Gulf General Atomic Incorporated

P.O. Box 608, San Diego, California 92112

AEC RESEARCH AND
DEVELOPMENT REPORT

GA-9753
UC-2, General
Miscellaneous, and
Progress Reports

INTEGRAL NEUTRON THERMALIZATION

ANNUAL SUMMARY REPORT

OCTOBER 1, 1968 THROUGH SEPTEMBER 30, 1969

Work done by:

G. M. Borgonovi
J. E. Cunningham
D. H. Houston
J. U. Koppel
J. M. Neill
E. L. Slaggie
D. Sprevak

Report written by:

G. M. Borgonovi
D. H. Houston
J. M. Neill
E. L. Slaggie

Prepared under
Contract AT(04-3)-167
Project Agreement No. 2
for the

San Francisco Operations Office
U. S. Atomic Energy Commission

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:
A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.
As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Gulf General Atomic Project 48.01

October 30, 1969

This document is

PUBLICLY RELEASABLE

Larry E. Williams
Authorizing Official

Date: 05/26/2006

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED
phil

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

ANNUAL REPORTS ON THIS PROGRAM

Annual Summary Report, October 1, 1960 through
September 30, 1961, USAEC Report GA-2544 (1961).

Annual Summary Report, October 1, 1961 through
September 30, 1962, USAEC Report GA-3542 (1962).

Annual Summary Report, October 1, 1962 through
September 30, 1963, USAEC Report GA-4059 (1963).

Annual Summary Report, October 1, 1963 through
September 30, 1964, USAEC Report GA-5798 (1964).

Annual Summary Report, October 1, 1964 through
September 30, 1965, USAEC Report GA-6824 (1965).

Annual Summary Report, October 1, 1965 through
September 30, 1966, USAEC Report GA-7480 (1966).

Annual Summary Report, October 1, 1966 through
September 30, 1967, USAEC Report GA-8280 (1967).

Annual Summary Report, October 1, 1967 through
September 30, 1968, USAEC Report GA-9036 (1968).

CONTENTS

	<u>Page</u>
1. INTRODUCTION AND SUMMARY	1
2. STUDIES IN COHERENT NEUTRON SCATTERING BY BERYLLIUM.	4
2.1 Angular Cross Section of a Beryllium Single Crystal.	4
2.2 Coherent Scattering Law of Polycrystalline Beryllium.	10
2.3 Approximate Calculation of the Coherent Scattering Law in Polycrystalline Beryllium	10
3. CENTRAL FORCE MODEL FOR URANIUM CARBIDE	20
4. COHERENT ELASTIC NEUTRON SCATTERING BY UO_2 AND UC	22
5. TEMPERATURE DEPENDENCE OF PHONON FREQUENCY DISTRIBUTIONS	28
6. CRYSTAL BINDING EFFECTS ON DOPPLER BROADENING OF NEUTRON ABSORPTION RESONANCES	40
6.1 Introduction	40
6.2 Formulas for Doppler Broadening	41
6.3 Numerical Results	47
6.4 Conclusions	55
7. MISCELLANEOUS STUDIES.	57
7.1 Total Cross Section of Zirconium Hydride	57
7.2 Analysis of Spectral Measurements	61
8. ADDITION OF THE SHORT COLLISION APPROXIMATION TO THE FLANGE CODE	65
9. ENDF ACTIVITIES.	74
9.1 UC, UO_2 and CH_2 Kernels	74
9.2 Miscellaneous Activities	75
REFERENCES	76

ILLUSTRATIONS

<u>Figure</u>		<u>Page</u>
2. 1	Basal plane of reciprocal lattice in beryllium	6
2. 2	Theoretical and experimental angular cross section of beryllium single crystal	9
2. 3	Total cross section of beryllium below the Bragg cutoff .	11
2. 4	Average dispersion relations for beryllium	15
2. 5	Calculated scattering laws for beryllium at $\beta = 1.75$ . .	17
2. 6	Calculated scattering laws for beryllium at $\beta = 2.25$ . .	18
2. 7	Calculations of the total inelastic cross section of beryllium	19
5. 1	Phonon frequency distributions for ZrH_2 at 300°K and 800°K	32
5. 2	Frequency distributions for parallel vibrations of the graphite lattice at room temperature and at 1300°K . .	38
6. 1	Weighted frequency spectrum for U in UO_2	49
6. 2	Normal mode frequency spectrum for UO_2	50
6. 3	Doppler broadened line shape for 6.65 eV resonance of ^{238}U at $T = 296^\circ\text{K}$	52
6. 4	Effective temperature as a function of temperature for U in UO_2	53
6. 5	Doppler broadened line shape for 6.65 eV resonance of ^{238}U at $T = 50^\circ\text{K}$	54
7. 1	Total cross section of $\text{ZrH}_{1.85}$	60
7. 2	Geometry for spectral studies in the ^{235}U sphere . . .	62
7. 3	High energy neutron spectrum $N(E)$ from position 3 in APFA III.	63
8. 1	Example of double differential cross section of H_2O with and without the s. c. t. approximation	66

ILLUSTRATIONS (Continued)

<u>Figure</u>		<u>Page</u>
8.2	Example of angular single differential cross section of H_2O with and without the s. c. t. approximation	69
8.3	Example of zeroth Legendre moment of the H_2O kernel with and without the s. c. t. approximation. . . .	70
8.4	Example of first Legendre moment of the H_2O kernel with and without the s. c. t. approximation. . . .	71
8.5	Total cross section of H_2O computed with and without the s. c. t. approximation	72

1. INTRODUCTION AND SUMMARY

This annual summary report describes the work performed on the Integral Neutron Thermalization Program from October 1, 1968 through September 30, 1969, 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 given in the eight annual reports listed on page ii.

The experimental and theoretical work performed under this program in previous years has succeeded in advancing significantly the description of neutron thermalization in a wide variety of reactor materials. The remaining tasks involve improved analysis of previous work and application of previously developed techniques to improve the models on which scattering laws are based. Consequently no new experimental work was undertaken this year. New theoretical work has largely involved improving already established methods. The exception to this trend was substantial theoretical work done on the temperature dependence of phonon frequency distributions. The goal of this work, however, was to check previously developed models in which the dependence was neglected. That this neglect was in fact justified has been confirmed for graphite and for zirconium hydride, as reported in Section 5.

The calculation of coherent inelastic scattering by polycrystalline beryllium has been performed by revised methods. A result of this work was the development of a code with greatly improved capability to make coherent inelastic scattering law calculations in reasonable times. This code is discussed in Section 2.2. Its application in a test of a method formerly used to approximate the coherent inelastic scattering law is described in Section 2.3. Section 2.1 reviews the detailed analysis performed

on beryllium single-crystal single-differential cross section measurements made during the previous contract year. A central force lattice dynamical model has been developed for uranium carbide, following methods used previously for zirconium hydride. This model is discussed in Section 3. Section 4 describes the development of a code to calculate the coherent elastic scattering from cubic crystals. This code was applied to uranium carbide and to uranium dioxide, both of which have cubic symmetry. Formerly coherent elastic scattering was calculated only for hexagonal crystals.

The availability of the new lattice dynamical model for uranium carbide and a previously developed model for uranium dioxide has made possible a definitive study of Doppler broadening of neutron absorption resources. This work also depended upon the use, in a slightly modified form, of the GASKET code,⁽¹⁾ developed at Gulf General Atomic in previous years for scattering law calculations. The work on Doppler broadening, reported in Section 6, is reassuring with regard to the accuracy of approximations usually made at room temperature. Important binding effects were seen at lower temperatures, but these effects are smaller than had been anticipated on the basis of prevalent misunderstandings concerning the proper use of normal mode frequency distributions.

Miscellaneous studies reported in Section 7 include the analysis of previously taken total cross section data for zirconium hydride and spectral measurements performed previously in a ^{235}U sphere. Code modifications to FLANGE are described in Section 8. This type of work — improvements in existing codes to extend their applicability and usefulness — will be undertaken more intensively in the coming year.

The dissemination of detailed information gathered under this program is accomplished primarily by inclusion in the ENDF together with descriptive reports. Section 9 describes activities pertaining to enlarging

and updating this file and revising the Kernel Book, which contains information pertinent to the data included in the ENDF. Information on work done under this program has also been disseminated by presentations at meetings, ^(2,3) journal articles, ^(4,5) and topical reports. ⁽⁶⁻¹¹⁾

2. STUDIES IN COHERENT NEUTRON SCATTERING BY BERYLLIUM

A number of studies on the coherent scattering of neutrons by beryllium have been performed during this contract year. The single differential angular cross section of a Be single crystal has been computed for different orientations of the crystal in order to make a comparison with measurements which were made at the end of the previous contract year. The agreement between theoretical and experimental results is not very satisfactory and seems to indicate that a higher angular resolution should be used for this type of experiments. The calculation of the angular cross section is described in Section 2.1. Very satisfactory results have been obtained in the exact calculation of the coherent one-phonon scattering law of beryllium. These results are fully described in separate reports^(10, 11) and are briefly summarized in Section 2.2. The exact treatment, while giving satisfactory results, has the drawback of requiring lengthy computational time. For this reason an approximation first proposed by Egelstaff⁽¹²⁾ for the coherent scattering law has been investigated, and is described in Section 2.3.

2.1 ANGULAR CROSS SECTION OF A BERYLLIUM SINGLE CRYSTAL

The purpose of this calculation was to make a comparison with experimental data obtained from a beryllium single crystal at the end of the previous contract year. The comparison for beryllium is of interest because the scattering is practically all coherent and therefore the calculation is based on the expression for the coherent scattering cross section which in thermalization work has received less attention than the incoherent scattering cross section.

