238}U (Section 7). The precision achieved in these spectral data and the sensitivity of the accompanying theoretical analyses to cross section sets

indicate the usefulness of these measurements. The sensitivity of the results of kinetic studies in a ^{238}U sphere to inelastic neutron scattering cross sections has been investigated by comparing measured results to calculations performed with a simplified Monte Carlo code (see Section 7.4). Finally, new analyses of thermal neutron data taken in previous contract years are described in Sections 2.2 and 2.4. A description of the over-all facility improvements is given in Section 9.

In view of the large amount of information accumulated during work performed under this program, it is important that these results be disseminated in an orderly and systematic fashion and that appropriate summaries of pertinent details be available in a form convenient for users. Completed scattering laws are submitted to the ENDF (Section 8.2) while details of their construction are summarized in the so-called kernel book, described in Section 8.3. The Spectrum Book, discussed in Section 8.4, provides a similar source of descriptions of important neutron spectra measurements. Information has also been disseminated by means of presentations at meetings, (8-12) topical reports, (13-24, 73) and journal articles. (25-30)

Discussions in this annual report have been confined to brief summaries, stressing the results and significance of the work, in those cases in which a detailed exposition is available in the form of a topical report. Work not covered in topical reports is described at greater length.

2. NEUTRON THERMALIZATION IN H_2O AND D_2O

During this year, no new models for H_2O or D_2O have been developed. This situation reflects the fact that the scattering laws generated for these moderators by Gulf General Atomic and placed on the ENDF/A file have not yet received widespread use. Studies this year have therefore concentrated on ancillary matters. Among these are the final analysis of spectrum measurements across a TRIGA fuel element surrounded by cadmium-poisoned D_2O (Section 2.3) and the measurement of $\bar{T}$ for H_2O (Section 2.1). Further calculations have been performed which indicate that the procedure used by Brugger⁽³¹⁾ to correct for multiple scattering in his H_2O samples may underestimate the necessary correction. This work is described in Section 2.4.

The most important study completed this year is the comparison of the short collision time approximation to the scattering law with an exact calculation. This study was motivated by the requirement that future scattering laws be generated appropriate for an incident neutron energy of up to 10 eV. The study shows that the approximation is generally reasonable above 2 eV for H_2O and should be more than adequate for reactor calculations.

A further refinement to the existing theoretical calculations of the water scattering law is the inclusion of diffusive modes, as described in Section 2.5. While this refinement leads to improved agreement with experiment for very low energy exchange it is not yet clear that a similar improvement could not have been obtained by adding low frequency modes to the frequency distribution or by introducing a model containing freely-recoiling aggregates of various molecular weights. More extensive use of the

theoretical scattering laws as well as more precise experimental data are needed to establish firmly the importance of the diffusive modes.

2.1 $\bar{T}$ FOR H_2O

In principle, it is possible to apply the Placzek⁽³²⁾ expansion to the measured total cross section for water, σ_T , to obtain $\bar{T}$, which is 2/3 of the average kinetic energy of the bound hydrogen atoms in H_2O . This expansion has the following form

$$\sigma_T = \sigma_O + 2\sigma_a + \sigma_s \quad (2.1a)$$

where

$$\sigma_s = 2\sigma_{fa} \left[1 + \frac{\bar{T}}{2ME} - \frac{1}{32} \left(\frac{M+1}{M} \right)^2 \frac{\bar{C}}{ME^3} + O(E)^{-4} \right] \quad (2.1b)$$

where

σ_T = total cross section for water

σ_O = total cross section for oxygen

σ_{fa} = free atom cross section for hydrogen

σ_a = absorption cross section for hydrogen

E = neutron energy

M = mass ratio of hydrogen to neutron.

We have used this expansion up to the E^{-3} term to make a least squares fit to the data measured by Russell.⁽³³⁾ The value of $\bar{C}$ obtained was found to vary considerably in both sign and magnitude depending on the sample thickness and the number of points used in the least squares fit. In addition, the statistical uncertainty of $\bar{C}$ was found to be larger than the magnitude of $\bar{C}$ itself. This indicates that the observed scattering cross section for H_2O does not exhibit the functional dependence given by the above equation out to E^{-3} . This lack of an observed E^{-3} dependence may be due either to insufficient precision of the data or alternatively

to the difficulties of fitting an observed quantity to an asymptotic expansion. For each energy E , there is an inherent error in the expansion, which is not necessarily reduced by adding further terms (including perhaps the E^{-3} term).

$\overline{C}$ can be computed from the expression below

$$\overline{C} = \frac{1}{2} \sum_i \lambda_i \omega_i^3 \coth\left(\frac{\omega_i}{2T}\right)$$

where

T = temperature

λ_i = weightings based on effective masses

ω_i = molecular excitation energies

We have used the parameters of the Nelkin model⁽³⁴⁾ to obtain a value for $\overline{C}$ which then allowed us to correct the observed total cross section for water. A linear least squares fit of σ_s to $1/E$ was then made, but the resulting $\overline{T}$ was found to be larger than the value obtained without this correction. This correction procedure, (which was used previously)⁽³²⁾ is considered unrealistic in view of the absence of an observed E^{-3} behavior.

A similar total cross section measurement was made in polyethylene by Armstrong⁽³⁵⁾ who also used the above correction procedure and also obtained a value that was greater than the uncorrected value. In addition, Armstrong's $\overline{T}$ for polyethylene was high compared to the predictions of the Koppel model.⁽³⁶⁾ Therefore, we have decided to use only the linear fit to σ_s . The coefficients were obtained from each sample thickness and appropriately weighted and summed for the data covering an energy range from 16 eV to 0.83 eV. This led to a value of $\sigma_{fa} = 20.366 \pm 0.076$ for hydrogen and a $\overline{T}$ given in Table 2.1.

Table 2.1
 $\bar{T}$ FOR H_2O

$\bar{T}$ (eV)	Source
0.1240 ± 0.0078	Measured here
0.134 ± 0.013	Melkonian ⁽³⁷⁾
0.1168	Nelkin Model
0.1204	Haywood II Model ^(38, 39)

$\bar{T}$ for the Young-Koppel anisotropic model for H_2O has not been calculated; however it is believed to be extremely close to the value for the Nelkin model. The measured $\bar{T}$ is in statistical agreement with the predictions of the Haywood II model, which is considered to be the most reliable theoretical scattering law for water developed at Gulf General Atomic. $\bar{T}$ for the McMurry-Russell model,⁽⁴⁰⁾ developed elsewhere, was not available for inclusion in this comparison. However, the measured value of $\bar{T}$ is probably not precise enough to serve as a strict test of this model.

2.2 COMPARISON OF EXACT AND SHORT COLLISION CALCULATIONS OF THE SCATTERING LAW FOR WATER

Since it is generally too expensive to extend the exact GASKET calculation⁽⁴¹⁾ of the scattering law for a given material beyond say 2.0 eV ($\beta \sim 80$ at room temperature), it has been standard practice to use the short collision time approximation⁽⁴²⁾ for all vibrational modes for the scattering law data calculated in the range 2.0-10.0 eV. Beyond 10.0 eV it is generally safe to ignore all binding effects.

The short collision approximation to the scattering law is given by the expression⁽⁴¹⁾

$$S_{SC}(\alpha, \beta) = e^{\beta/2} \frac{\exp \left[-\frac{T}{4\alpha\bar{T}} (\beta + \alpha)^2 \right]}{\sqrt{4\pi\alpha T \bar{T}}}, \quad (2.2)$$

where $\bar{T}$ as in Section 2.1 is two thirds of the average kinetic energy of the scattering nuclei (setting Boltzmann's constant $k_B = 1$).

The dimensionless variables α and β are defined as

$$\alpha = \frac{K^2}{2MT} \quad (2.2a)$$

$$\beta = \frac{\varepsilon}{T} \quad (2.2b)$$

where K and ε are respectively the neutron momentum and energy exchanges, M the atomic mass of the scatterer, and T the temperature in energy units. The expression (Eq. 2.2) is meant only for downscattering ($\beta < 0$) since it does not satisfy detailed balance

$$S_{SC}(\alpha, \beta) \neq S_{SC}(\alpha, -\beta). \quad (2.3)$$

This lack of symmetry is not important, however, since the short collision approximation is to be used for large values of $|\beta|$ where upscattering ($\beta > 0$) becomes very small. In order to obtain a double differential cross section which nevertheless does satisfy detailed balance one uses the following relations:

$$\sigma(E \rightarrow E', \theta) = \frac{\sigma_b}{4\pi} \sqrt{\frac{E'}{E}} e^{|\beta|/2} S_{SC}(\alpha, -|\beta|) \text{ for downscattering } (\beta < 0) \quad (2.4)$$

and

$$\sigma(E \rightarrow E', \theta) = \frac{\sigma_b}{4\pi} \sqrt{\frac{E'}{E}} e^{-|\beta|/2} S_{SC}(\alpha, -|\beta|) \text{ for upscattering } (\beta > 0). \quad (2.5)$$

Since H_2O is of all moderators the one with the highest vibrational frequencies ($\omega \approx .44$ eV) it probably constitutes the most severe check of the adequacy of the short collision approximation. Thus, we extended the exact calculations of the H_2O scattering law up to 3 eV ($\beta = 120$) and compared it to the results obtained using Eq. 2.2. This comparison is shown in Figs. 2.1, 2.2, and 2.3, for three different values of α . It is seen that the agreement is reasonable in the region of the maximum of $e^{-\beta/2}S(\alpha, \beta)$, i. e. when $|\beta|$ approximately equals α , and improves with increasing α . This is the expected behavior since the RHS of Eq. 2.2 is the first term of an asymptotic expansion valid for large α and $\beta \sim \alpha$.

The short collision data are commonly used only for $\beta > 80$ or $\alpha > 320$ (at room temperature). Thus, it is believed that the procedure is satisfactory since large discrepancies will occur only when $e^{\beta/2}S(\alpha, \beta)$ is small.

2.3 REANALYSIS OF THE SPECTRUM MEASUREMENTS ACROSS A TRIGA FUEL ELEMENT SURROUNDED BY CADMIUM POISONED D_2O

Spectral measurements were made previously⁽⁶⁾ across a TRIGA fuel element surrounded by cadmium-poisoned D_2O . These measurements were designed to check the feasibility of performing spectral measurements in strongly heterogeneous assemblies and also to check the adequacy of the scattering descriptions for D_2O and zirconium hydride. (The latter material is contained in the fuel element.) The earlier analysis⁽⁶⁾ of these data indicated an extreme sensitivity of the flux to the spatial distribution of the source. This observation was not checked at the time because our computing equipment and computational methods were being changed. During this year we have reanalyzed these data using the discrete ordinates code 1DF⁽⁴³⁾ instead of GAPLSN⁽⁴⁴⁾ and a revised version of the DSZ code⁽²⁰⁾ to calculate the space and energy dependent

Figure 2.2. Comparison of exact and approximate (short collision) scattering law for H_2O (Haywood II frequency spectrum).^(38, 39)

Figure 2.3. Comparison of exact and approximate (short collision) scattering law for H_2O (Haywood II frequency spectrum). (38, 39)

slowing down source. The newer DSZ code⁽²⁰⁾ is described in Section 6.2. The resulting comparison of theory and measurement is given in Fig. 2.4. This comparison differs little from the previous one and has therefore been incorporated into the Spectrum Book.⁽⁴⁵⁾ The effect of varying the spatial source distribution was also studied and no strong sensitivity was found. It was concluded that the previous observations on this sensitivity were erroneous and were due to the inadequacy of the convergence of the GAPLSN calculation for the flux. These experimental data have now been written up along with other heterogeneous measurements in an article which has been accepted for publication by Nukleonik.