A code which computes the single differential cross section for a particular orientation of a beryllium single crystal has been written, it is based on the model used by Slutsky and Garland⁽¹³⁾ to describe the characteristic vibrational frequencies of the crystal. Figure 2.1 contains several features that are useful in the discussion of the single differential cross section. The figure shows the basal plane of the reciprocal lattice for a beryllium crystal with the contours of the Brillouin zones. The line K'K defines the direction of the incident neutrons. The triangular shaded region near the 200 reciprocal lattice point is an irreducible part of the section of the Brillouin zone. Once the frequencies and the polarization vectors are known within this area, they can be obtained in any other point of the plane by means of symmetry transformations.

A coherent one-phonon scattering process takes place when the two conditions of energy and momentum conservation are satisfied:

$$E_1 - E_0 = h\nu_j$$

$$\vec{k}_1 - \vec{k}_0 = \vec{\tau} + \vec{q} = \vec{Q}, \quad (2.1)$$

where ν_j and $\vec{q}$ are, respectively, the frequency and the wavevector of the phonon, E_0 and $\vec{k}_0$ are the energy and the wavevector of the incoming neutron, E_1 and $\vec{k}_1$ are the same quantities for the neutron after scattering, $\vec{\tau}$ is a reciprocal lattice vector, $\vec{Q}$ is the momentum transfer, h is Planck's constant, and j is the index of the branch.

When the wave vectors $\vec{k}_0$ and $\vec{k}_1$ are limited within a plane, the conditions (2.1) are satisfied along scattering lines. In the case of beryllium there will be in general twelve different scattering lines, corresponding to the six branches of the dispersion relations and to the conditions of energy gain and energy loss.

Figure 2.1. Basal plane of reciprocal lattice in beryllium.

The intensity of the one-phonon scattering process is given by the expression, ⁽¹⁴⁾

$$\left(\frac{d\sigma}{d\Omega}\right)_{j,\mu} = \frac{k_1}{k_0} \frac{h}{4v_j} \frac{\coth(hv_j/k_B T) - \mu}{\left|1 - \mu \frac{k_1 \cdot \text{grad}_{\vec{q}} v_j}{k_1 V}\right|} \frac{b^2 e^{-2W}}{M} |F(\vec{q}, \vec{\tau})|^2, \quad (2.2)$$

where k_B is the Boltzmann constant, T is the absolute temperature of the scatterer, μ is equal to -1 for energy gain processes and to $+1$ for energy loss processes, V is the velocity of the scattered neutron, b is the coherent bound scattering amplitude of beryllium, W is an isotropic Debye Waller factor, M is the mass of the beryllium atom; $F(\vec{q}, \vec{\tau})$, given by the expression,

$$F(\vec{q}, \vec{\tau}) = \sum_{\ell=1}^2 \left(\vec{Q} \cdot \vec{C}_{\ell, \vec{q}}^j \right) e^{2\pi i \vec{\tau} \cdot \vec{r}(\ell)}, \quad (2.3)$$

is the inelastic structure factor. In Eq. 2.3 $\vec{C}_{\ell, \vec{q}}^j$ is the polarization vector relative to the phonon of frequency v_j and wave vector $\vec{q}_j$, and $\vec{r}(\ell)$ is the position vector of the Be atom in the unit cell.

For each incident energy at a fixed scattering angle, we have to compute the contribution of the six different branches. The frequencies, the polarization vectors, and the components of the gradient of the frequency with respect to $\vec{q}$ were determined for a mesh in the irreducible part (1/12) of the Brillouin zone-section. This information has been calculated for a triangular mesh of 40×24 points and written on a magnetic tape. The different branches have been ordered according to increasing frequency.

At a fixed scattering angle and for a given incident wave vector the code explores a range of final wave vectors, checking the conservation conditions (2.1). Whenever these conditions are satisfied (within a preset interval ΔE) the contribution to the single differential cross section is computed using Eq. 2.2. In the hypothesis of ideal resolution, any branch

should give at maximum two contributions (energy loss and energy gain). However, because of the relaxation that has to be imposed on the delta function conditions in a numerical calculation, a branch may actually contribute more than twice. In such a case the average value of the contributions is taken.

The experimental setup which was used in obtaining the measured single differential cross sections from a beryllium single crystal did not differ substantially from the setup used for the determination of the single differential cross sections of water,⁽¹⁵⁾ benzene,⁽¹⁶⁾ and zirconium hydride,⁽¹⁷⁾ apart from the fact that a longer flight path, 1510 cm instead of 1256 cm, was used. The measurements were analyzed following a standard technique and the cross sections were put on an absolute basis by using as a reference the scattering from a small disc of polyethylene having approximately the size of the single crystal of beryllium.

Four distributions were measured at angles ψ (the angle ψ , as visible in Fig. 2.1, gives the orientation of the crystal with respect to the incident neutron beam) of 55, 60, 65, and 70 degrees.

Figure 2.2 shows the measured and calculated single differential cross sections for the orientation defined by $\psi = 70$ degrees as a function of the orientation of the incident neutrons. The parameters used in the calculation were $\Delta k_0 = 0.02 \text{ \AA}^{-1}$, $\Delta k_1 = 0.005 \text{ \AA}^{-1}$, and $\Delta E = 0.002 \text{ eV}$, where Δk_0 , Δk_1 , and ΔE denote respectively the increments in incident wave vector, the increment in final wave vector, and the energy range of acceptance.

The scattering angle Φ was 90° . In order to account for the finite angular resolution of the scattered beam, the calculated curve is actually a weighted average of calculations performed at scattering angles of 89° , 90° , and 91° . The cross section assumes large values when the orientation of the crystal is such that reciprocal lattice points fall close to the line of elastic scattering. The position of these large values of the cross section is predicted correctly by the calculation; however, the over-all

Figure 2. 2. Theoretical and experimental angular cross section of beryllium single crystal.

agreement between theory and experiment is not as satisfactory as it was in a previous measurement in MgO.⁽¹⁸⁾ The reason for this seems to be that the experimental resolution was not fine enough to pick up the sudden variations of the cross section. These sudden variations are due to the structure of beryllium, which is more complex than that of MgO, and to the complicated behavior of the polarization vectors inside the Brillouin zone.

2.2 COHERENT SCATTERING LAW OF POLYCRYSTALLINE BERYLLIUM

A code which computes the coherent one-phonon scattering law for polycrystalline beryllium has been written. The descriptions of the method of calculation and of the code itself have been written up in separate documents,^(10, 11) a combination of which is being submitted to Nuclear Science and Engineering for publication. The code is an improvement over previous codes which compute coherent cross sections,⁽¹⁹⁾ in the sense that the results obtained show a limited dependence on the size of the mesh in reciprocal space which is required for the calculation. This is essentially an exact calculation and therefore requires a long computational time. About 40 minutes were required on the CDC-6600 computer to calculate the scattering law up to $\alpha = .6$ and the time of computation increases approximately as $\alpha^{3/2}$. A significant improvement over the incoherent approximation has been obtained in the calculation of the total cross section below the Bragg cutoff. Figure 2.3 shows the total cross section below the Bragg cutoff calculated from the coherent scattering law and using the incoherent approximation.

2.3 APPROXIMATE CALCULATION OF THE COHERENT SCATTERING LAW IN POLYCRYSTALLINE BERYLLIUM

Since the exact calculation of the coherent one-phonon scattering law for beryllium is very time consuming it is interesting to explore the possibilities of approximate treatments. Egelstaff⁽¹²⁾ made an attempt to treat the coherent cross section of a polycrystal by writing the

Figure 2.3. Total cross section of beryllium below the Bragg cutoff.

coherent cross section as the product of the incoherent cross section and a structure factor which was obtained on the basis of an elastic continuum. A generalization of Egelstaff's approximation, introducing a more realistic dispersion of sound waves, was made by Kroo, et al., ⁽²⁰⁾ in analyzing the scattering of neutrons from condensed argon. Recently, deWette and Rahman⁽²¹⁾ have carried out a calculation of the coherent cross section for polycrystals of noble gases both exactly and in the Egelstaff's approximation. From the comparison of the two calculations the latter authors conclude that the approximation is not adequate. However, it seems to us that when the comparison is made at the level of coherent scattering laws which are characterized by the presence of numerous peaks and valleys, some disagreement should be expected. The question therefore should be put in a different way; namely, is the approximate coherent treatment an improvement over the incoherent approximation in the calculation of integral quantities? It was briefly mentioned in Section 2.2 that the total cross section below the Bragg cutoff calculated with the coherent code fits experimental data significantly better than the same quantity predicted by the incoherent approximation. If a similar improvement could be obtained by using the approximate coherent treatment, this treatment would have a practical value, since the time of computation required by the approximate code is about one hundred times shorter than the one required by the exact treatment.

The standard form of the scattering law $S(\alpha, \beta)$, coherent or incoherent, is defined by its relation to the double differential cross section $\frac{\partial^2 \sigma}{\partial \Omega \partial E}$ in the following way:

$$\frac{\partial^2 \sigma}{\partial \Omega \partial E} = \frac{\sigma_b}{4\pi k_B T} \sqrt{\frac{E}{E_0}} e^{-\beta/2} S(\alpha, \beta) \quad (2.4)$$

where E and E_0 are the energies of the neutrons after and before scattering and α and β are related to the squared momentum exchange $\hbar^2 \kappa^2$ and to the energy exchange $\hbar\omega$ by:

$$\alpha = \frac{\hbar^2 \kappa^2}{2Mk_B T} \quad \beta = \frac{\hbar\omega}{k_B T} \quad (2.5)$$

where M is the mass of the scattering atom.

In the incoherent approximation, the one-phonon part of the scattering law for a cubic Bravais crystal can be written:

$$S_{\text{inc}}^{(1)}(\alpha, \beta) = \frac{k_B T \kappa^2}{4M\omega \sinh \frac{\beta}{2}} e^{-2W} f(\omega) \quad (2.6)$$

Here W is an isotropic Debye-Waller factor and $f(\omega)$ is the normalized frequency distribution.

Egelstaff⁽¹²⁾ writes the one-phonon part of the coherent scattering law in the following way:

$$S_{\text{coh}}^{(1)}(\alpha, \beta) = \frac{k_B T \kappa^2}{4M\omega \sinh \frac{\beta}{2}} e^{-2W} f(\kappa, \omega) \quad (2.7)$$

where

$$f(\kappa, \omega) = f(\omega) Z(\kappa, \omega) \quad (2.8)$$

and the structure factor $Z(\kappa, \omega)$ is given by

$$Z(\kappa, \omega) = \sum_{\tau} \frac{\pi^2 F_{\tau}}{B \tau \kappa q(\omega)} \quad (2.9)$$

Here τ is the length of a reciprocal lattice vector, F_τ is the number of equivalent lattice planes associated with τ , B is the volume per atom, and, for a given ω , κ has to satisfy the inequality:

$$\tau - q(\omega) \leq \kappa \leq \tau + q(\omega) \quad (2.10)$$

It is easy to see how to apply Eq. 2.9 to an elastic continuum or to a medium with a uniquely defined dispersion relation $q(\omega)$. However, in the case of beryllium we have six anisotropic branches in the dispersion relation. As a means of generalizing Egelstaff's work to the more elaborate case, the Slutsky and Garland⁽¹³⁾ model which has been consistently applied to coherent studies in beryllium can be used to define average optical and acoustic dispersion curves. For each wave vector we have three acoustic and three optical branches. The distinction between optical and acoustic is made according to the phase difference between the largest Cartesian components of the polarization vectors. Suppose that, for a given wave vector, $\vec{C}(1)$ and $\vec{C}(2)$ are the (complex) polarization vectors for atom 1 and atom 2 in the beryllium unit cell. Suppose then that $|C_x(1)| = |C_x(2)|$ is the magnitude of the largest Cartesian component. Then, if the phase difference between the two complex numbers $C_x(1)$ and $C_x(2)$ is less than $\pi/2$, we classify the mode as acoustic, otherwise we classify it as optical. A scanning of the irreducible part of the Brillouin zone has been made and an average of the acoustic and optical frequencies has been performed. The result is the average acoustic and optical dispersion relations $q_{ac}(\omega)$ and $q_{op}(\omega)$, which are illustrated in Fig. 2.4.

The next step is a generalization of Eq. 2.9 in the form

$$Z(\kappa, \omega) = \frac{1}{2} \sum_{\tau} \Delta_{\tau, \omega}^{ac}(\kappa) \frac{\pi^2 F_\tau}{B \tau q_{ac}(\omega)} + \frac{1}{2} \sum_{\tau} \Delta_{\tau, \omega}^{op}(\kappa) \frac{\pi^2 F_\tau}{B \tau q_{op}(\omega)} \quad (2.11)$$

Figure 2.4. Average dispersion relations for beryllium.

where $\Delta_{\tau, \omega}^{\text{ac}}(\kappa)$ and $\Delta_{\tau, \omega}^{\text{op}}(\kappa)$ are defined by:

$$\Delta_{\tau, \omega}^{\text{ac}}(\kappa) = \begin{cases} 1 & \text{for } \tau - q_{\text{ac}}(\omega) \leq \kappa \leq \tau + q_{\text{ac}}(\omega) \\ 0 & \text{otherwise} \end{cases} \quad (2.12)$$

$$\Delta_{\tau, \omega}^{\text{op}}(\kappa) = \begin{cases} 1 & \text{for } \tau - q_{\text{op}}(\omega) \leq \kappa \leq \tau + q_{\text{op}}(\omega) \\ 0 & \text{otherwise} \end{cases}$$

Formulas 2.7, 2.8, 2.11 and 2.12 have been used to compute an approximate coherent scattering law. Figures 2.5 and 2.6 show two examples of scattering laws for $\beta = 1.75$ and $\beta = 2.25$. The results of the exact calculation, of the incoherent approximation, and of the generalized Egelstaff's approximation are shown.

Figure 2.7 shows the total scattering cross section below the Bragg cutoff as calculated using the approximate coherent treatment, the incoherent approximation, and the exact one-phonon coherent treatment. It appears that the generalized Egelstaff's approximation is not much improvement over the incoherent approximation even in the calculation of integral quantities. The conclusion is that, when accurate predictions are required for coherent moderators, one has to resort to the exact calculation.

Figure 2.5. Calculated scattering laws for beryllium at $\beta = 1.75$.

Figure 2.6. Calculated scattering laws for beryllium at $\beta = 2.25$.

Figure 2. 7. Calculations of the total inelastic cross section of beryllium.

3. CENTRAL FORCE MODEL FOR URANIUM CARBIDE

The potential usefulness of uranium carbide as a fuel for advanced reactors makes it desirable to develop theoretical models to describe thermal neutron scattering from this substance. Uranium monocarbide (UC) has a cubic structure of the NaCl type, with a lattice constant $a = 4.948 \pm 0.001$.⁽²²⁾ A preliminary calculation of a UC neutron scattering kernel in the incoherent approximation was made the previous year based on a Debye model for the distribution of normal mode frequencies in the lattice. However, recent neutron scattering data⁽²³⁾ have indicated clearly the importance of optical vibrations not taken into account by a pure Debye model. These vibrations can be included in a phenomenological frequency spectrum by combining an optical peak (of Gaussian shape, for example) at 0.045 eV with a low energy Debye spectrum to represent acoustic modes.⁽²³⁾ It is then necessary to assign arbitrarily the relative weighting of acoustic and optical mode contributions to the neutron scattering by the two different types of atoms in the crystal. (A discussion of the need for this weighting is included in Section 6.)

A somewhat similar situation exists for zirconium hydride, a reactor moderator for which a phenomenological Gaussian-plus-Debye frequency spectrum leads to a fairly good description of neutron scattering measurements.⁽²⁴⁾ It has been shown in the case of zirconium hydride that this type of frequency spectrum can be explained on more nearly fundamental terms by a central force lattice dynamical model,⁽²⁵⁾ from which the weightings needed for neutron scattering calculations can be obtained directly without the need for an arbitrary assignment. The agreement obtained between central force model predictions and experimental specific heat and neutron scattering data for ZrH_2 is an improvement over previous phenomenological results.

There is thus considerable incentive to construct a similar lattice dynamical model for UC. Such a calculation has been performed this year and is described at length in a topical report.⁽²⁶⁾ The calculation of the normal mode frequency distributions is similar in most respects to that for ZrH_2 ,⁽²⁵⁾ except for some reduction in complexity permitted by the simpler lattice structure of UC. However, uncertainties in specific heat data and the scarcity of neutron scattering data make it impossible at present to establish the values of the force constants for UC with the precision attainable for ZrH_2 . The calculated frequency spectrum is qualitatively in accord with the phenomenological model,⁽²³⁾ though it has much more structure. Neutron data presently available are not sufficiently precise to confirm the presence of this structure. A quantitative, but not very sensitive, test of the model has been made by comparing measured and calculated specific heat from 0°K to 360°K. The agreement is satisfactory.⁽²⁶⁾

The considerable effort and expense required to calculate coherent inelastic scattering (see Section 2.2) do not seem to be warranted by the neutron data for UC presently available for comparison. For reactor studies a thermal neutron scattering law calculated in the incoherent approximation is adequate. Such a scattering law has been obtained (see Section 9.1) by using the weighted frequency distributions for U and C as input to the code GASKET.⁽¹⁾ Coherent elastic scattering for UC has also been obtained by a code described in the following section.

4. COHERENT ELASTIC NEUTRON SCATTERING BY UO_2 AND UC

The code QSCAT has been developed to compute the coherent elastic neutron scattering cross section for a cubic lattice. The n^{th} Legendre moment of this cross section per atom for neutrons of energy E scattered by polycrystalline material is given by the expression: ⁽²⁷⁾

$$\sigma_n(E) = \frac{\hbar^2 (2\pi)^3}{4rmeV} \sum_{\vec{\tau}}^{\tau \leq \frac{4\pi}{\lambda}} \frac{P_n(\bar{\mu})}{\tau} \left| \sum_{k=1}^r a_k \exp \left[\frac{-\hbar^2 \tau^2 \gamma_k^2}{4M_k} \right] e^{-i\vec{\tau} \cdot \vec{r}_k} \right|^2 \quad (4.1)$$

where $\hbar$ is Planck's constant divided by 2π , r is the number of atoms per unit cell, m is the neutron mass and V is the volume of a unit cell; λ denotes the neutron wavelength,

$$\lambda = (2\pi^2 \hbar^2 / mE)^{\frac{1}{2}} \quad (4.2)$$

$\vec{\tau}$ denotes the modulus of a reciprocal lattice vector $\vec{\tau}$ and the summation is carried out over all such vectors satisfying the condition

$$\tau < 4\pi/\lambda \quad (4.3)$$

$P_n(\bar{\mu})$ denotes the n^{th} Legendre polynomial, with argument $\bar{\mu}$ given by the expression

$$\bar{\mu} = 1 - \tau^2 \lambda^2 / 8\pi^2 \quad (4.4)$$

Within the absolute value signs in Eq. 4.1 the summation is over all atoms in the unit cell, a_k denotes the bound coherent scattering length for the k^{th} atom of mass M_k , $\vec{r}_k$ is the position vector of the atom within the unit cell.