2.4 MULTIPLE SCATTERING AS A FUNCTION OF NEUTRON PATH LENGTH

This past year a further study of multiple scattering in neutron double differential water measurements was undertaken to investigate the use of special sample designs in such measurements. By "special samples" we mean samples of a relatively complicated geometry incorporating neutron absorbing materials for the purpose of cutting down multiple scattering. An example would be a framework of parallel aluminum tubes containing water and separated by gadolinium foils. Such a sample was used in measurements recently reported by Brugger,⁽³¹⁾ who obtained the somewhat surprising result that at low momentum transfer the scattering law for room temperature water is linear in the momentum transfer rather than quadratic as expected on rather fundamental grounds. The multiple scattering corrections in this experiment were based on an extrapolation procedure involving data taken with different spacing of the foils in the special sample. No real justification was given for the extrapolation procedure except that the resulting corrections were "in the right direction." Thus, there remains the possibility that the apparent unusual behavior of the observed scattering law is a consequence of inaccuracies in the multiple scattering correction.

Figure 2.4. Comparison of measured and calculated neutron spectra across a TRIGA fuel element surrounded by D_2O .

The results of recent work at Gulf General Atomic suggest strongly that such a conclusion should be drawn. Our calculations were based on a modification of the MUSE code⁽⁴⁶⁾ to permit description of a slab sample in which multiply-scattered neutrons are absorbed after traveling a certain path length within the sample. This path length plays the role of the foil separation in a realistic sample. By varying the path length we were thus able to get quantitative estimates of the foil separations required to achieve varying degrees of suppression of multiple scattering effects. It was found that a substantial amount of multiple scattering involves neutron path lengths shorter than the smallest spacings that seem reasonably attainable in actual samples. Extrapolation procedures based on the use of larger foil separations were shown in pertinent cases to underestimate seriously the multiple scattering correction, particularly for low momentum transfer data. Details of the work are given in a topical report.⁽²¹⁾

2.5 DIFFUSIVE MODES IN H₂O*

Although the very low frequency end of the generalized frequency spectrum (spectrum of the velocity auto-correlation) for water is not very important for calculating neutron spectra in nuclear reactors, it strongly affects double differential neutron scattering experiments with very small energy transfer.

The generalized frequency spectrum $f(\omega)$ for H bound in H₂O has been measured by a number of workers using the technique of inelastic neutron scattering, either by Egelstaff's extrapolation technique⁽⁴⁷⁾ or simply by approximating the double differential cross section by the one-phonon (incoherent) term, which is adequate for small values of the variable α ,

$$\alpha = \frac{\hbar K^2}{2M\omega_c} \quad , \quad (2.6)$$

* Much of the work in this section was supported by the Office of Saline Water under Contract 14-01-0001-1705. It is reported here as well as in reports associated with that project (G. M. Borgonovi, GA-8558, 2-14-69) because the computer codes essential to this work were developed with AEC support under Project 48.01 and because the study of diffusive modes is an important contribution to the understanding of neutron thermalization in water.

where K is the momentum exchange, M is the mass of the scattering atoms (in the present case protons) ω_c is the cut off frequency and $\hbar$ is Planck's constant.

In principle it is possible to determine from these measurements the diffusion coefficient D through the relation

$$D = \frac{\pi T}{2M} f(0) , \quad (2.7)$$

where $f(\omega)$ is the frequency spectrum associated with the scatterer of mass M , T is the temperature and Boltzmann's constant has been set to unity. However, more accurate values of D can be obtained from other measurements such as tracing techniques or nuclear magnetic resonance methods.

In the Gaussian approximation the intermediate scattering function⁽⁴¹⁾ is

$$X(K^2, t) = \exp \left[- \frac{\hbar^2 K^2}{2M} \gamma(t) \right] , \quad (2.8)$$

where

$$\gamma(t) = \frac{1}{\hbar} \int_0^\infty f(\omega) \left[\coth \frac{\omega}{2T} (1 - \cos \omega t) - i \sin \omega t \right] \frac{d\omega}{\omega} , \quad (2.9)$$

and $f(\omega)$ is normalized to 1.

The frequency spectrum can always be broken into two terms

$$f(\omega) = f_D(\omega) + f_H(\omega) \quad (2.10)$$

such that

$$f_D(\omega) \leq f(\omega) \quad (2.11)$$

and

$$f_D(0) = f(0) , \quad (2.12)$$

$f_D(\omega)$ can then be interpreted as the diffusive component of $f(\omega)$ while $f_H(\omega)$ describes the vibrational (harmonic) component.

For a given $f(\omega)$ this decomposition is of course not unique. It becomes well determined, however, once a model for the diffusion process is chosen.

We then have

$$X = X_D X_H , \quad (2.13)$$

where

$$X_D(K^2, t) = \exp \left[- \frac{\hbar^2 K^2}{2M^*} \gamma_D(t) \right] , \quad (2.14)$$

and where $\gamma_D(t)$ is given by Eq. 2.9 with $f(\omega)$ replaced by

$$f_D^{\text{norm}}(\omega) = \frac{f_D(\omega)}{\int_0^\infty f_D(\omega) d\omega} = f_D(\omega) \frac{M^*}{M} . \quad (2.15)$$

Similarly

$$X_H(K^2, t) = \exp \left[- \frac{\hbar^2 K^2}{2} \left(\frac{1}{M} - \frac{1}{M^*} \right) \gamma_H(t) \right] \quad (2.16)$$

where $\gamma_H(t)$ is also given by Eq. 2.9 with $f(\omega)$ replaced by

$$f_H^{\text{norm}}(\omega) = \frac{M^*}{M^* - M} f_H(\omega) . \quad (2.17)$$

Both f_D^{norm} and f_H^{norm} are normalized to 1. X_D describes the diffusion of particles or aggregates of mass M^* and is characterized by its asymptotic behavior

$$X_D(K^2, t) \xrightarrow{t \rightarrow \infty} e^{-K^2 D t} . \quad (2.18)$$

The simplest analytical form of X_D which satisfies this condition as well as the sum rules for the zeroth and first moments of its Fourier transform (Placzek sum rules)⁽³²⁾ was proposed by Egelstaff⁽⁴⁷⁾

$$X_D(K^2, t) = \exp \left\{ -K^2 D \left[\left(t^2 - \frac{i\hbar t}{T} + \frac{\hbar^2 c^2}{T^2} \right)^{\frac{1}{2}} - \frac{\hbar c}{T} \right] \right\} , \quad (2.19)$$

where D is the diffusion constant and c is a parameter describing the time delay before classical diffusion sets in. The corresponding f_D^{norm} is

$$f_D^{\text{norm}}(\omega) = \frac{4}{\pi} \frac{M^* D}{T} \left(c^2 + \frac{1}{4} \right)^{\frac{1}{2}} \sinh \frac{\hbar \omega}{2T} K_1 \left[\left(c^2 + \frac{1}{4} \right)^{\frac{1}{2}} \frac{\hbar \omega}{T} \right] . \quad (2.20)$$

Actually for this expression to be normalized to one the parameter c must have the value

$$c = M^* D / \hbar . \quad (2.21)$$

This also ensures that $X_D(K^2, t)$ satisfies the first two Placzek sum rules for scattering by (diffusing) particles of mass M^* .

Presently the best values for the diffusion coefficient of water are those obtained by Simpson and Carr⁽⁴⁸⁾ by the technique of nuclear magnetic resonance (NMR). Their results for different temperatures between 0°C and 100°C are given in Table 2. 2.

Table 2. 2
DIFFUSION COEFFICIENT FOR H₂O

Temperature (°C)	$D \times 10^5$ (cm ² /sec)	Temperature (°C)	$D \times 10^5$ (cm ² /sec)
0	0.97	55	4.37
5	1.16	60	4.82
10	1.36	65	5.30
15	1.58	70	5.78
20	1.85	75	6.27
25	2.13	80	6.81
30	2.46	85	7.26
35	2.70	90	7.75
40	3.14	95	8.20
45	3.52	100	8.65
50	3.94		

Because of the crudeness of the model the values of c are not so well defined as those of D . They can be obtained by different methods, which however do not give accurate quantitative agreement with each other.

One method of obtaining an experimental value of c is through Eyring's⁽⁴⁹⁾ formula for the diameter ($2r$) of the diffusing particle in terms of the viscosity η

$$2r = \frac{T}{\eta D} . \quad (2.22)$$

If a^3 is the volume of one H_2O molecule the radius of the diffusing particle (or globule) can also be expressed as

$$r = \left(\frac{M^*}{18M} \cdot \frac{3}{4\pi} \right)^{1/3} a, \quad (2.23)$$

where M is the proton mass. Then, using Eq. 2.21 it follows that

$$r = \left(\frac{c\hbar}{24\pi DM} \right)^{1/3} a \quad (2.24)$$

and introducing this result into Eq. 2.22 we get

$$c = \frac{\pi DM_W}{6\hbar} \left(\frac{T}{\eta D_a} \right)^3, \quad (2.25)$$

where $M_W = 18 M$ is the mass of one H_2O molecule.

The values of c can also be obtained directly from neutron scattering experiments by fitting the curve which gives the width of the quasi-elastic peak of the scattering law $S(K^2, \epsilon)$ as a function of K^2 .

Table 2.3 gives the values of c obtained by the two methods⁽⁵⁰⁾ and using different sets of experimental data.

It is seen that there is fair agreement between the results of Eq. 2.25 and the neutron scattering data obtained by Sakamoto⁽⁵¹⁾ who used the rotating crystal method. Larsson's data,⁽⁵²⁾ which were obtained by the beryllium filter technique, lead to a much higher value of c . However, due to the higher energy resolution at low β Sakamoto's data are probably more reliable.

Having found reasonable values for D and c we can determine the mass of diffusing particles from Eq. 2.21.

$$\frac{M^*}{M} = \frac{\hbar c}{MD}. \quad (2.26)$$

Table 2.3
VALUES OF THE TIME-DELAY PARAMETER c

<u>Method</u>	<u>Temperature ($^{\circ}\text{C}$)</u>	<u>$\hbar c/T$ (10^{-12} sec)</u>	<u>c</u>
Equation 2.25	0	1.3	46.5
	100	7.8	380
Neutron Scattering ⁽⁵¹⁾ (Sakamoto Data)	24	1.1	42.7
	75	1.5	68.3
Neutron Scattering ⁽⁵²⁾ (Larsson Data)	22	2.6	100
	70 - 92	~ 3.5	

For room temperature ($\sim 20^{\circ}\text{C}$) we find using $c \cong 40$ and $D = 1.85 \times 10^{-5}$ cm^2/sec

$$M^* \cong 1365 M \cong 76 M_W \quad (2.27)$$

Once D and c have been fixed the diffusive factor X_D of the intermediate scattering function X can be evaluated. To determine the vibrational factor X_H we need an estimate of the vibrational part $f_H(\omega)$ of the generalized frequency spectrum. For water it is known from optical measurements that there are three internal vibrations of the H_2O molecules with frequencies of 0.205, 0.43, and 0.44 eV, each one having a weight⁽⁵³⁾

$$\omega_1 = 1/6 . \quad (2.28)$$

Due to the relatively high values of the forces corresponding to these frequencies as compared to the intermolecular forces, the internal vibrations have rather sharp lines and can be approximated by δ -functions. In addition to the internal vibrations $f_H(\omega)$ must account for those vibrational modes in which the H_2O molecules move as a whole: a band of hindered rotations and a band of translational vibrations with a total weight

$$w_r + w_t = 1/2 - M/M^* , \quad (2.29)$$

such that

$$\int_0^\infty f_H(\omega) d\omega = \sum_{i=1}^3 w_i + w_r + w_t = 1 - M/M^* \quad (2.30)$$

in agreement with Eq. 2.17.