Finally, the quantity γ_k is the Debye-Waller integral, given by the expression

$$\gamma_k = \int_0^\infty d\omega \frac{\rho_k(\omega)}{\hbar\omega} \coth\left(\frac{\hbar\omega}{2k_B T}\right) \quad (4.5)$$

where T is the temperature of the scattering material and $\rho_k(\omega)$ is the normal mode frequency distribution for the crystal appropriately weighted by the squared modulus of the polarization vector for the motion of the k^{th} atom (see Section 6). We note for completeness that Eq. 4.1 would be made valid for arbitrary polycrystalline material by the replacement

$$\exp\left[\frac{-\hbar^2 \tau^2 \gamma_k}{4M_k}\right] \rightarrow e^{-W_k} \quad (4.6)$$

where

$$W_k = \frac{\hbar}{4NM_k} \sum_{\vec{q}, j} \frac{\left| \vec{\tau} \cdot \vec{C}\left(k \middle| \vec{q} \middle| j\right) \right|^2}{\omega(\vec{q}, j)} \coth\left[\frac{\hbar\omega(\vec{q}, j)}{2T}\right] \quad (4.7)$$

In 4.7 $\omega(\vec{q}, j)$ denotes the j^{th} normal mode frequency of the crystal associated with the wave vector $\vec{q}$; $\vec{C}\left(k \middle| \vec{q} \middle| j\right)$ is the polarization vector for the k^{th} atom, normalized as follows:

$$\sum_{k=1}^r \left| \vec{C}\left(k \middle| \vec{q} \middle| j\right) \right|^2 = 1 \quad (4.8)$$

and N is the number of atoms in the crystal. The summation over $\vec{q}$ is restricted to the first Brillouin zone while j is summed from 1 to $3r$. For the special case of a crystal (not necessarily a Bravais crystal) with cubic symmetry W_k reduced to $\hbar^2 \tau^2 \gamma_k / 4M_k$.

For cubic crystals with a face-centered cubic primitive lattice, of which UO_2 and UC are examples, the reciprocal lattice vectors are given by: ⁽²⁵⁾

$$\vec{\tau} = \frac{2\pi}{\tilde{a}} (\ell_1 \hat{i} + \ell_2 \hat{j} + \ell_3 \hat{k}) \quad (4.9)$$

where $\tilde{a}$ denotes the lattice constant; $\hat{i}$, $\hat{j}$, and $\hat{k}$ are unit vectors for a Cartesian coordinate system; and the (ℓ_1, ℓ_2, ℓ_3) 's are sets of positive and negative integers which are all even or all odd. The summation in Eq. 4.1 then becomes a summation over all such triplets of integers satisfying the condition

$$\ell_1^2 + \ell_2^2 + \ell_3^2 < \frac{\tilde{a}^2}{\pi \lambda} \quad (4.10)$$

which is obtained at once from Eqs. 4.3 and 4.9. A program which actually carries out the summation over all these triplets would require considerable memory and lengthy running time. For example, ℓ -values of up to 40 are permitted by Eq. 4.3 when E is the order of 1 eV. A considerable simplification is possible for particular lattice structures, however, and it has proved convenient to specialize the code to take full advantage of this simplicity. For example, for UO_2 , which has the CaF_2 (fluorite) structure like ZrH_2 , we can adopt coordinates such that

$$\vec{r}_1 = \vec{0}, \quad \vec{r}_2 = -\vec{r}_3 \quad (4.11)$$

where $k = 1$ designates the uranium atom and $k = 2, 3$ designate oxygen atoms so that the scattering lengths a_2 and a_3 are equal. It is then straightforward to show that the absolute square in Eq. 4.1 is a function only of $(\ell_1^2 + \ell_2^2 + \ell_3^2)$ and $\cos [\frac{\pi}{2} (\ell_1 + \ell_2 + \ell_3)]$. Hence all contributions to the sum over $\vec{\tau}$ in Eq. 4.1 are obtained by considering only ℓ -values such that

$$\ell_1 \geq \ell \geq \ell_3 \geq 0 \quad (4.12)$$

Considering the above one can show that the absolute square in Eq. 4.1 satisfies for UO_2 the equation

$$\left| \sum_{k=1}^r a_k \exp \left[\frac{-\hbar^2 \vec{\tau}^2 \gamma_k}{4M_k} \right] e^{-i\vec{\tau} \cdot \vec{r}_k} \right|^2 = 4a_2^2 e^{-2\vec{\tau}^2 \gamma_2'} \left| \frac{a_1}{2a_2} e^{-\vec{\tau}^2 (\gamma_1' - \gamma_2')} + \cos(\vec{\tau} \cdot \vec{r}_2) \right|^2 \quad (4.13)$$

where

$$\gamma_k' = \frac{\hbar^2 \gamma_k}{4M_k}, \quad k = 1, 2; \quad (4.14)$$

$$\cos(\vec{\tau} \cdot \vec{r}_2) = 0 \text{ when } \ell_1 + \ell_2 + \ell_3 \text{ is odd; and} \quad (4.15)$$

$$\cos(\vec{\tau} \cdot \vec{r}_2) = (-1)^{(\ell_1 + \ell_2 + \ell_3)/2} \text{ when } \ell_1 + \ell_2 + \ell_3 \text{ is even.} \quad (4.16)$$

The method of weighting contributions to the sum which arise from points on the boundary of the region in ℓ -space defined by Eq. 4.12 has been described elsewhere.⁽²⁵⁾ A program incorporating this weighting and using Eq. 4.12 has been written and used to calculate $\sigma_n(E)$ for room temperature UO_2 , using previously derived γ_k' s.⁽²⁸⁾ The weights are given by⁽²⁵⁾

$$W = \frac{3! 2^{\text{NNO}}}{(3 - \text{NNI})!} \quad (4.17)$$

where

NNO = number of ℓ 's (ℓ_1, ℓ_2, ℓ_3) that are not zero

NNI = number of nonrepeated components of (ℓ_1, ℓ_2, ℓ_3)

The coherent elastic cross section of UC has been calculated also by means of the code QSCAT. The summation over reciprocal lattice vectors can be restricted to the same limited region of reciprocal space

as that used in the UO_2 calculation, although the expression for the structure factor is slightly different.

For the UC lattice structure

$$\vec{r}_1 = \vec{0} \quad \vec{r}_2 = (1, 0, 0)$$

where $k = 1$ designates the uranium atom and $k = 2$ designates carbon.

Hence

$$\left| \sum_{k=1}^2 a_k \exp \left[\frac{-\hbar^2 \tau^2 \gamma_k}{4M_k} \right] e^{-i\vec{\tau} \cdot \vec{r}_k} \right|^2 = a_1^2 e^{-2\tau^2 \gamma_1} + a_2^2 e^{-2\tau^2 \gamma_2} + 2a_1 a_2 e^{-\tau^2 (\gamma_1 + \gamma_2)} (-1)^{\ell_1} \quad (4.18)$$

which also considerably simplifies the calculation of Eq. 4.1 for UC.

Table 1 lists the values used in Eq. 4.1 for UO_2 and UC. Values of γ_1 and γ_2 for UC used in Eq. 4.18 were obtained from results of the central force model study⁽²⁶⁾ described in Section 3.

Table 1
VALUE OF PARAMETERS IN COHERENT ELASTIC SCATTERING
CALCULATIONS FOR UO_2 AND UC

Values Used in Equation 4.1	UO_2	UC
$4\pi a_1^2$ - (barns)	9.0	9.0
$4\pi a_2^2$ - (barns)	4.2	5.5
M_1 - mass in grams $\cdot 10^{-24}$	394.50	394.50
M_2 - mass in grams $\cdot 10^{-24}$	26.510	19.901
γ_1 - Debye-Waller integral in eV^{-1}	277.87	362.72
γ_2 - Debye-Waller integral in eV^{-1}	48.294	39.558
a - unit cell dimension in $\text{\AA}$	5.469	4.9554
maximum value of ℓ_i	39	35
maximum energy considered (eV)	1.0	1.0

5. TEMPERATURE DEPENDENCE OF PHONON FREQUENCY DISTRIBUTIONS

The theoretical thermal scattering laws for polycrystalline moderators described in the Kernel Book⁽²⁹⁾ (see Section 9.2) are based on lattice dynamical models in the strict harmonic approximation. In this approximation the lattice constant and the distribution of phonon frequencies are independent of temperature. Temperature-dependence is introduced into thermal neutron scattering only by the change in population of the vibrational excited states. In an actual crystal, however, the presence of anharmonic forces gives rise to thermal expansion of the crystal; the resulting change of interatomic distances alters the harmonic force constants and changes the phonon frequencies. The resulting change with temperature in the frequency distributions and weighted distributions introduces additional temperature dependence in the neutron scattering properties of the material. A theoretical study has been performed to evaluate the importance of this dependence, since sufficient experimental data at different temperatures are not available.

Theoretical descriptions in the literature concerning the connection between phonon frequencies and thermal expansion have only limited applicability to nonprimitive lattices, which are the structures of primary interest in reactor calculations. Therefore an effort to extend and improve the theoretical formulation of this problem was begun at the end of the previous contract year. This work has now been completed and is described in detail in a topical report,⁽⁹⁾ a condensed version of which is to be submitted for publication. The report also contains a description of the application of the theory to the calculation of phonon frequency shifts in

zirconium hydride. We shall review these zirconium hydride results and give, in addition, more recently obtained results concerning the temperature-dependence of the frequency spectrum for graphite.

Zirconium hydride was selected for the first numerical calculation based on this theory for a number of reasons. First of all, it was expected from the relatively large thermal expansion of zirconium hydride that the crystal anharmonicity would be fairly substantial and lead to significant shifts in the phonon frequency distributions. Second, a recently written code, ELSC, based on a central force model for ZrH_2 ,^(25,30) was available and could be readily extended to perform the complicated calculations involved in a test of the thermal expansion formalism. Finally, the formalism, while complicated even for ZrH_2 , was nonetheless considerably simplified by cubic symmetry. On the basis of the rigorous calculation made for zirconium hydride, approximate methods were developed for estimating the order of magnitude of the temperature-dependence of the phonon frequency distribution. These methods have been applied to the study of graphite. The results confirm, as we shall see below, that the temperature-dependence of the graphite frequency distribution does not significantly affect integral neutron thermalization parameters.

Beginning with zirconium hydride, we note that the limitations of available data are such that the anharmonic force constants must be selected solely on the basis of rather imprecise thermal expansion measurements. Thus, it is convenient and reasonable to select the anharmonic constants in such a way that they are given in terms of the harmonic force constants by a single ratio parameter, which can be adjusted if necessary to make the calculated thermal expansion agree with experiment. An initial choice of the ratio parameter can be made by assuming a physically reasonable force law for the interatomic potential and using this potential to determine the anharmonic constants. (The harmonic constants for ZrH_2 are taken from previous work,⁽²⁵⁾ in which they were obtained by fitting neutron and specific heat data.) Thus, a

simple Lennard-Jones form for the central force interatomic potential $\phi(r^2)$ written as a function of the square of the interatomic distance, r , namely,

$$\phi(r^2) = ar^{-12} - br^{-6} \quad (5.1)$$

where a and b are adjustable constants, leads to the result

$$\tilde{r}^2 \phi'''(\tilde{r}^2) = -12 \phi''(\tilde{r}^2) \quad (5.2)$$

where $\tilde{r}$ denotes an equilibrium interatomic separation distance at which the interatomic forces vanish:

$$\phi'(\tilde{r}^2) = 0 \quad (5.3)$$

In Eqs. 5.2 and 5.3, the primes refer to differentiation with respect to r^2 .

Equation 5.2 suggests for the ratio parameter $A^{(9)}$ the choice,

$$A = -12.0$$

which leads to the following calculated values of the thermal expansion:⁽⁹⁾

$$\alpha = 1.125 \cdot 10^{-5}/^\circ\text{K at } T = 300^\circ\text{K} \quad (5.4)$$

$$\alpha = 2.28 \cdot 10^{-5}/^\circ\text{K at } T = 800^\circ\text{K} \quad (5.4)$$

The most nearly comparable experimental measurements have been made by Kempter, Elliott, and Gschneider⁽³¹⁾ for $\text{ZrH}_{1.96}$ in the slightly tetragonal ϵ -phase. For randomly oriented polycrystalline material between 297°K and 573°K , they observed an average linear expansion coefficient of $0.93 \cdot 10^{-5}/^\circ\text{K}$, with which our calculated values are in satisfactory qualitative agreement. By varying the value of A within reasonable limits, we could obtain an improved fit to the observed α , but in view of the average nature of the data, a precise fit would not be particularly meaningful. (As an example of dependence of α on A , we

note that a calculation with $A = -10.0$ gave a value of $0.86 \cdot 10^{-5}/^{\circ}\text{K}$ for α at 300°K .) Figure 5.1 shows phonon frequency distributions for ZrH_2 based on 520- $\vec{q}$ -point calculations at 300°K and 800°K with $A = -12.0$. A shift to lower frequencies is apparent at the higher temperature, the central frequency of the optical part being decreased by about fifteen percent. (The distribution at 300°K calculated here is not in agreement with neutron data, since the harmonic force constants have not been reevaluated to provide fits to the neutron data in the presence of anharmonicity. The frequency shifts introduced by the anharmonicity displace the optical peak shown here from its observed position centered around 0.14 eV. We have not adjusted the force constants to restore agreement since our purpose here is to illustrate the effects of thermal expansion rather than provide accurate phenomenological fits.)

The squared-amplitude-weighted distributions used in neutron scattering calculations have not been calculated, but are expected to be shifted by about the same amount. Of course, these shifts do not give a complete specification of the lowest order effects of anharmonicity on neutron scattering since we have not included purely anharmonic phonon frequency shifts of the type calculated by Maradudin and Fein⁽³²⁾ and by Cowley.⁽³³⁾ In their work, the third and fourth order purely anharmonic shifts turn out to be somewhat smaller than the thermal expansion shifts but of the same order of magnitude. However, the third and fourth order shifts are of opposite sign, the third order shift having the same sign as the thermal expansion shift, so that there is a partial cancellation of the purely anharmonic shifts. Thus, a thermal expansion calculation such as we have performed for ZrH_2 probably gives a good estimate of the over-all magnitude of the temperature dependence of the phonon frequencies as they appear in neutron scattering calculations.

Temperature-dependent shifts in phonon frequencies have been detected experimentally in the past, e.g., for aluminum⁽³⁴⁾ and lead.⁽³⁵⁾

Figure 5.1. Phonon frequency distributions for ZrH_2 at 300°K and 800°K .

Shifts of the magnitude illustrated in Fig. 5.1 could no doubt be detected by precise double-differential measurements at elevated temperatures. However, it is not a priori clear how important such shifts are likely to be in thermalization calculations. To investigate this question without having to develop a full-scale anharmonic-model kernel, we prepared a modified scattering law for H in ZrH_2 at 800°K by arbitrarily shifting the optical part of the weighted frequency spectrum for H, $\rho_H(\omega)$, as given by the harmonic central force model.⁽²⁵⁾ We imposed a shift agreeing in order of magnitude with the calculated thermal expansion shifts of Fig. 5.1 by using the following rule:

$$\rho_{\text{Hs}}(\omega) = \rho_H(\omega + 0.15 \omega) \quad (5.5)$$

where $\rho_{\text{Hs}}(\omega)$ denotes the new shifted weighted frequency spectrum. The modified scattering law at 800°K was then calculated by using $\rho_{\text{Hs}}(\omega)$ in the code GASKET.⁽¹⁾ This scattering law was used in the code BSPEC⁽³⁶⁾ to calculate the infinite medium neutron spectrum in an erbium-poisoned ZrH_2 assembly at 800°K , the erbium poisoning being 3b/H atom. A similar calculation was made using the ENDF scattering law for ZrH_2 , which is based on the unshifted room-temperature weighted central-force-model phonon frequency spectrum.⁽²⁵⁾ The differences between the two calculated neutron spectra were essentially negligible. Physically, one expects scattering laws and neutron spectra at elevated temperatures to be somewhat insensitive to the details of the phonon frequency distribution, since as the temperature of the moderator increases its neutron scattering properties become more gas-like. Our results indicate that this insensitivity is sufficient to keep thermal expansion shifts of the magnitude seen in Fig. 5.1 for ZrH_2 from influencing neutron thermalization calculations.

In view of the above results, the effect of frequency distribution temperature dependence on neutron scattering and thermalization by graphite is not expected to be of great significance. The thermal expansion

of artificially produced (reactor grade) graphite at room temperature lies roughly in the range ⁽³⁷⁾

$$\begin{aligned}\alpha_{\parallel} &= 0.18 \text{ to } 0.435 \times 10^{-5} / ^{\circ}\text{K} \\ \alpha_{\perp} &= 0.48 \text{ to } 0.685 \times 10^{-5} / ^{\circ}\text{K}\end{aligned}\tag{5.6}$$

where $\alpha_{\parallel}$ and $\alpha_{\perp}$ denote respectively the coefficients of linear expansion parallel and perpendicular to the axis of extrusion. Thus the upper limit for $\alpha_{\perp}$ is less than two-thirds the room temperature thermal expansion obtained by the anharmonic calculation for ZrH_2 ⁽⁹⁾ on which the frequency shifts seen in Fig. 5.1 are based. Accordingly, one would expect the shifts with temperature in the phonon frequency distribution for graphite to be much less than the shifts seen for ZrH_2 . However, since the structure of graphite and the over-all shape of its frequency distribution in the harmonic model ⁽³⁸⁾ is drastically different from ZrH_2 , it is desirable to make a quantitative investigation.

The structure of graphite consists of weakly coupled planar sheets of hexagonally-arrayed atoms. A force model involving four interatomic force constants was developed by Yoshimori and Kitano ⁽³⁸⁾ and found by Young and Koppel ⁽³⁹⁾ and by Takahashi ⁽⁴⁰⁾ to lead to good agreement with specific heat data and with neutron data, respectively. In this model the forces act either parallel or perpendicular to the planar sheets. The two parallel, or in-plane forces, are associated with: (a) changes in the bond angles, proportional to a force constant μ ; and (b) changes in bond length between nearest neighbors in the same plane, proportional to a force constant K . The perpendicular forces are: (c) a central force due to changes in the bond length between nearest neighbors in two adjacent planes, proportional to a force constant K' ; and (d) a force due to the bending of the planes and proportional to the displacement perpendicular to the planar sheets of any atom relative to its nearest neighbors in the

same plane. A force constant μ' is associated with this last force. The values of these constants as selected by Young and Koppel⁽³⁹⁾ are

$$\begin{aligned}
 (a) \quad \mu &= 3.6 \cdot 10^4 \text{ dyne/cm} \\
 (b) \quad K &= 4.3615 \cdot 10^5 \text{ "} \\
 (c) \quad K' &= 5.86521 \cdot 10^3 \text{ "} \\
 (d) \quad \mu' &= 2.92 \cdot 10^5 \text{ "}
 \end{aligned} \tag{5.7}$$

The change in phonon frequencies associated with thermal expansion comes about physically from the fact that as the lattice expands and the atoms move farther apart, the interatomic forces change with increasing separation. A rigorous lattice dynamical treatment of these changes requires a formal and numerical treatment of the type we have described above for ZrH_2 . We can, however, estimate the order of magnitude of the frequency shifts by evaluating the change in the harmonic force constants on the basis of an assumed form for the interatomic potential. This procedure, which we describe more fully below, has been checked by using it to estimate the frequency shifts for ZrH_2 and comparing with the rigorously calculated results. A hand calculation of frequencies at the center of the Brillouin zone for ZrH_2 indicated that the approximately calculated shifts have the correct order of magnitude.