The rotational and translational part of $f_H(\omega)$ have been measured by several authors using the method of inelastic neutron scattering and Egelstaff's extrapolation technique or else the one-phonon approximation to the scattering law. Table 2.4 gives the results obtained by Haywood⁽⁵⁴⁾ expressed in terms of the variable $\beta = \omega/T$ and with the normalization

$$\int f_{\text{Haywood}}(\beta) d\beta = 0.5 .$$

In principle the neutron scattering experiment leads to the total frequency spectrum $f_D + f_H$. However, because of resolution problems the experimental results are not very reliable at very low frequencies where f_D is significantly different from zero. Hence, it is preferable to split X into the two factors of Eq. 2.13 and determine the parameters c and D as described above. We can then disregard the very low frequency part of the experimental $f(\omega)$ and extrapolate it smoothly to zero. Actually this is what probably was done with the data leading to Table 2.4 since it is seen that $f_{\text{Haywood}}(0) = 0$.

Having determined the parameters c and M/M^* and knowing $f_H(\omega)$ we can calculate the scattering law using the code GASKET.⁽⁴¹⁾ With the values

Table 2.4
HAYWOOD⁽⁵⁴⁾ FREQUENCY SPECTRUM FOR H₂O

<u>β</u>	<u>$f(\beta)$</u>	<u>β</u>	<u>$f(\beta)$</u>
0.0	0.00	3.7	0.068
0.1	0.006	3.8	0.064
0.2	0.024	3.9	0.060
0.3	0.037	4.0	0.057
0.4	0.043	4.1	0.054
0.5	0.053	4.2	0.052
0.6	0.053	4.3	0.050
0.7	0.041	4.4	0.048
0.8	0.031	4.5	0.047
0.9	0.039	4.6	0.045
1.0	0.044	4.7	0.044
1.1	0.039	4.8	0.042
1.2	0.037	4.9	0.041
1.3	0.040	5.0	0.039
1.4	0.043	5.1	0.039
1.5	0.047	5.2	0.039
1.6	0.051	5.3	0.038
1.7	0.055	5.4	0.038
1.8	0.061	5.5	0.038
1.9	0.068	5.6	0.038
2.0	0.075	5.7	0.037
2.1	0.083	5.8	0.037
2.2	0.091	5.9	0.037
2.3	0.100	6.0	0.037
2.4	0.108	6.1	0.037
2.5	0.114	6.2	0.036
2.6	0.120	6.3	0.036
2.7	0.120	6.4	0.036
2.8	0.120	6.5	0.036
2.9	0.116	7.0	0.036
3.0	0.108	7.5	0.036
3.1	0.102	8.0	0.036
3.2	0.095	8.5	0.036
3.3	0.089	9.0	0.036
3.4	0.083	9.5	0.036
3.5	0.078	10.0	0.036
3.6	0.073		0.0

$$c = 40$$

$$\frac{M^*}{M} = 1365 ,$$

we obtained the curves of Figs. 2.5, 2.6, and 2.7, which also show a comparison with Haywood's measurements and with previous calculations* using a model of freely recoiling molecules of mass 18. It is clearly seen that for low energy exchange the present results represent an improvement. For large energy exchange there should be no significant difference between the two models.

*All calculations are for H-scattering only; scattering by oxygen has not been included.

Figure 2.5. Scattering law for H_2O ($\beta = 0.2$).

Figure 2.6. Scattering law for H_2O ($\beta = 0.40$).

Figure 2.7. Scattering law for H₂O ($\beta = 1.0$).

3. NEUTRON THERMALIZATION IN DIPHENYL AND POLYETHYLENE

3.1 INTRODUCTION

The neutron scattering by diphenyl has been studied in the past⁽⁵⁵⁻⁵⁷⁾ using models based on the dynamical properties of the benzene molecule. Evidence that this is not a good approximation⁽⁵⁷⁾ has motivated the development of a new model based entirely on the dynamic properties of diphenyl itself. The work done on this topic and the results obtained are summarized in Section 3.2.

Goldman and Federighi⁽⁵⁸⁾ presented a model for polyethylene in which the molecular dynamics is simulated by four harmonic oscillators and free recoiling CH_2 radicals. However, the frequencies assigned to the oscillators are in contradiction to the experimental data obtained by optical methods.⁽⁵⁹⁾ A second model, developed by Koppel and Young,⁽³⁶⁾ obtains the frequency spectrum from the dispersion relations (calculated by Lin and Koenig)⁽⁶⁰⁾ for an infinite chain of CH_2 radicals. The weights given to the normal modes in the weighted frequency spectrum used for neutron scattering calculations were determined from the assumptions that the carbon and the hydrogen atoms move with equal amplitudes in the modes involving skeletal deformations and that the carbon atoms are nearly at rest in the remaining optical modes. It is surprising that the model of Goldman and Federighi gives a better prediction of the total cross section than the Koppel-Young model, which has a more physical basis. However, a refined version of the Koppel-Young model, developed this year and described in Section 3.3, gives a greatly improved prediction of the total cross section.

3.2 NEUTRON SCATTERING BY DIPHENYL

The scattering by diphenyl was parametrized in terms of a harmonic model in which the oscillations of the hydrogen and carbon atoms are determined by the interatomic forces. A set of force constants was found by using a least squares technique that gave a best fit to the vibrational frequencies of the molecule measured by optical methods. The initial values for the force constants were taken from the forces computed for benzene in previous work.^(29, 61) For the modes corresponding to in-plane vibrations the force field for diphenyl is determined by 17 parameters while only nine parameters were required to describe the out-of-plane vibrations. The hindered translations and rotations corresponding to the liquid state can be treated as free translations of the whole molecule with an effective mass. Comparisons between predictions based on the model and the experimental neutron scattering results show very good agreement. The predictions of this model have also been compared with the results calculated with the old model based on benzene dynamics. The comparisons seem to indicate that as far as the integral data are concerned the two models will produce very similar results. The main differences will be found in the double differential cross section at low energy transfer.

A complete report on diphenyl has been written⁽¹³⁾ and has been accepted for publication in Nuclear Science and Engineering.

3.3 NEUTRON SCATTERING BY POLYETHYLENE

The dispersion relations for the infinite chain of CH_2 radicals, as well as the polarization vectors for each normal frequency, have been calculated using the set of force constants determined by Lin and Koenig. The weighted frequency spectrum for neutron scattering calculations was then obtained from the computed dispersion relations with appropriate weightings based on the computed amplitude vectors, rather than the

assumed amplitudes used in the Koppel-Young model.⁽³⁶⁾ The scattering law for polyethylene was determined (in the incoherent approximation) by using this weighted spectrum as input to the code GASKET.⁽⁴¹⁾ Comparison of neutron cross sections and neutron spectra based on this scattering law with experimental data indicates that this model is satisfactory. In particular, the predictions of the total cross sections and neutron spectra obtained from this model are in much better agreement with the experimental data than the ones obtained with the models of Refs. 36 and 58.

A report on polyethylene⁽¹⁴⁾ has been written and has been accepted for publication in Nukleonik.

4. NEUTRON THERMALIZATION IN BERYLLIUM AND BERYLLIUM OXIDE

4.1 INTRODUCTION

The theoretical study of the coherent moderators beryllium and beryllium oxide has been pursued in an attempt to produce thermal cross sections that depend on a minimum number of arbitrary parameters.

Three different approaches, which correspond to different stages in the understanding of the thermalization process, have been used. The first approach is the calculation of the lattice dynamics of beryllium starting from first principles. A summary of the results and conclusions obtained with this type of calculation is given in Section 4.2. Here the main conclusion seems to be that the simplest (and unrealistic) free electron model gives a better agreement than practical calculations using the exact electronic band structure of beryllium. The second approach is a calculation of the coherent scattering cross section for a polycrystalline beryllium specimen. A new technique for calculating the one-phonon coherent cross section is being used and is described in Section 4.3. The main advantage of the new technique over the one used previously is that more accurate results can be obtained using a shorter computer time.

The third approach consists of a phenomenological lattice dynamical calculation, using the formalism of the shell model, of the vibrational frequencies of beryllium oxide, and the use of the incoherent approximation to calculate the scattering cross section. A noteworthy improvement has been obtained over the previously used rigid ion model. A summary of this work is reported in Section 4.4.

4.2 FIRST PRINCIPLES CALCULATION OF PHONON DISPERSION CURVES OF BERYLLIUM

The ab initio calculation of phonon dispersion curves in beryllium has been completed and a detailed account of the theory and results has been published.⁽²³⁾ The main results of this study are described here:

1. Despite the rather distorted Fermi surface of beryllium, the calculated phonon frequencies are in fair agreement with experiment if the free electron screening function is used in conjunction with a self-consistent bare ion potential to describe the motion of the conduction electrons. The agreement is particularly good for the hardest of the optical branches. Also the elastic constants C_{11} , C_{33} , and C_{44} (beryllium has the hcp crystal structure) are quite well predicted. For other branches and elastic constants the agreement is less satisfactory.
2. Calculations using a dielectric screening matrix obtained by considering the exact electronic band structure of beryllium run into severe convergence problems. Good convergence can be obtained only when the off-diagonal elements of the dielectric matrix are neglected, but the corresponding phonon frequencies and elastic constants are in poorer agreement with experiment than the free electron values.

Conclusions to be drawn from these results are: (1) the effect of the beryllium lattice on the screening properties of the conduction electrons cannot be adequately described by only the diagonal elements of the screening matrix, and (2) the off-diagonal elements, despite their relative smallness, give an important compensating contribution to the phonon frequencies and to the elastic constants. However, an accurate calculation of the off-diagonal contribution to the screening would require extending the reciprocal lattice sums over more shells than can be handled in a reasonable time by present day computers.

The first and second Brillouin zones and two cuts through the Fermi surface (in the extended zone) are shown in Figs. 4.1 through 4.4.

Figure 4.1. First Brillouin zone of the hexagonal closed packed structure.

Figure 4.2. Second Brillouin zone of the hexagonal closed packed structure.

Figure 4.3. Fermi surface of beryllium: cut AHK Γ .

Figure 4.4. Fermi surface of beryllium: cut $ALM\Gamma$.

It is seen that the free area of the Fermi surface is much reduced and that the contact with the zone boundary is very extensive. Thus, the good results obtained from the free electron model are quite surprising and lead to the reasonable conjecture that, in general, even for strongly distorted Fermi surfaces the dispersion curves for any metal (except perhaps the transition metals), can be fairly well predicted by the free electron model. This knowledge would be of value in cases where accurate measurements have not yet been made or in the case of an incoherent neutron scatterer, for which the usual coherent inelastic neutron scattering technique for measuring the dispersion curves fails.