Since bond angles and plane-bending will not be affected by expansion of the lattice, we expect that the constants μ and μ' in (5.7) will not undergo effective changes due to thermal expansion. The forces (b) and (c) associated with changes in bond length can be described in terms of central force potentials $\Phi_b(r^2)$ and $\Phi_c(r^2)$ written, as in Eq. 5.1, as a function of the square of the interatomic distance r^2 .⁽⁴¹⁾ Let $\tilde{r}_b$ and $\tilde{r}_c$ denote the room temperature equilibrium bond lengths appropriate to these forces. Then, if we require

$$\Phi'_b(\tilde{r}_b^2) = \Phi'_c(\tilde{r}_c^2) = 0 \tag{5.8}$$

to insure that the net forces on the atoms vanish at equilibrium, we can relate the force constants K and K' to the interatomic potential as follows:

$$K = B \tilde{r}_b^2 \Phi''(\tilde{r}_b^2)$$

$$K' = C \tilde{r}_c^2 \Phi''(\tilde{r}_c^2)$$
(5.9)

on using the formulation of the central force model given by Born and Huang,⁽⁴¹⁾ where B and C are temperature-independent proportionality constants.

At a temperature ΔT above room temperature, the equilibrium bond lengths $\tilde{r}(\Delta T)$ will be given approximately by

$$\tilde{r}_b(\Delta T) = \tilde{r}_b \cdot (1 + \Delta T \cdot \alpha_{\parallel})$$

$$\tilde{r}_c(\Delta T) = \tilde{r}_c \cdot (1 + \Delta T \cdot \alpha_{\perp})$$
(5.10)

where $\alpha_{\parallel}$ and $\alpha_{\perp}$ denote the average coefficients of linear expansion parallel and perpendicular, respectively, to the planar sheets. Equation 6 gives the numerical magnitudes for these coefficients. From Eq. 5.9 we can estimate the change in the force constant K , for example, by writing

$$dK = B d\tilde{r}_b^2 \left[2 \tilde{r}_b \Phi''(\tilde{r}_b^2) + 2 \Phi'''(\tilde{r}_b^2) \tilde{r}_b^3 \right]$$
(5.11)

where from Eq. 5.10

$$d\tilde{r}_b = \tilde{r}_b \cdot \Delta T \cdot \alpha_{\parallel}$$
(5.12)

If we assume that the potential is given by Eq. 5.1, which as we saw above gave good results for thermal expansion in ZrH_2 , we obtain from Eqs. 5.2 and 5.11 the result

$$dK = B \frac{dr_b}{r_b} \left[2 \tilde{r}_b^2 \tilde{\Phi}_b''(\tilde{r}_b^2) - 24 \tilde{r}_b^2 \tilde{\Phi}_b''(\tilde{r}_b^2) \right] = -22 B \tilde{r}_b \tilde{r}_b \tilde{\Phi}_b''(\tilde{r}_b^2) \quad (5.13)$$

where we have used Eqs. 5.9 and 5.12 to write the final result in (5.13).

Consider graphite at a temperature in the vicinity of 1300°K, for which

$$\Delta T \approx 1000^\circ K. \quad (5.14)$$

With the choice

$$\alpha_{||} = 0.40 \cdot 10^{-5} \quad (5.15)$$

from the range given by Eq. 5.6, we obtain from Eqs. 5.13 and 5.14

$$dK \approx -0.088 K \quad (5.16)$$

Substituting from Eq. 5.7 gives

$$K(1300^\circ C) \approx K + dK = (4.3615 - 0.384) 10^5 = 3.9775 \cdot 10^5 \text{ dyne/cm} \quad (5.17)$$

Similarly,

$$\begin{aligned} K'(1300^\circ C) &= K' - 0.088 K' = (5.86521 - 0.51614) 10^3 \\ &= 5.34907 \cdot 10^3 \text{ dyne/cm.} \end{aligned} \quad (5.18)$$

The normal mode frequency distribution for graphite in the Yoshimori-Kitano model is the weighted sum of the independent frequency distributions for parallel and perpendicular vibrations. These distributions are calculated with root sampling techniques by the program CAFS,⁽³⁹⁾ originally written for the IBM 7044 computer and now converted to run on the UNIVAC 1108. The parallel frequency distributions, based on constants K and μ given by Eq. 5.7 and on $K(1300^\circ K)$ given by Eq. 5.17 and μ given by Eq. 5.7, are shown in Fig. 5.2, taken from a CAFS calculation based on 10,261 q-points. Even though the temperature increase considered in this case is twice as large as the increase investigated for zirconium

Figure 5.2. Frequency distributions for parallel vibrations of the graphite lattice at room temperature and at 1300°K (solid line is the room temperature distribution).

hydride, the shift in the frequency distribution for graphite is seen to be quite small. The perpendicular frequency distribution, not shown in the figure, is not very sensitive to changes in K' , particularly at higher frequencies, and consequently exhibits even smaller shifts.

Thus, in view of the general insensitivity of high temperature thermalization calculations to the detailed structure of the moderator frequency distribution, as illustrated above by calculations for ZrH_2 , it can be concluded that the temperature dependence of the graphite phonon frequencies can be neglected in such calculations.

6. CRYSTAL BINDING EFFECTS ON DOPPLER BROADENING OF NEUTRON ABSORPTION RESONANCES

6.1 INTRODUCTION

The effect of target particle thermal motion on the shape of neutron absorption resonances (Doppler broadening) was first treated by Bethe and Placzek,⁽⁴²⁾ who considered free gas targets with a Maxwellian velocity distribution. Their treatment turns out to be approximately valid even for crystalline targets, when certain "weak binding" conditions introduced originally by Lamb⁽⁴³⁾ are satisfied, provided that an effective temperature is substantiated for the actual temperature. The free gas model for Doppler broadening, even without the effective temperature refinement, has proved fairly satisfactory in a wide variety of applications, particularly in reactor physics.⁽⁴⁴⁾ Nevertheless, it was recognized that for certain targets — in particular, for tightly bound oxides of uranium — chemical binding effects might affect the line shape significantly. A general formalism for obtaining resonance line shapes in terms of functions describing noninterference slow neutron scattering by the target material was given by Nelkin and Parks.⁽⁴⁵⁾ However, the numerical results obtained with this formalism were originally limited by the difficulty of obtaining complete and accurate double-differential thermal scattering cross sections. This difficulty has been largely eliminated by the availability of convenient computer codes⁽¹⁾ for calculating these cross sections for polycrystalline material, using appropriately weighted phonon frequency distributions, and by the recent development of lattice dynamical models for calculating these distributions for a number of important reactor materials. It is thus possible for these materials to test line shapes obtained by

approximate methods for comparison to essentially exact results. It will be seen below that such comparisons tend to confirm the accuracy of the free gas model at and above room temperature. At low temperatures ($\sim 50^\circ\text{K}$), however, it becomes important to have a lattice dynamical description of the crystal binding effects. This need is particularly apparent in the study of polyatomic targets, like UO_2 , for which the weighted frequency distributions (see below) differ drastically from the unweighted normal mode distributions. The importance of this difference has not always been recognized in the past, and as a result the effects of crystalline binding have sometimes been significantly overestimated. (46)

A major purpose of this section is to stress the importance of using the correct weighted frequency distributions in Doppler broadening calculations. Some general formulas are reviewed in the following section. Comparisons are given between approximate line shapes and exact results based on detailed scattering law and lattice dynamical calculations for uranium dioxide.

6.2 FORMULAS FOR DOPPLER BROADENING

Lamb's weak binding condition for a crystal containing one type of atom is given as follows: (43)

$$\frac{1}{2}\Gamma + \left(\frac{m}{M} E \bar{\epsilon} \right)^{\frac{1}{2}} \gg \theta \quad (6.1)$$

where Γ is the natural width of the resonance, E is the kinetic energy of the neutron (in the laboratory system), m and M are respectively the neutron and target masses, $\bar{\epsilon}$ is the average energy per vibrational degree of freedom of the crystal, and θ is the Debye temperature of the crystal in energy units. The broadened absorption cross section $\sigma_{n,\gamma}(\Delta, E)$ as a function of E and the Doppler width Δ is given (47) for a free gas target by the integral

$$\sigma_{n, \gamma}(\Delta, E) = \frac{1}{\sqrt{\pi}\Delta} \frac{1}{\sqrt{E}} \int_0^\infty dE' \sqrt{E'} \sigma_{n, \gamma}(E') \exp \left[-\frac{(E' - E)^2}{\Delta^2} \right] \quad (6.2)$$

where

$$\Delta = 2(k_B T_{Em}/M)^{\frac{1}{2}}; \quad (6.3)$$

$\sigma_{n, \gamma}(E')$ is the unbroadened neutron absorption cross section, and T denotes the temperature of the gas. For a crystalline target satisfying the weak binding conditions the Doppler broadened cross section is still given by Eq. 6.2, but T in Eq. 6.3 must be replaced⁽⁴³⁾ by the "effective temperature" $\bar{T}$, where

$$k_B \bar{T} = \bar{\epsilon} \quad (6.4)$$

The effective temperature $\bar{T}$ is given in terms of $\rho(\omega)$, the normalized distribution of normal mode frequencies, by the integral

$$\bar{T} = (2k_B)^{-1} \int_0^\infty \rho(\omega) \omega \coth \left(\frac{\omega}{2kT} \right) d\omega \quad (6.5)$$

in units such that $\hbar = 1$. As T becomes large the properties of the crystal become more gas-like until in the limit Eq. 6.5 gives the result

$$\bar{T} \rightarrow T \text{ as } T \rightarrow \infty \quad (6.6)$$

Thus at high temperature the effective temperature model is equivalent to the free gas model.