4.3 ONE-PHONON COHERENT CROSS SECTION FOR Be

The scattering of slow neutrons by beryllium has been treated in the past using the incoherent approximation.⁽⁶¹⁾ For small values of the momentum exchange this treatment is not adequate for Be since this material has a large coherent cross section. In previous years an attempt was made to compute the coherent one phonon inelastic cross section of Be.⁽⁶³⁾ However, the method of computation required a large amount of computing time and therefore was not suitable for computing the scattering law for a reasonable mesh of energy and momentum transfers. Thus, a more efficient procedure of computing one-phonon coherent cross sections had to be developed.

The coherent one-phonon double-differential cross section (per unit cell) can be written in the form

$$\frac{d^2\sigma}{dE d\Omega} = \frac{(2\pi)^3}{2MN} \frac{a^2 \hbar^2}{V} \frac{k}{k_0} \sum_{\vec{q}, \vec{t}, \theta} \left| \sum_{\vec{\ell}} e^{-W_{\vec{\ell}}} e^{i\vec{\alpha} \cdot \vec{r}_{\vec{\ell}}} \cdot \vec{\alpha} \cdot \vec{C} \left(\vec{\ell} \middle| \begin{smallmatrix} \vec{q} \\ \vec{j} \end{smallmatrix} \right) \right|^2 \cdot \frac{1}{\theta \hbar \omega_j(\vec{q})} \frac{1}{\frac{-\hbar \omega_j(\vec{q})}{k_B T} \theta} \delta(\epsilon + \theta \hbar \omega_j(\vec{q})) \delta(\vec{\alpha} - 2\pi \vec{q} - 2\pi \vec{t}) , \quad (4.1)$$

1-e

where W_ℓ , the exponent of the Debye-Waller factor, is given by

$$W^\ell = \frac{\hbar}{4NM} \sum_{\vec{q}} \frac{|\vec{\kappa} \cdot \vec{C}(\ell | \vec{q}_j)|^2}{\omega_j(\vec{q})} \coth\left(\frac{\hbar \omega_j(\vec{q})}{2k_B T}\right) \quad (4.2)$$

Planck's constant divided by 2π is denoted by $\hbar$, $\hbar k_0$ and $\hbar k$ are the initial and final momentum of the neutron respectively, a is the scattering length, M is the mass of the scatterer, N is the number of cells of the crystal, V is the volume of the unit cell, $\hbar \vec{\kappa} = \hbar(\vec{k} - \vec{k}_0)$ is the momentum transfer, $\omega_j(\vec{q})$ and $\vec{C}(\ell | \vec{q}_j)$ are the frequency and the amplitude vector of the ℓ -th atom in the mode j with wave vector $\vec{q}$, T is the temperature, k_B the Boltzmann's constant, $\vec{r}$ is a vector of the reciprocal lattice, and ℓ is the energy transfer defined as $\ell = E - E_0$, where E_0 and E are the initial and final neutron energies. The symbol θ is equal to plus or minus one according to whether a phonon is created or destroyed. It should be noticed that the structure factor in Eq. 4.1, namely

$$\left| \sum_{\ell} e^{-W_\ell} \vec{\kappa} \cdot \vec{C}(\ell | \vec{q}_j) e^{i\vec{\kappa} \cdot \vec{r}_\ell} \right|^2 \quad (4.3)$$

differs from the one given in Ref. 63 in the sign of the wave vector that identifies the polarization vector. The difference is due to a mistake in the derivation of the cross section given in Ref. 63.

The cross section can be written in terms of the scattering law

$$\frac{d^2\sigma}{dE d\Omega} = a^2 \frac{k}{k_0} e^{-\beta/2} \frac{S(\vec{\kappa}, \beta)}{k_B T} \quad (4.4)$$

with

$$S(\vec{\kappa}, \beta) = \frac{(2\pi)^3 \hbar^2}{2MNV} \sum_{\vec{q} \vec{t} \theta} \left| \sum_{\ell} e^{-W_{\ell}} e^{i\vec{\kappa} \cdot \vec{r}_{\ell}} \vec{\kappa} \cdot \vec{C}(\ell | \vec{q}_j) \right|^2 \cdot \frac{k_B T}{\hbar \omega_j(\vec{q})} \frac{1}{1 - e^{-\frac{\hbar \omega_j(\vec{q})}{k_B T}}} \delta(\ell + \theta \hbar \omega_j(\vec{q})) \delta(\vec{\kappa} - 2\pi \vec{q} - 2\pi \vec{t}), \quad (4.5)$$

where β is defined by Eq. 2.2b.

Performing the sum over $\vec{t}$ we have

$$S(\vec{\kappa}, \beta) = \frac{1}{2M} \sum_{j\theta} \left| \sum_{\ell} e^{-W_{\ell}} e^{-i\vec{\kappa} \cdot \vec{r}_{\ell}} \vec{\kappa} \cdot \vec{C}(\ell | \vec{\kappa}_j) \right|^2 \cdot \frac{e^{\beta/2}}{(e^{\beta} - 1)\beta} \delta(\beta T k_B - \theta \hbar \omega_j(\vec{\kappa})) \quad (4.6)$$

To obtain the cross section for polycrystalline Be we have to average over all possible orientations of the crystal, or equivalently, over all possible orientations of $\vec{\kappa}$. Thus for the polycrystal

$$S(\alpha, \beta) = \frac{1}{2M} \frac{1}{N_1 N_2} \sum_{\Phi \mu} \sum_{j\theta} \left| \sum_{\ell} e^{-W_{\ell}} e^{-i\vec{\kappa} \cdot \vec{r}_{\ell}} \vec{\kappa} \cdot \vec{C}(\ell | \vec{\kappa}_j) \right|^2 \cdot \frac{e^{\beta/2}}{(e^{\beta} - 1)\beta} \delta(\beta k_{\beta} T + \theta \omega_j(\vec{\kappa})) , \quad (4.7)$$

where α is defined by Eq. 2.2a and

where we have made the approximation of computing the average over-orientation using a mesh of spherical coordinates with N_1 indicating the number of azimuthal angles Φ and N_2 the number of polar angles θ ; the mesh adopted for the calculation was uniform in Φ and $\mu = \cos\theta$. Furthermore, the delta function was replaced by a box function, that is

$$\begin{aligned}\delta(\beta k_B T - \omega_j(\vec{k})) &= \frac{1}{2\Delta} \text{ for } \hbar\omega_j - \Delta \leq k_B T \beta \leq \hbar\omega_j + \Delta \\ &= 0 \text{ for } \omega_j - \Delta > \beta k_B T > \hbar\omega_j + \Delta .\end{aligned}\quad (4.8)$$

With this approximation a computer program COBE has been written to compute Eq. 4.7.

To compare the results of this work with the incoherent scattering law we had to recompute $S(\alpha, \beta)$ for beryllium in the incoherent approximation because of an error found in the computation of the frequency spectrum that was used in Ref. 62. The correct frequency spectrum is shown in Fig. 4.5. However, the differences from the previous (incorrect) spectrum are not very large and lead only to minor corrections of the previously calculated incoherent scattering law.

In Figs. 4.6, 4.7, and 4.8 we compare the coherent scattering law computed here with experimental data and with the incoherent approximation. Clearly there is no reasonable way by which the experimental data and the calculation using the incoherent approximation can be reconciled. On the other hand, resolution broadening of the coherent results can be expected to bring the sharp calculated peaks into at least qualitative agreement with the broad experimental results. To this extent, then, these preliminary results are encouraging and seem to indicate the correctness of the present calculation.

Due to the complicated structure of the scattering law for low values of β , a large number of points are needed in order to define the function. Since the present method still requires a large amount of

Figure 4.5. Frequency spectrum for Be.

Figure 4.6. Scattering law for Be.

Figure 4.7. Scattering law for Be.

Figure 4.8. Scattering law for Be.

computing time, several techniques are now being considered to make the code faster. Even in the present stage, however, this code is much faster than the one reported in Ref. 63.

4.4 LATTICE DYNAMICS AND NEUTRON SCATTERING PROPERTIES OF BeO

The moderator beryllium oxide has a relatively wide use in nuclear reactor design; therefore, it is important to have a good theoretical description of its neutron scattering properties. In the past, calculations have been made based on crude estimates of the lattice vibrational spectrum. Earlier calculations were made using a Debye frequency spectrum and a rigid ion model with short range interactions extending to the first neighbors.⁽⁶⁴⁾ More refined calculations have become possible recently as a result of the availability of new data. These data consist of elastic constants measured from single crystals,⁽⁶⁵⁾ new measurements of Raman frequencies⁽⁶⁶⁾ and coherent neutron scattering experiments from single crystals.⁽⁶⁷⁾ The rigid ion calculation proved not to be flexible enough to explain the detailed structure of the atomic motions in BeO. For this reason the more elaborate shell model approach was chosen. A complete shell model formulation of the BeO lattice dynamics, assuming polarizable oxygen atoms and second neighbor short range forces, was prepared during this contract year and is described in a topical report.⁽²⁴⁾

A set of parameters was subsequently chosen to fit the elastic constants and the Raman frequencies. An attempt was made to fit the low lying optical branches observed in the coherent neutron scattering experiments,⁽⁴⁾ but it was found impossible to obtain a fit for the observed values. There is some doubt in our mind about the real significance of the observed branches and any further modification of the model should in our opinion wait for an additional confirmation of the existence of the low lying branches. All together the calculation is a considerable improvement over the previous ones, as is shown by the agreement with the elastic constants and Raman frequencies.

The frequency spectrum was calculated and then the standard technique⁽⁴¹⁾ was used to derive the scattering law in the incoherent approximation. The total cross section has been calculated, including a separate calculation of the coherent elastic part, and shows good agreement with the experimental data. (24)

5. NEUTRON THERMALIZATION IN URANIUM DICARBIDE, URANIUM DIOXIDE AND ZIRCONIUM HYDRIDE

5.1 INTRODUCTION

Single and double differential cross section measurements, performed at this and other laboratories, have shown that the central force lattice dynamical model for zirconium hydride is an improvement over the previously used Gaussian-plus-Debye model. Comparison of predictions by the central force model with integral data are shown below in Section 5.2. The results of this comparison warrant the inclusion of the central force model scattering law in the ENDF.

In a different area, the study of the cross sections of possible fuel materials has been pursued both experimentally and theoretically. Experimentally, the total cross section of uranium dicarbide has been measured up to an energy of 10 eV. Theoretically, a lattice dynamical shell model calculation has been used to compute the frequency spectrum of uranium dioxide and from this the scattering law. The elastic scattering has been treated in the incoherent approximation but work is now in progress to calculate the coherent elastic cross section as has been standard procedure for the hexagonal systems.

5.2 COMPARISON OF CENTRAL FORCE MODEL RESULTS WITH INTEGRAL DATA FOR ZIRCONIUM HYDRIDE

We have tested the central force lattice dynamical model for zirconium hydride, described in previous project reports⁽⁷⁾ and in a topical report,⁽⁶⁸⁾ by making further comparisons with integral data

and with the Gaussian-plus-Debye spectrum model^(68, 69) presently used in the ENDF. The theoretical calculations were done with the codes GASKET⁽⁴¹⁾ and FLANGE.⁽⁵⁾ Since the over-all characteristics, though not the detailed structure, of the frequency spectrum calculated from the central force model are similar to those of the Gaussian-plus-Debye spectrum, no striking differences were anticipated or obtained in the integral results. The small differences that were observed indicate for the most part that the central force model leads to improved agreement with integral data.