General formulas for Doppler broadening when the weak binding approximation does not apply were given (for the case of an isolated absorption resonance) in a particularly convenient form by Nelkin and Parks.⁽⁴⁵⁾ They noted that for a crystalline target the resonance line shape $W(E)$, normalized to unit area, for absorption by atoms of type k can be written in the form

$$W(E) = \frac{\Gamma}{2\pi} \int_{-\infty}^{\infty} d\omega \tilde{S}_k(\vec{p}, \omega) [(E - E_0 - \omega)^2 + \frac{1}{4}\Gamma^2]^{-1} \quad (6.7)$$

where E_0 is the resonance energy and $\vec{p}$ is the momentum of the incident neutron of energy E ,

$$p = |\vec{p}| = \sqrt{2mE} \quad (6.8)$$

The function $\tilde{S}_k(\vec{p}, \omega)$ contains the dependence of the lineshape on the atomic system and is given most conveniently as the Fourier transform of a so-called intermediate scattering function, $\chi_k(\vec{p}, t)$, as follows:

$$\tilde{S}_k(\vec{p}, \omega) = (2\pi)^{-1} \int_{-\infty}^{\infty} dt e^{-it\omega} \chi_k(\vec{p}, t) \quad (6.9)$$

Nelkin and Parks⁽⁴⁵⁾ restrict themselves to the case of a cubic Bravais lattice for which $\chi(\vec{p}, t)$ (dropping the subscript k) is independent of the direction of $\vec{p}$ and can be given as follows:

$$\chi(\vec{p}, t) = \chi(p, t) = \exp \{ R[\gamma(t) - \gamma(0)] \} \quad (6.10)$$

where

$$R = p^2/2M \quad (6.11)$$

and

$$\gamma(t) = \int_0^{\infty} [\coth(\omega/2k_B T) \cos \omega t + i \sin \omega t] \rho(\omega) \omega^{-1} d\omega \quad (6.12)$$

This equation can be derived readily from expressions given by Sjölander.⁽⁴⁸⁾ For nonprimitive lattices these expressions lead to the result,

$$\chi_k(\vec{p}, t) = \exp \left\{ \sum_{\vec{q}, j} \frac{|\vec{p} \cdot \vec{c}(k | \vec{q}_j)|^2}{4N\omega_j(\vec{q})M_k \sinh [\omega_j(\vec{q})/2k_B T]} \sum_{\eta=\pm 1} e^{-\eta\omega_j(\vec{q})/2k_B T} \begin{pmatrix} -i\eta\omega_j(\vec{q})t \\ e^{-1} \end{pmatrix} \right\} \quad (6.13)$$

In Eq. 6.13 $\vec{c}(k | \vec{q}_j)$ denotes the polarization vector for the motion of the k^{th} atom of mass M_k in the unit cell in the j^{th} mode associated with wave vector $\vec{q}$ and having frequency $\omega_j(\vec{q})$. The crystal is considered to consist of N unit cells and has periodic boundary conditions. The wave vectors satisfy the basic normalization condition, (48)

$$\sum_{k=1}^r \vec{c}^*(k | \vec{q}_j) \cdot \vec{c}(k | \vec{q}_i) = \delta_{ij} \quad (6.14)$$

where r denotes the number of atoms per unit cell and δ_{ij} is the Kronecker delta function. The summation over $\vec{q}$ in Eq. 6.13 includes the wave vectors lying in the first Brillouin zone; j is summed from 1 to $3r$.

For a large class of nonprimitive lattices having cubic symmetry it can be shown by using the symmetry transformation formalism of Maradudin and Vosko⁽⁴⁹⁾ that $\chi_k(\vec{p}, t)$ is independent of the direction of $\vec{p}$. We shall restrict ourselves to this class, which includes crystals having the fluorite structure (e.g., CaF_2 , ZrH_2 , UO_2) and the NaCl structure (e.g., UC). The generalization of Eqs. 6.10 and 6.12 for such crystals becomes

$$\chi_k(\vec{p}, t) = \chi_k(p, t) = \exp \{R[\gamma_k(t) - \gamma_k(0)]\} \quad (6.15)$$

where

$$\gamma_k(t) = \int_0^\infty [\coth(\frac{\omega}{2kT}) \cos \omega t + i \sin \omega t] \rho_k(\omega) \omega^{-1} d\omega \quad (6.16)$$

In (6.16) $\rho_k(\omega)$ represents the normalized weighted frequency distribution,⁽²⁸⁾

$$\rho_k(\omega) = \lim_{\Delta\omega \rightarrow 0} \frac{\left(\sum_{\vec{q}} \sum_j |\vec{c}(k|\vec{q}_j)|^2 \right)_{\Delta\omega}}{3N\Delta\omega} \quad (6.17)$$

where the subscript $\Delta\omega$ indicates that only those terms in the summation are retained for which $\omega_j(\vec{q})$ lies in the interval ω to $\omega + \Delta\omega$. In Eq. 6.16 the upper limit on the integration is given formally as ∞ , but in fact the integral is cut off at an upper limit determined by the largest normal mode frequency for which the distribution $\rho_k(\omega)$ is nonvanishing.

For the restricted class of nonprimitive cubic crystals which we are considering, we can rewrite Eq. 6.7 without the dependence on the direction of the neutron momentum,

$$W(E) = \frac{\Gamma}{2\pi} \int_{-\infty}^{\infty} d\omega \tilde{S}_x(p, \omega) [(E - E_0 - \omega)^2 + \frac{1}{4}\Gamma^2]^{-1} \quad (6.18)$$

where in this special case Eq. 6.9 can be written as

$$\tilde{S}_k(p, \omega) = (2\pi)^{-1} \int_{-\infty}^{\infty} dt e^{-it\omega} \chi_k(p, t) \quad (6.19)$$

with $\chi_k(p, t)$ given by Eqs. 6.15 and 6.16 in terms of the weighted frequency distribution. The function $\tilde{S}_k(p, \omega)$ is related as follows to the scattering law $S_k(\alpha, \beta)$ ⁽⁵⁰⁾ describing the noninterference slow neutron scattering by k-atoms of the crystal in the Fermi pseudopotential approximation:

$$\tilde{S}_k(p, \omega) = e^{-\beta/2} S_k(\alpha, \beta) \quad (6.20)$$

where

$$\alpha = p^2 / (2M_k k_B T) = (mE) / M_k k_B T \quad (6.21)$$

and

$$\beta = -\omega / k_B T \quad (6.22)$$

Definition 6.22 appears to differ from Eq. 2.5 by a sign; this difference stems from a difference in the definition of ω , which is chosen in this section to agree with the notation of Nelkin and Parks.⁽⁴⁵⁾

Substitution of 6.19 in 6.18 enables the line shape, $W(E)$ to be written in the form⁽⁴³⁾

$$W(E) = (2\pi)^{-1} \int_{-\infty}^{\infty} dt \exp [i(E_0 - E)t - \frac{1}{2}\Gamma |t|] \chi_k(p, t) \quad (6.23)$$

It is thus possible to calculate the line shape either as a convolution, or by numerically evaluating the time integral in Eq. 6.23. Normally, the latter procedure would be a difficult numerical problem. However, the code GASKET,⁽¹⁾ which calculates the scattering law^{*} by numerically evaluating the Fourier transform in Eq. 6.19, can be relatively easily modified to include an additional time dependence $\frac{1}{2}\Gamma |t|$ in the exponent in order to perform lineshape calculations with Eq. 6.23.

For a crystal which is not monatomic, the effective temperature model and the weak binding condition⁽⁴³⁾ must be modified so that $\bar{\epsilon}$ and θ in Eq. 6.1 refer respectively to the average energy and Debye temperature associated with the vibrations of the type of atom for which the resonant cross sections are being studied. An obvious generalization of Eq. 6.1 for a nonprimitive crystal is

---

* The scattering law calculated by GASKET⁽¹⁾ is equivalent to the scattering law defined by Egelstaff⁽⁵⁰⁾ times the inverse of the temperature in energy units.

$$\frac{1}{2}\Gamma + \left(\frac{m}{M_k} E \bar{\epsilon}_k \right)^{\frac{1}{2}} \gg \theta_k \quad (6.24)$$

where

$$\bar{\epsilon}_k = k\bar{T}_k = \frac{\hbar}{2k_B} \int_{-\infty}^{\infty} \rho_k(\omega) \omega \coth\left(\frac{\omega}{2k_B T}\right) \quad (6.25)$$

In Eq. 6.24 θ_k , the "Debye temperature for atoms of type k", can be meaningfully interpreted only in a somewhat loose fashion by selecting a Debye frequency distribution that most closely approximates the actual weighted distribution, $\rho_k(\omega)$. A precise definition of θ_k is not really needed, since weak binding conditions like Eqs. 6.1 and 6.24 are not particularly stringent, as will be illustrated below.

6.3 NUMERICAL RESULTS

We have calculated the lineshape for the 6.65 eV resonance for ^{238}U in UO_2 using the code LAMB, a modified version of GASKET⁽¹⁾, to evaluate the integral in Eq. 6.23. As a check on the correctness of the modification and the adequacy of the integration meshes, some separate calculations were done with a code written to evaluate the convolution integral, Eq. 6.18. These results confirmed the accuracy of the modified GASKET code, which is somewhat simpler to use than the convolution method, since only an appropriate frequency spectrum is required as input and an independent calculation of the scattering law is not necessary before computing the line shape. It also turns out in the calculations described below that it was possible in evaluating Eq. 6.23 for different values of E to neglect the dependence of p on E and simply evaluate $\chi_k(p, t)$ at

$$p = \sqrt{2mE_0} \quad (6.26)$$

At the resonance peak this approximation is of course exact. The ^{238}U resonance falls off sufficiently rapidly that the lineshape is essentially unaffected by the dependence of $\chi_k(p, t)$ on E . Considerable computation time is saved by this approximation.

Our purpose in studying the ^{238}U lineshape in UO_2 was to make a direct comparison between the effective temperature model and the more general calculation taking complete account of crystalline binding. A lattice dynamical model for UO_2 developed at Gulf General Atomic⁽²⁸⁾ provided the weighted frequency spectrum for U in UO_2 necessary for the calculation of $\bar{T}_k$ in Eq. 6.5 and $\chi_k(p, t)$ in Eq. 6.23 for $k = \text{U}$. This spectrum is shown in Fig. 6.1. Figure 6.2 shows the unweighted normal mode frequency spectrum for UO_2 . It is clear from comparing these figures that the higher frequency modes are primarily associated with the motion of the oxygen atoms, while the vibrations affecting the Doppler broadening of ^{238}U cross sections are primarily those of lower energies (< 0.04 eV). Calculations⁽⁴⁶⁾ have sometimes been based on the normal mode spectrum, the distribution obtained from specific heat studies, which clearly gives too high a Debye temperature and average energy for the vibrations of the heavy uranium atom in oxides like UO_2 or U_3O_8 .