Figure 5.1 shows a typical comparison between single differential cross section data⁽⁷⁰⁾ and theoretical results. The scattering at backward angles is better represented by the central force model. In Fig. 5.2 results are compared for the average cosine of the scattering angle ($\bar{\mu}$) as a function of incident neutron energy. Neither model represents the data well above 0.1 eV, though the central force model appears to come closer to the dip in the data at about 0.17 eV. Further studies, both of the experimental data and of the numerical procedures involved in calculating $\bar{\mu}$, are needed to evaluate fully the significance of the discrepancies observed in Fig. 5.2. It is also not unlikely that at higher incident neutron energies it will be necessary to consider anharmonic effects (see Section 6.1) to give a good theoretical description of the neutron scattering.

In view of the over-all results of tests of the central force model, we have submitted to the ENDF a scattering law based on this model in place of the Gaussian-plus-Debye scattering law. The models show a significant difference in double differential results. Though at present there are no experimental double differential data precise enough to be decisive, recent results cited in Ref. 69 tend to favor the central force model. Specific heat comparisons⁽⁶⁹⁾ definitely favor central force model results. The over-all results for integral data comparisons are comparable

Figure 5. 1. Single differential scattering by zirconium hydride.

Figure 5.2. Average cosine of the scattering angle for $\text{ZrH}_{1.85}$.

in quality for both models, with differences generally favoring the central force predictions, as noted above.

5.3 TOTAL NEUTRON CROSS SECTION OF UC_2

In some reactors significant thermalization can occur in the fuel when it comprises a light element bound to the heavy fissile or fertile element, for example, UO_2 , UC , or UC_2 . In such reactors it is possible that the lattice dynamics of the fuel material has a significant effect on the thermalization. As an experimental contribution to a program to study thermalization in fuel systems we have measured the total cross section of UC_2 . The description of the measurement and the results obtained are fully described elsewhere.⁽¹⁵⁾ The measurement of the total cross section from samples of depleted UC_2 is straightforward. However, it was found difficult to obtain an accurate value of the total scattering cross section due to the relative importance of the absorption in ^{235}U and to the uncertainty in the content of this isotope. A tabulation of the value of the total cross section of UC_2 up to an energy of 10 eV is reported in Ref. 15.

5.4 SCATTERING KERNEL FOR UO_2

A shell model for the lattice dynamics of UO_2 was given by Dolling et al.⁽⁷¹⁾ The corresponding parameters and force constants were obtained by them by fitting the dispersion curves measured by coherent inelastic neutron scattering. Using this shell model (after correcting several misprints and minor errors) we obtained by the method of root sampling the amplitude weighted frequency spectra required for computing the inelastic neutron scattering cross section. The details of this calculation can be found in Ref. 72. However, for convenience, we reproduce here the general form of the secular equation used for determining the vibrational frequencies and amplitudes

$$\det |R + Z^+ CZ - (R + Z^+ CY)(\kappa + R + Y^+ CY)^{-1}(R + Y^+ CZ) - M^+ \omega^2 I| = 0. \quad (5.1)$$

All quantities in this expression, except the frequency ω , are matrices. R describes the nearest neighbor ion-ion, shell-shell and ion-shell interactions; κ is a diagonal matrix describing the interaction between core and shell of the same ion; C is a matrix describing the Coulomb interaction between ions of ($\pm$) unit charge, Z and M correspond to the ionic charges and masses, Y corresponds to the shell charges and I is the identity. Since the UO_2 lattice has cubic symmetry the elements of the matrix C , as computed by Ewald's method, take the relatively simple form given below in standard notation.

$$\begin{aligned} C_{\alpha\beta} \begin{pmatrix} q \\ ss' \end{pmatrix} = & \frac{4\pi}{\Omega} \sum_G \frac{(\vec{q} + \vec{G})_\alpha (\vec{q} + \vec{G})_\beta}{|\vec{q} + \vec{G}|^2} \exp \left[-\frac{|\vec{q} + \vec{G}|^2}{4\eta^2} + i \vec{G} \cdot \vec{X} \begin{pmatrix} 0 \\ ss' \end{pmatrix} \right] \\ & - \sum_l e^{-i\vec{q} \cdot \vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}} \left\{ \vec{X}_\alpha \begin{pmatrix} l \\ ss' \end{pmatrix} \vec{X}_\beta \begin{pmatrix} l \\ ss' \end{pmatrix} \left[\frac{4\eta^3 e^{-|\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|^2 \eta^2}}{\sqrt{\pi} |\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|^2} \right. \right. \\ & + \left. \frac{6\eta e^{-|\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|^2 \eta^2}}{\sqrt{\pi} |\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|^4} + \frac{3 \operatorname{erfc}(\eta |\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|)}{|\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|^5} \right] \\ & - \delta_{\alpha\beta} \left[\frac{2\eta e^{-|\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|^2 \eta^2}}{\sqrt{\pi} |\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|^2} + \frac{\operatorname{erfc}(\eta |\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|)}{|\vec{X} \begin{pmatrix} l \\ ss' \end{pmatrix}|^3} \right] \Bigg\} \\ & + \delta_{ss'} \delta_{\alpha\beta} \frac{4\eta^3}{3\sqrt{\pi}}. \end{aligned} \quad (5.2)$$

Having obtained the weighted frequency spectra, we computed the inelastic scattering cross section for UO_2 in the incoherent approximation using the code GASKET.⁽⁴¹⁾

Since both O and U are coherent scatterers, the elastic scattering from UO_2 should be calculated by evaluating the Legendre moments of the coherent elastic angular cross section. This, however, has not yet been done, since the only code presently available for coherent elastic scattering (HEXSCAT)⁽⁷³⁾ is restricted to the case of lattices with hexagonal symmetry. Hence, while a new code, QSCAT, for coherent elastic scattering by cubic crystals is being written, the elastic scattering for UO_2 has been computed in the incoherent approximation. The corresponding Legendre moments, together with the inelastic scattering law have been sent to Brookhaven to be stored on the ENDF system. However, as soon as the QSCAT becomes operational the elastic scattering data will be re-evaluated.

6. MISCELLANEOUS THEORETICAL STUDIES

6.1 PHONON FREQUENCY SPECTRUM TEMPERATURE DEPENDENCE

The neutron scattering properties of reactor moderators often need to be known over a wide range of temperatures. However, the scattering laws developed for the ENDF are usually based on a limited number of neutron measurements at very few temperatures, or even a single temperature. From a theoretical model a phonon frequency spectrum is then obtained, with parameters chosen to fit the available data, and from this spectrum the scattering law at various temperatures is calculated. This procedure is consistent within the framework of the harmonic approximation, in which the normal mode frequencies are independent of temperature. However, when anharmonic terms are introduced into the expansion for the potential energy of the lattice, the frequency spectrum becomes temperature dependent.

It is difficult to say precisely how important this temperature dependence is, since no extensive theoretical or experimental studies of the problem appear to have been made. Nevertheless, anharmonic terms clearly play an important role in the lattice dynamical description of solids, since anharmonic phenomena like thermal expansion – which does not occur in the harmonic approximation – are among the prominent characteristics of solids. ⁽⁷⁴⁾

It is possible to relate theoretically the phonon frequency shifts to the anharmonicity required to explain the observed thermal expansion. However, the explicit formulation of this problem has been done only for primitive lattices. ⁽⁷⁵⁾ Moreover, a proper description

invariant under rigid-body rotations of the crystal was not carried out. Nevertheless, if we ignore these defects and limitations for the moment and, for want of something better, apply the equations in Ref. 75 to a moderator like zirconium hydride, we find that between 300°K and 800°K the normal mode frequencies decrease by twenty to thirty percent. This result indicates that anharmonic effects may well have a significant effect on the frequency spectra of important reactor moderators. (Difficulties observed previously⁽³⁹⁾ in the fittings of zirconium hydride total cross sections and neutron spectra have also suggested the possibility that a pure harmonic model is inadequate for the treatment of this moderator.)

In view of the potential significance to ENDF users of sizeable shifts of frequency distributions with temperature, we have begun the theoretical formulation of this problem for a nonprimitive lattice, using a formalism which is manifestly invariant to rigid-body rotations of the crystal. The initial steps in the development of this formalism are described in detail in the following discussion. The first objective is to obtain an explicit description of a distorted crystal by introducing into the expansion for the potential energy a set of parameters describing the relationship of a distorted lattice to an undistorted or reference configuration.

Let us consider a crystal consisting of N unit cells in which $X_{\alpha}^{(\ell)}(k)$ denotes the α component of a vector specifying the position of the kth atom in the ℓ th unit cell. The index ℓ stands for a triplet of integers, (ℓ_x, ℓ_y, ℓ_z) , such that the corners of the ℓ th unit cell are at a point $\vec{X}(\ell)$ where

$$\vec{X}(\ell) = \ell_x \vec{a}_1 + \ell_y \vec{a}_2 + \ell_z \vec{a}_3, \quad (6.1)$$

the $\vec{a}$'s being the basis vectors defining the lattice structure.⁽⁴⁾ Then

$$\vec{X}(\ell)_k = \vec{X}(\ell) + \vec{X}(k) , \quad (6.2)$$

where $\vec{X}(k)$ is one of a set of r vectors, r being the number of atoms per unit cell. In the following, we shall refer to the lattice specified by $\vec{X}(\ell)_k$ as the undistorted lattice.

We now expand the potential energy of the crystal in powers of displacements of the atoms from the undistorted lattice positions

$$\Phi = \sum_{n=0}^{\infty} \frac{1}{n!} \sum_{\substack{\ell_1 \dots \ell_n \\ k_1 \dots k_n \\ \alpha_1 \dots \alpha_n}} \tilde{\Phi}_{\alpha_1 \dots \alpha_n} \binom{\ell_1 \dots \ell_n}{k_1 \dots k_n} \prod_{i=1}^n \epsilon_{\alpha_i} \binom{\ell_i}{k_i} . \quad (6.3)$$

In Eq. 6.3 $\epsilon_{\alpha} \binom{\ell}{k}$ denotes the α -component of the displacement of atom $\binom{\ell}{k}$. The summation over $\ell_1 \dots \ell_n$ stands for a summation from 1 to $N^{1/3}$ of each integer in the triplet denoted by each ℓ_i . The k 's are summed from 1 to r and the α 's are summed from 1 to 3. The coupling parameters (CP), $\tilde{\Phi}_{\alpha_1 \dots \alpha_n} \binom{\ell_1 \dots \ell_n}{k_1 \dots k_n}$, are defined as follows:

$$\tilde{\Phi}_{\alpha_1 \dots \alpha_n} \binom{\ell_1 \dots \ell_n}{k_1 \dots k_n} \equiv \left\{ \frac{\partial^n \Phi}{\partial \epsilon_{\alpha_1} \binom{\ell_1}{k_1} \dots \partial \epsilon_{\alpha_n} \binom{\ell_n}{k_n}} \right\}_0 , \quad (6.4)$$

where the bracket and subscript indicates that the partial derivatives are to be evaluated at $\epsilon_{\alpha} \binom{\ell}{k} = 0$. For $n = 0$ we write the CP as $\tilde{\Phi}_0$. If the undistorted lattice represents a configuration in which the potential energy is a minimum, we have

$$\tilde{\Phi}_{\alpha}^{(\ell)}(k) = 0 . \quad (6.5)$$

However, we shall not restrict our formal development by imposing this restriction.

Next, we consider the displacement $e_{\alpha}^{(\ell)}(k)$ to consist of two parts, as follows:

$$e_{\alpha}^{(\ell)}(k) = \epsilon_{\alpha\beta} X_{\beta}^{(\ell)}(k) + d_{\alpha}^{(\ell)}(k) . \quad (6.6)$$

where in Eq. 6.6 and the following we adopt the convention that a repeated Greek index is summed from 1 to 3. The first term of Eq. 6.6 represents a homogeneous deformation of the undistorted lattice, the $\epsilon_{\alpha\beta}$'s being constants describing the deformation. The second term represents a further deformation which we shall later divide into a static and dynamic part

$$d_{\alpha}^{(\ell)}(k) = d'_{\alpha}^{(\ell)}(k) + u_{\alpha}^{(\ell)}(k) , \quad (6.7)$$

where $d'_{\alpha}^{(\ell)}(k)$ on the RHS of Eq. 6.7 represents a static deformation and is assumed to be expressible in the form

$$d'_{\alpha}^{(\ell)}(k) = d'_{\alpha}(\ell) + d'_{\alpha}(k) . \quad (6.8)$$

We require Eq. 6.8 to satisfy the condition that the static array of atoms at the positions

$$X'_{\alpha}^{(\ell)}(k) = X_{\alpha}^{(\ell)}(k) + \epsilon_{\alpha\beta} X_{\beta}^{(\ell)}(k) + d'_{\alpha}^{(\ell)}(k) \quad (6.9)$$

have the periodicity property necessary for being a distorted lattice.

We could now substitute Eqs. 6.6 and 6.7 into Eq. 6.8 and obtain a formal expansion for Φ in powers of the dynamic displacement, $u_{\alpha}^{(\ell)}(k)$, from the distorted lattice positions, with expansion coefficients given as functions of $\epsilon_{\alpha\beta}, d_{\alpha}^{(\ell)}(k)$, and the CP's for the undistorted configuration. Such an expansion is not particularly useful, however, because the physically-required invariance of the potential energy to rigid-body rotations of the crystal is not manifest. In other words, the homogeneous deformation described by the first term in Eq. 6.6 includes rigid body rotations (for example, $\epsilon_{\alpha\beta}$ antisymmetric and infinitesimal) which contribute formally to the expansion for Φ , though in fact this contribution is physically required to vanish.

To recast the expression for Φ in a form independent of crystal orientation it is necessary to introduce the symmetric strain parameters, $\{\eta_{\alpha\beta}\}$, defined as follows:

$$\eta_{\alpha\beta} = \frac{1}{2} (\epsilon_{\alpha\beta} + \epsilon_{\beta\alpha} + \epsilon_{\gamma\alpha} \epsilon_{\gamma\beta}) \quad (6.10)$$

In a lattice subjected to an external strain such that the lattice points are displaced by an amount given by the first term on the right side of Eq. 6.6, the separation between any two lattice points is determined by the parameters $\{\eta_{\alpha\beta}\}$.⁽⁷⁶⁾ We observe that a rigid rotation of the crystal, which of course does not change lattice point separation, is described by a set of parameters $\{\epsilon_{\alpha\beta}\}$ such that $\{\delta_{\alpha\beta} + \epsilon_{\alpha\beta}\}$ are the components of an orthogonal matrix, i. e.,

$$(\delta_{\alpha\beta'} + \epsilon_{\beta'\alpha})(\epsilon_{\beta'\beta} + \epsilon_{\beta\beta}) = \delta_{\alpha\beta}.$$

so that, using Eq. 6.10, $\eta_{\alpha\beta} = 0$. Thus, a rigid rotation is clearly equivalent to vanishing strain in a formalism expressed in terms of $\{\eta_{\alpha\beta}\}$.

Since the static and dynamic displacements represented respectively by $d_{\alpha}^{(\ell)}(k)$ and $u_{\alpha}^{(\ell)}(k)$ in Eq. 6.7 are referred to a particular orientation of the crystal the orientation-independent representation of a general strain requires in conjunction with $\{\eta_{\alpha\beta}\}$ the use of the parameters $\bar{d}_{\alpha}^{(\ell)}(k)$ and $\bar{u}_{\alpha}^{(\ell)}(k)$, given as follows: ⁽⁷⁶⁾

$$\bar{d}_{\alpha}^{(\ell)}(k) = d_{\alpha}^{(\ell)}(k) + \epsilon_{\beta\alpha} d_{\beta}^{(\ell)}(k) \quad (6.11a)$$

$$\bar{u}_{\alpha}^{(\ell)}(k) = u_{\alpha}^{(\ell)}(k) + \epsilon_{\beta\alpha} u_{\beta}^{(\ell)}(k) \quad (6.11b)$$

Our goal now is to obtain a formal expansion for Φ in powers of $\bar{u}_{\alpha}^{(\ell)}(k)$. The coefficients in this expansion will themselves be given in terms of expansions in powers of the distortion parameters, $\eta_{\alpha\beta}$ and $\bar{d}_{\alpha}^{(\ell)}(k)$, with coefficients which can be related to the CP's, $\mathfrak{P}_{\alpha_1 \dots \alpha_n}^{(\ell \dots)}(k \dots)$, and positions $X_{\alpha}^{(\ell)}(k)$ in the undistorted configuration. For generality and clarity we shall work with complete formal expansions like Eq. 6.3. Moreover, since the displacements in a homogeneous strain (first term of Eq. 6.6) become large as the distance from the origin increases, it is by no means clear at first that it would be legitimate to work with only the first few terms of an expansion like Eq. 6.3 when Eq. 6.6 is substituted. It turns out, however, that conditions on the CP's imposed by translation and rotation invariance ⁽⁷⁴⁾ make the expansions possible and remove the apparent origin-dependence.

Using Eq. 6.7 in Eq. 6.3 we can write

$$\Phi = \sum_{n=0}^{\infty} \frac{1}{n!} \sum_{\substack{\ell_1 \dots \ell_n \\ k_1 \dots k_n}} \mathfrak{P}_{\alpha_1 \dots \alpha_n}^{(\ell_1 \dots \ell_n)}(k_1 \dots k_n) \prod_{i=1}^n \left[\epsilon_{\alpha_i \beta} X_{\beta}^{(\ell_i)}(k_i) + d_{\alpha_i}^{(\ell_i)}(k_i) \right] \quad (6.12a)$$

$$\begin{aligned}
&= \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \frac{1}{m! n!} \sum_{\substack{\ell_{n+1} \dots \ell_{n+m} \\ k_{n+1} \dots k_{n+m}}} \left\{ \frac{\partial^{n+m} \Phi}{\partial \eta_{\alpha_1 \beta_1} \dots \partial \eta_{\alpha_n \beta_n} \partial \bar{d}_{\alpha_{n+1}} \binom{\ell_{n+1}}{k_{n+1}} \dots \partial \bar{d}_{\alpha_{n+m}} \binom{\ell_{n+m}}{k_{n+m}}} \right\}_0 \\
&\cdot \eta_{\alpha_1 \beta_1} \dots \eta_{\alpha_n \beta_n} \bar{d}_{\alpha_{n+1}} \binom{\ell_{n+1}}{k_{n+1}} \dots \bar{d}_{\alpha_{n+m}} \binom{\ell_{n+m}}{k_{n+m}}, \quad (6.12b)
\end{aligned}$$

where Eq. 6.12b represents a formal expansion of Φ in terms of the distortion parameters. To determine the formal coefficients in this expansion in terms of the Φ 's and $X_{\alpha} \binom{\ell}{k}$'s, we substitute for $\eta_{\alpha\beta}$ and $\bar{d}_{\alpha} \binom{\ell}{k}$ the definitions Eqs. 6.10 and 6.11 and rewrite Eq. 6.12b as an expansion in powers of $\epsilon_{\alpha\beta}$ and $d_{\alpha} \binom{\ell}{k}$. Then since the $\epsilon_{\alpha\beta}$'s and $d_{\alpha} \binom{\ell}{k}$'s are arbitrary, we can equate like powers in Eq. 6.12a and Eq. 6.12b, thereby obtaining the desired expressions for the formal coefficients.

Our next goal is to obtain lowest order values for $\eta_{\alpha\beta}$ and $\bar{d}_{\alpha} \binom{\ell}{k}$ which minimize the free energy F of the lattice. We can find an expression for F by retaining in Eq. 6.12b only terms quadratic in $\bar{u}_{\alpha} \binom{\ell}{k}$ (the "quasi-harmonic approximation").⁽⁷⁴⁾ The coefficients of these terms will then depend on the CP's for the undistorted lattice and on the distortion parameters. These coefficients then play the role of CP's with respect to the distorted lattice, and normal mode frequencies can be obtained in terms of them. These frequencies will now depend on the distortion parameters and will introduce this dependence into F when they are substituted into the expression for F in terms of normal mode frequencies.⁽⁷⁴⁾

We are carrying out the mathematical development described in the above paragraph, in addition to deriving the necessary explicit expressions for the formal coefficients in Eq. 6.12b. Results of this work will be reported during the coming contract year.

6.2 COMPUTATION OF THE SLOWING-DOWN SOURCES

The distributed slowing-down sources for the calculation of spatially dependent thermal neutron spectra have been computed in the past by the code DSZ.⁽²⁾ This code was written to provide P_0 and P_1 sources in a format suitable for the S_n transport theory code GAPLSN.⁽⁴⁴⁾ This code has now been replaced by the FORTRAN code 1DF⁽⁴³⁾ which requires a different input format. In addition, DSZ was originally written only for a one-region system. Since DSZ was not very well described in Ref. 2, we have rewritten the code during this year and documented it properly.⁽²⁰⁾

The following brief specifications apply to the revised DSZ code:

1. P_0 and P_1 slowing-down sources are calculated and either punched out or written on tape in a format suitable for 1DF.
2. The sources are computed for multi-region systems (up to 10 regions).
3. The flux energy behavior used in calculating these sources is assumed to have an E^{-k} behavior. The exponent k is obtained from one of the following procedures:
 - a. A fit is made to the observed flux in each region, or
 - b. k is read in, or
 - c. k is determined from Honeck's formulation.⁽²⁾
4. The sources are integrated over the broad energy groups. This is an improved procedure over the earlier version of the code, which merely evaluated the source term at the energy group midpoint and multiplied it by the group width. In addition, this new procedure corrects the previous overestimate of the contributions to the source terms in the lowest energy group possible for each nuclide.
5. The free gas treatment for hydrogen is retained.

6. The input guessed flux spectrum is now required at the energy group boundaries instead of the midpoints.

The following assumptions are made in the formulation of DSZ:

1. Diffusion theory applied at energies above the cutoff energy E^* .
2. The flux is separable in space and energy.
3. The spatial distribution is equal to the indium foil activities.
4. The nuclides present have constant scattering and zero absorption cross sections above energy E^* .

The treatment of the sources is reasonable if restrictions 1 through 4 above are maintained. It is worth noting that conditions are commonly encountered where these restrictions are violated. Diffusion theory is inadequate when strong flux gradients occur, such as at an external boundary. The P_1 source would generally be underestimated there with this treatment. Chemical binding effects can extend to energies much higher than commonly used cutoff values. An ideal minimum cutoff energy E^* would be 5 eV. However, it is not always practical or feasible to generate a bound atom scattering kernel up to that energy, (a free gas tack-on from a lower energy is usually made). In some cases it is necessary to start the thermal calculation at cutoff energies of the order of 1 eV, in which case the free gas treatment for the bound hydrogen atom may lead to errors of up to 5% in the P_0 source.⁽⁷⁷⁾ Finally, some systems contain resonance absorbers that give rise to substantial flux depressions. If these depressions occur just above the cutoff energy E^* , the source terms will be overestimated. This can be avoided by selecting E^* judiciously. It might be possible in the future to modify DSZ to permit the calculated or measured energy dependence of the flux above E^* to be used instead of assumed E^{-k} behavior. If this modification were made, absorption terms could also then be included.

The measured indium foil activities are often somewhat scattered. A code PREDSZ was written previously⁽⁵⁾ to smooth this measured distribution, compute its gradient, and provide punched card output suitable for DSZ. This code has been revised during this quarter to provide double, second order, five-point, least squares smoothing. In addition, the code computes the local buckling and the interval midpoints, and interpolates the measured activity data. The interval boundaries are punched out in a format suitable for 1DF. This code has now been documented with the new DSZ.

7. STUDIES IN ^{235}U AND ^{238}U

7.1 INTRODUCTION

During the previous contract year, spectrum and kinetic measurements were performed in ^{238}U and ^{235}U spheres. These measurements were essentially an extension of the techniques successfully applied at thermal neutron energies to obtain integral check of the cross sections of the materials under study. No further fast neutron studies, however, have been or are to be performed under this program. Consequently only the analysis of existing data was carried out during this year.

The analysis of the early time behavior of the excited ^{238}U sphere was found to depend strongly on the inelastic scattering of ^{238}U . This work has now been completed. The neutron spectrum measurements have been partially analyzed and the initial results reported at the Chicago ANS Meeting and the Conference on Neutron Cross Sections in Washington. Sections 7.2 and 7.3 describe these results. The comparison of measured and calculated spectra for both ^{235}U and ^{238}U indicate a sensitivity to the cross section set used. However, detailed final conclusions will depend strongly on the calibration of the intermediate energy detector used for the measurements. This calibration has been performed recently under another program and topical reports containing final conclusions on these two experiments will be released soon.

7.2 NEUTRON SPECTRA IN A ^{235}U SPHERE

During this year, an analysis of previous fast spectral measurements⁽⁷⁾ in a ^{235}U sphere was performed. These measurements were designed to check reactor calculational methods and cross section sets

for ^{235}U . Figure 7.1 shows the setup for these measurements. The data had been previously analyzed⁽⁷⁾ assuming that the background consisted primarily of a constant background of delayed neutrons and gamma rays, and a time-dependent gamma background emanating from the sphere itself. This assumption was checked by examining the raw time-of-flight data taken with the NE-213 detector (see Fig. 7.2 for example). The counts in early channels resulted from the reactor fission and capture gamma rays and could, in fact, be used to determine the dieaway time at each measurement position. Fast neutrons did not begin to arrive at the detector until about channel 140 (~ 10 MeV). The counts per channel between the disappearance of the reactor gamma rays and the arrival of the fast neutrons are roughly the same as those at large channel numbers, thus supporting the background assumption.

The reactor and pulsed source intensities were monitored by means of a sulphur foil and room return fission chambers, in addition to the normal reactor instrumentation. The high and low energy measurements were performed in two different experiments with different detectors, (NE-213 and lithium glass detectors respectively). The adequacy of the overlap of the spectral data at each measurement position indicated that the monitoring was satisfactory. This was surprising since both reactor and external neutrons were being measured and the reactivity level was changing from one position to another.

We have compared the measured data at the center (Position 3) and the surface (Position 6) with the theoretical values calculated previously⁽⁶⁾ for a similar sphere using the code GAPLSN⁽⁴⁴⁾ and the cross sections for ^{235}U evaluated by Joanou and Drake.⁽⁷⁸⁾ This comparison is shown in Fig. 7.3 with the experimental values integrated over the broad energy groups of the calculation. The intensities of the measured and calculated fluxes are in reasonable agreement. The measured spectrum, however, is considerably softer than the theory at both the surface and the center but the discrepancy is larger at the surface.

Figure 7.1. Geometry for spectral studies in a ^{235}U sphere.

Figure 7.2. Uncorrected time-of-flight data from the surface of the ^{235}U sphere.

Figure 7.3. Comparison of ²³⁵U sphere spectra with GAPLSN calculation.

Further calculations have been performed using the 1DF code⁽⁴³⁾ and ENDF/B cross sections for ^{235}U and ^{238}U . These calculations were performed to determine the effect of air scattering and room return. The results are summarized in Table 7.1. The flux at the reactor center is little affected by the room return. Changes in the spectrum at the surface are observed; however, they are small (less than 5%) until below 50 keV. The room return is clearly the cause of the long tail in the measured spectrum shown in Fig. 7.4. The differences shown in Fig. 7.3 between theory and experiment are therefore believed to be associated with either incorrect inelastic scattering cross sections or an incorrect fission source spectrum. The discrepancies are not believed to be due to the presence of higher order spatial harmonics since the shape of the neutron spectrum would then have exhibited a strong spatial dependence which was not observed. The analysis of these data is not yet complete since one important uncertainty remains. This is the uncertainty in the energy dependence of the efficiency of the lithium glass detector. This detector is being calibrated under another program. Subsequently this experiment is to be written up for publication.

7.3 NEUTRON SPECTRA IN A ^{238}U SPHERE

Spectral data were taken previously⁽⁷⁾ in a 20-inch diameter depleted uranium sphere for the purpose of providing an integral check of calculational methods and existing cross section sets for ^{238}U . These measurements were studied further during this year. Figure 7.5 illustrates the sphere and the measurement positions. The monitoring was analyzed carefully and found to be satisfactory since the data from the high energy and low energy runs overlapped in a consistent fashion at each position both in intensity as well as shape. Figure 7.6 shows the experimental data taken at various positions corresponding to a constant radius of 6.5 in. but at varying angles to the radius vector. These data

Table 7. 1
COMPARISON OF NEUTRON SPECTRA IN THE APFA
REACTOR CALCULATED WITH AND WITHOUT ROOM RETURN

Group No.	Upper Energy Boundary (eV)	Lethargy Increment (ΔU)	Calculated Scalar Fluxes per Unit Lethargy $\phi(U)$			
			Bare APFA		APFA With Concrete Walls	
			Center	Surface	Center	Surface
1	1.492+7	0.4	2.348-2	5.499-3	2.348-2	5.552-3
2	1.000+7	0.4	2.988-1	6.936-2	2.988-1	7.005-2
3	6.703+6	0.4	1.329+0	3.031-1	1.329+0	3.062-1
4	4.493+6	0.4	3.019+0	6.653-1	3.019+0	6.725-1
5	3.012+6	0.4	4.309+0	9.340-1	4.310+0	9.442-1
6	2.019+6	0.4	4.752+0	1.032+0	4.754+0	1.043+0
7	1.353+6	0.4	5.125+0	1.115+0	5.128+0	1.127+0
8	9.072+5	0.4	5.408+0	1.162+0	5.413+0	1.174+0
9	6.081+5	0.5	4.668+0	9.473-1	4.675+0	9.572-1
10	3.688+5	0.5	2.956+0	5.609-1	2.961+0	5.680-1
11	2.237+5	0.6	1.447+0	2.529-1	1.449+0	2.574-1
12	1.228+5	0.6	5.879-1	9.537-2	5.891-1	9.797-2
13	6.738+4	0.75	1.962-1	2.988-2	1.966-1	3.153-2
14	3.183+4	0.75	5.020-2	7.338-3	5.035-2	8.495-3
15	1.503+4	1.0	3.898-3	5.489-4	3.944-3	1.429-3
16	5.531+3	1.25	4.803-4	6.982-5	4.901-4	8.082-4
17	1.585+3	1.5	3.069-5	4.409-6	3.101-5	5.518-4
18	3.536+2	1.75	1.330-6	1.980-7	1.333-6	4.070-4
19	6.144+1	2.25	2.239-7	3.630-8	2.242-7	2.316-4
20	6.476+0	2.5	8.809-8	1.294-8	8.831-8	1.727-3
	5.316-1

Figure 7.4. Measured neutron spectra at the center of a ^{235}U sphere.

Figure 7.5. Experimental hole pattern in the depleted uranium sphere.

NEUTRON
FLUX X ENERGY
(RELATIVE UNITS)

Figure 7.6. Experimental angular fluxes in the ^{238}U sphere at a radius of 6.5 in.

are entirely monitor normalized and show a consistent pattern of high flux anisotropy at high energy and near isotropy at energies below 50 keV.

An analysis was performed of this experiment using the 1DF transport theory code⁽⁴³⁾ and two cross section sets, one due to Joanou and Stevens⁽⁷⁹⁾ and the other being the ENDF set.⁽⁸⁰⁾ We have integrated the measured fluxes over the broad energy group boundaries of the calculation and using a common normalization factor have compared the experimental data to the theory at Positions B, D and F. These comparisons are shown in Figs. 7.7, 7.8, and 7.9, respectively. Examination of these figures indicates that there is a greater depletion of high energy neutrons than that predicted by the theory, no matter which cross section set is used. There are important differences between theory and measurement, but these comparisons appear to show that the ENDF cross sections provide a better fit. The general trend is for the theoretical fluxes to be more anisotropic than the measured values.

In the analysis of the experiment, the source was treated as isotropic and spatially uniform over a 0.8-in. -diam volume at the center. The source spectrum was taken to be equal to the observed leakage spectrum⁽⁷⁾ from the bare target. We have modified the spectrum of the source so that the calculated and measured leakage spectrum from the bare target are in agreement. The shape of the spectrum above 1 MeV was found to be affected very slightly. On the other hand the changes to the spectrum below 1 MeV were found not to affect the computed spectra very much in the 20-in. -diam sphere at distances far from the source. Before finalizing the comparison it would be desirable to examine the effect of varying the spatial distribution of the calculated source. Also the final energy calibration of the sensitivity of the lithium glass detector should be included in the data reduction of spectra measured both from the 20-in. -diam sphere and from the bare target. At that time these data will be written up for journal publication. These comparisons already

NEUTRON
FLUX X ENERGY
(RELATIVE UNITS)

Figure 7.7. Comparison of theoretical and measured neutron spectra at Position B in the U^{238} sphere ($R = 6.5$ in., $\mu = 1.000$)

Figure 7.8. Comparison of theoretical and measured neutron spectra at Position D in the U^{238} sphere ($R = 6.5$ in., $\mu = 0.978$)

NEUTRON
FLUX X ENERGY
(RELATIVE UNITS)

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Figure 7.9. Comparison of theoretical and measured neutron spectra at Position F in the U^{238} sphere ($R = 6.5$ in., $\mu = 0.876$)

indicate that the theoretical spectra are sensitive to the choice of cross section sets and that the experimental data are of sufficient quality to specify which choice should be made.

7.4 KINETIC EXPERIMENTS IN A ^{238}U SPHERE

Detailed experimental measurements were made during the previous contract year⁽⁷⁾ of the neutron kinetics in a non-multiplying ^{238}U sphere. These studies were performed in order to obtain an integral check of the ^{238}U cross sections and calculational methods for neutron dynamics. During this year, the measurements have been analyzed and the results presented in a topical report⁽²¹⁾ and as a journal article. The theoretical analysis depends primarily on the use of a simplified semi-multigroup Monte Carlo code which gave good agreement with the early time behavior of the neutrons as observed by an ^{237}Np fission threshold detector. The experiment-theory comparison showed that kinetic behavior coupled with a suitable detector can serve as a good integral check of the inelastic scattering cross section of the material comprising the system under study. These results and others appeared in the joint APS-ANS 1968 neutron cross section and technology conference.⁽¹⁸⁾

8. DATA DISSEMINATION TO THE ENDF AND OTHER USERS

This year several new scattering laws have been submitted to the National Neutron Cross Section Center at BNL for inclusion in the ENDF/A. Also a tentative compilation of the scattering reference book has been prepared, as described below in Section 8.3. Some additions to the spectrum book are mentioned in Section 8.4.

8.1 ENDF CONTRIBUTIONS

8.1.1 Benzene

The scattering law data for the C_6H_6 molecule were calculated with GASKET⁽⁴¹⁾ for the following temperatures: 296, 350, 400, 450, 500, 600, 800, and 1000°K. The amplitude-weighted frequency spectra used for the hydrogen and carbon atoms were obtained from a normal mode analysis.⁽⁶¹⁾ An 80 x 80 α and β mesh was used to cover the energy range up to 2.0 eV.

8.1.2 Zirconium Hydride

New scattering law data have been generated separately for Zr and H. The weighted frequency spectra calculated with the central force lattice dynamical model^(27, 68) discussed in Section 5.2 was used in the code GASKET⁽⁴¹⁾ to compute the incoherent elastic data. The code ZREND⁽⁸¹⁾ was used to compute the incoherent elastic data. The temperatures were: 296, 400, 500, 600, 700, 800, 1000, and 1200°K. The hydrogen data are given in a 40 x 150 α and β mesh. For zirconium, a 40 x 118 α and β mesh was used. The energy range covered extends up to 1.0 eV for both elements.

8.1.3 Uranium Dioxide

The scattering law data for the UO_2 molecule were calculated with the codes GASKET⁽⁴¹⁾ and ZREND⁽⁸¹⁾ for temperatures of 296, 400, 500, 600, 700, 800, 1000, and 1200°K. The weighted frequency spectra of the uranium and oxygen atoms were obtained from a best fit lattice dynamical shell model described in Section 5.4. It should be noted here that only data for incoherent scattering was sent to ENDF. The calculation of the coherent elastic data falls within the work scope of the next contract year and the improved data will be sent then. A $50 \times 80 \alpha$ and β mesh covering an energy range up to 1.0 eV was used.

8.1.4 Beryllium Oxide

The code GASKET was used to compute the incoherent inelastic scattering law for a BeO molecule. Weighted frequency spectra for beryllium and oxygen atoms were obtained from a lattice dynamical shell model calculation after fitting the frequencies to elastic, Raman, and neutron scattering experiments. The code HEXSCAT⁽⁷³⁾ computed the coherent elastic data. This improved scattering law was generated for temperatures of 296, 400, 500, 600, 700, 800, 1000 and 1200°K. A $50 \times 80 \alpha$ and β mesh was used to cover the energy range up to 1.0 eV.

8.2 KERNEL BOOK

A tentative version of the ENDF Scattering Reference Book has been compiled. It describes reference information on the data submitted by Gulf General Atomic to the Evaluated Nuclear Data File (ENDF). The data submitted to ENDF include thermal inelastic incoherent scattering laws and Legendre moments through P5 of the elastic incoherent and coherent scattering by various moderators. The report describes the data available for each moderator, and the model and codes that have been utilized to compute these data, and includes some relevant examples.

The data for each moderator are included in a loose leaf format similar to the Gulf General Atomic Spectrum Book. This permits the book to be easily revised and kept up to date. For each moderator there is a description of the model with enough physical content to satisfy the casual user plus all pertinent references. An illustration of the frequency spectrum used is included. Following the physics description is a listing of the card input to all the codes used to generate the ENDF data for that particular moderator. This should permit anyone to regenerate the data or modify model parameters relatively easily. Following this listing are typical plots of the scattering law and as total scattering cross section and average cosine of such the scattering angle. There will also be some comments describing the accuracy of the data. The moderators included so far are the following: H_2O , polyethylene, D_2O , graphite, beryllium, zirconium hydride, and BeO . The formal version of the Kernel Book will be published as Gulf General Atomic Report GA-8774.

8.3 SPECTRUM BOOK

The reanalysis of spectra measurements across a TRIGA fuel element surrounded by poisoned D_2O (see Section 2.4) was completed during this past year and the nine spectra were distributed to Spectrum Book holders.

There still exists an extensive backlog of neutron spectra to be included in the Spectrum Book. Work is in progress on analyzing or reanalyzing the data.

9. IMPROVEMENT IN LINAC AND EXPERIMENTAL CAPABILITIES

9.1 LINAC FACILITY IMPROVEMENTS

During this year construction of the low level cell adjacent to the single section machine has been completed. A 90° bending magnet has been installed to transport the beam into this cell. The program that was begun three years ago to build new high power modulators, upgrade and relocate others, has been essentially completed. The multiple switching tubes in each modulator have been replaced by a single more reliable high power tube. One modulator remains inside the LINAC as a spare. An improved regulating system for the modulators will be constructed to provide better pulse stability. Other improvements to the LINAC this year include the upgrading of the magnet controls and the interlock systems.

Next year the three-section machine will be improved by the completion of a high power injector system that will be compatible with the system in the single stage machine. A solid state CW oscillator is also to be installed which will provide an improved RF source and hence better energy resolution. In addition, the two machines will be synchronized by the construction of a master trigger unit. This will reduce interference effects caused by separate experiments. In addition, the trigger unit will provide greater versatility and reduced time jitter.

The accelerator is being used more frequently in the four-stage configuration. Typical conditions achieved for many studies utilizing the 220-meter flight path now include a pulse 50-nsec wide of 7 amps current with an energy of 54 MeV at 180 pps. The corresponding instantaneous

neutron yield rate is 1.7×10^{18} neutrons/sec, which is considerably higher than what we could regularly attain one year ago.

The AEC-owned computer supplied under this program as the core of a data acquisition system has received more use under this and other AEC programs. It has greatly enhanced our ability to record data with more speed and flexibility and to utilize the improved capabilities of the LINAC. The present system is described in Section 9.3.

9.2 INTERMEDIATE ENERGY NEUTRON DETECTOR

An intermediate energy neutron detector (IED) has been designed and constructed at Gulf General Atomic for the purpose of measuring neutron spectra in the intermediate energy range. A complete report, including the details of the design and operation of this detector, has been published. ⁽⁸²⁾

The motivation for designing a new detector came from the need to measure intermediate energy neutron spectra from fast multiplying assemblies. A detector was required which had reasonable timing resolution (30 nsec), high sensitivity to neutrons, and low sensitivity to gamma rays. A way of meeting these conditions was to surround a large conversion (n, γ) plate with an organic fluor capable of absorbing a significant fraction of the energy of the resulting gamma rays. By accepting counts which result in the deposition of gamma-ray energies greater than 2 or 3 MeV and by requiring coincidences between adjacent halves of the fluor, rejection of background gammas would be easily achieved.

Conversion plates of several materials and configurations were tried, and in addition the IED was used to study the spectra in a 20-in. - diam ^{238}U sphere. However, these studies revealed that neutrons, both fission and scattered neutrons, were being thermalized in the organic fluor and being reabsorbed at a later time in the conversion plate. These faults were reduced by the introduction of B_4C shielding and a nonfissioning

conversion plate. The new conversion plate contains a mixture of europium oxide, terbium oxide, and tantalum oxide in the isotopic ratio Eu: Ta of 3:1:1. This mixture was designed to give a fairly smooth absorption probability in the intermediate energy range on the basis of the Garrison⁽⁸³⁾ data.

The IED with the improved conversion plate and shielding has been found to have an excellent discrimination against background compared to the NE-213, B₄C and lithium glass detectors. However, the sensitivity was not as high as anticipated.

It proved necessary for the total energy absorbed to be greater than 2.2 MeV, which is the gamma-ray energy from absorption of a thermal neutron by hydrogen. An improved efficiency seemed obtainable by demanding a 2.4-MeV bias on the sum of the output from the two halves instead of a 1.2-MeV bias on each half. A threefold coincidence requirement permitted the bias between the two halves of the detector to be set well below the 1.2-MeV level which had been previously necessary to eliminate scattered neutrons. The change allowed us to count when the total gamma-ray energy deposited in the detector exceeded 2.2 MeV, even though as little as ~0.5 MeV may have been deposited in one half. Though these modifications increased the efficiency by about a factor of four, the originally anticipated efficiency was still not achieved. Hence we redesigned the shape of the central absorber, which was originally three inches thick and five inches in diameter. The new sample is made in two pieces, each 2 inches thick and 7.5 inches in diameter. The powdered Eu-Tb-Ta mixture was packed to a less dense consistency in this configuration to facilitate escape of the capture gamma rays from the sample for detection. Separation of a two-piece sample permits the gamma rays to escape even more readily and allows insertion of various materials between the samples, such as borated polyethylene, etc. The efficiency of the IED was determined from about 20 MeV to about 100 eV

by calibrating it against other detectors at Gulf General Atomic. This efficiency as a function of neutron energy is given in Fig. 9.1. After calibration the detector was used to measure the neutron spectrum from the three-inch-diameter water-cooled uranium target in use at the LINAC. These data compare satisfactorily with data taken previously with other detectors.

9.3 ON-LINE COMPUTER

During the past year usage of the CDC-1700 on-line acquisition computer⁽⁷⁾ has increased considerably; several new data collection programs have been written and used; interfacing for the two-parameter bin has been completed and two-parameter data are now being taken; and additions to the computer are being planned.

New and modified programs added during this year include: (1) an arbitrary time boundary (corresponding to either constant ΔE or ΔU) program with two banks of 1024 channels; (2) a core-resident two-parameter program which accumulates 512-channel spectra of one parameter for five selected intervals of another; (3) a two-parameter program which accumulates two in-core spectra for live display while writing a filtered vision of all the data taken on the disk for later analysis; and (4) a two-parameter analysis program which plays back selected data from the disk to form pulse height spectra for arbitrary time intervals. The amount of data to be printed or punched in two-parameter analysis makes the use of the 1700 computer extremely time consuming, because of its very slow typing and punching. The interim solution has been to copy the contents of the disk onto magnetic tape, which can be read by the UNIVAC 1108, at the CDC Miramar Road plant. A recently acquired magnetic tape drive has been installed on the 1700 computer to handle this disk-to-tape conversion.

Figure 9.1. Efficiency of the new rare earth neutron detector.

Control Data Corporation has been unable to deliver the mass storage operating system in a form that will run with our small memory (12,000 words). In order to get the promised system operating, CDC has extended its current loan of 4,000 words of memory for another year, and an additional 4,000 words will be bought. The resultant 16,000-word machine will be a considerably more powerful tool for both in-core and mass storage data acquisition and analysis.

A Houston Scientific Omnigraphic incremental plotter has been purchased and is being interfaced to the computer. All new interfacing is being designed around a general purpose input-output driver interface, which will allow us to add other equipment fairly inexpensively (such as a card reader and line printer).

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