Figure 6.3 shows the broadened lineshape for ^{238}U in UO_2 at $T = 296^\circ\text{K}$ (solid line) compared with ^{238}U as a free gas at $T = 296^\circ\text{K}$ (dotted line) and ^{238}U as a free gas at $\bar{T}_U = 311.1^\circ$, the value obtained from Eq. 6.5 by substituting the weighted frequency distribution of Fig. 6.1 for $\rho_k(\omega)$ and setting $T = 296^\circ\text{K}$. The resonance parameters E_0 and $\Gamma/2$ were taken to have the values

$$E_0 = 6.65 \text{ eV}$$

$$\Gamma/2 = 0.014 \text{ eV}$$

The free gas calculations are also made with GASKET by calculating an intermediate scattering function $\chi_u(p, t)$ appropriate to a free gas.⁽⁴⁵⁾

Figure 6.1. Weighted frequency spectrum for U in UO₂.

Figure 6.2. Normal mode frequency spectrum for UO_2 .

This choice can be shown from Eq. 6.18 or 6.23 to lead essentially to Eq. 6.2, where

$$\sigma_{n,\gamma}(E) \propto \Gamma [(E - E_0)^2 + \Gamma^2/4]^{-1} \quad (6.26)$$

The curves in Fig. 6.3 agree within a few percent, indicating that the free gas method of Doppler broadening is quite accurate at this temperature. If we check the weak binding condition, Eq. 6.24 for U in UO_2 we find for $E = 6.675$ eV (laboratory energy corresponding to a resonance energy of 6.65 eV) the result,

$$\frac{1}{2}\Gamma + \left(\frac{m}{M} E \bar{\epsilon}_U\right)^{\frac{1}{2}} = 0.042 \text{ eV at } T = 296^\circ\text{K} \quad (6.22)$$

where $\bar{\epsilon}_U = k\bar{T}_U = 0.265$ eV for $\bar{T}_U = 311.1^\circ\text{K}$.

A rough estimate of a suitable Debye temperature θ_U can be obtained from Fig. 6.1, where it is seen that the bulk of the uranium weighted spectrum lies below 0.025 eV. The comparison of 0.042 eV with 0.025 eV indicates in view of the success of the free gas model in this case that the weak binding condition need not be satisfied too strictly. At higher temperature $\bar{\epsilon}_U$ will increase, improving the satisfaction of this condition. Figure 6.4 shows $\bar{T}_U$ as a function of T computed from Eq. 6.25; $\bar{T}_U$ is seen to approach T asymptotically at higher temperatures.

When we look at lower temperatures, however, the situation changes. At $T = 50^\circ\text{K}$ we obtain from Fig. 6.4 the value

$$\bar{T}_U = 110.2^\circ\text{K (corresponding to } \bar{\epsilon}_U = 0.00946 \text{ eV)}$$

with the result for $E = 6.675$ eV,

$$\frac{1}{2}\Gamma + \left(\frac{m}{M} E \bar{\epsilon}_U\right)^{\frac{1}{2}} = 0.031 \text{ eV} \quad (6.28)$$

The weak binding condition in this case is evidently not satisfied, since as Fig. 6.5 shows, there are major differences between the correct crystalline-binding lineshape calculation (solid line) and the free gas

Figure 6.3. Doppler broadened line shape for 6.65 eV resonance of ^{238}U at $T = 296^\circ \text{K}$.

Figure 6.4. Effective temperature as a function of temperature for U in UO_2 .

Figure 6.5. Doppler broadened line shape for 6.65 eV resonance of ^{238}U at $T = 50^\circ\text{K}$.

(dotted line) and the free-gas-with-effective-temperature model (broken line). The effective temperature model is seen to give a significantly better approximation to the crystalline calculation than the simple free gas model. A fourth curve (second broken line) shows the results of performing the crystalline binding calculation incorrectly by using the unweighted frequency spectrum of Fig. 6.2. Such a procedure greatly overestimates the effects of the binding on Doppler broadening. A lattice dynamical model is also available for uranium monocarbide (UC)⁽⁶⁾ in addition to UO_2 and has been used to calculate the lineshape of the 6.65 eV resonance for ^{238}U at 296°K and 50°K . Although the normal mode distribution for UC is quite different from that for UO_2 , the weighted distribution for U in UC is qualitatively similar to that for U in UO_2 and almost identical lineshapes were obtained.

6.4 CONCLUSIONS

At and above room temperature the free gas model for Doppler broadening, with or without introduction of the effective temperature, appears to give accurate lineshapes. The high Debye temperatures of tightly bound systems like uranium dioxide do not imply failure of the gas model, since it is the weighted frequency distribution, Eq. 6.17, for the heavy atom that determines the applicability of approximate methods rather than the normal mode distribution. A characteristic of these weighted distributions, as illustrated by results for UO_2 and UC, is that most of the heavy atom vibrations have energies at or below about .025 eV. The higher frequencies, which give rise to the high Debye temperatures determined by specific heat measurements, are associated with vibrations of the lighter atom and are not relevant to broadening of the heavy atom absorption lines.

At low temperatures the chemical binding of the heavy atom has a significant effect on the lineshape. An effective temperature model gives much better results than a pure free gas model, but is still significantly

different from exact results. Actually, the effective temperature approximation for nonmonatomic crystals is not much easier to use than exact calculations, since a detailed lattice dynamical calculation to provide the weighted frequency distribution is necessary for either procedure. For primitive lattice targets the effective temperature is easier to obtain, since the weighted and normal mode distributions are identical and can often be obtained to the degree of accuracy necessary for lineshape calculations by fitting specific heat data. (51)

7. MISCELLANEOUS STUDIES

7.1 TOTAL CROSS SECTION OF ZIRCONIUM HYDRIDE

A measurement of the total cross section of zirconium hydride was performed during the previous contract year. The experimental technique was the same as that used for the measurement of the total cross sections of benzene⁽⁵²⁾ and uranium dicarbide.⁽⁵³⁾ The data have been analyzed during this contract year.

The sample used was $\text{ZrH}_{1.81}$ in powdered form with impurity content less than 0.1%. Two samples were used, with the following characteristics:

<u>Sample</u>	<u>Thickness (cm)</u>	<u>Density (g/cm³)</u>	<u>Transmission at 0.05 eV</u>
1	0.970	3.015	0.20
2	0.452	3.79	0.38

The data were analyzed with the technique described in Ref. 52. Corrections for in-scattering and multiple scattering were applied according to Eqs. 1 to 11 of Ref. 54 and the value of the cross section was normalized to the free atom cross section.

Table 2 gives our best evaluation of the measured total cross section of $\text{ZrH}_{1.81}$, obtained from an average of four measurements. In this table the cross section is given in barns per hydrogen atom.

Figure 7.1 shows a comparison of the measured total cross section with the prediction of the central force lattice dynamical model.⁽²⁵⁾ Since the calculated value was available for the 1.85 composition, the experimental data have also been reduced to this value. The agreement of the measured value with the theoretical one is satisfactory except at

TABLE 2
EXPERIMENTAL TOTAL CROSS SECTION OF $ZrH_{1.81}$
PER HYDROGEN ATOM

<u>E(eV)</u>	<u>σ_T (barns)</u>	<u>$\Delta\sigma_T$ (barns)</u>	<u>E(eV)</u>	<u>σ_T (barns)</u>	<u>$\Delta\sigma_T$ (barns)</u>
0.0094	72.41	0.762	0.0809	34.16	0.257
0.0099	72.25	0.724	0.0858	32.56	0.252
0.0105	73.87	0.707	0.0912	31.25	0.249
0.0111	73.60	0.680	0.0971	30.20	0.248
0.0117	72.09	0.675	0.103	28.75	0.247
0.0124	69.82	0.632	0.110	27.52	0.248
0.0131	70.30	0.616	0.118	26.55	0.251
0.0138	68.31	0.583	0.127	25.66	0.257
0.0146	68.31	0.562	0.137	26.95	0.274
0.0155	66.20	0.529	0.146	27.94	0.341
0.0164	67.01	0.540	0.154	29.69	0.362
0.0173	66.53	0.524	0.163	29.09	0.374
0.0182	65.50	0.502	0.172	30.16	0.388
0.0193	64.74	0.485	0.182	29.30	0.398
0.0204	63.66	0.464	0.193	29.09	0.407
0.0216	62.80	0.446	0.204	28.07	0.410
0.0229	60.42	0.442	0.217	27.49	0.416
0.0241	60.10	0.432	0.232	26.62	0.414
0.0254	59.67	0.420	0.247	25.13	0.411
0.0269	58.37	0.404	0.264	25.54	0.415
0.0284	56.86	0.387	0.283	26.39	0.424
0.0301	56.16	0.376	0.305	26.31	0.423
0.0320	55.02	0.364	0.328	26.30	0.423
0.0340	53.08	0.347	0.354	25.01	0.414
0.0361	52.27	0.354	0.384	24.47	0.408
0.0380	51.03	0.343	0.417	24.66	0.407
0.0401	49.89	0.332	0.455	24.82	0.410
0.0424	48.54	0.322	0.498	23.96	0.406
0.0448	47.41	0.312	0.547	24.22	0.410
0.0476	46.22	0.303	0.604	24.27	0.410
0.0505	45.09	0.284	0.652	23.41	0.555
0.0537	43.57	0.285	0.688	24.20	0.565
0.0573	41.63	0.274	0.726	24.00	0.559
0.0612	40.06	0.264	0.768	24.03	0.565
0.0649	38.66	0.274	0.813	24.54	0.561
0.0685	37.99	0.272	0.860	24.08	0.558
0.0723	35.83	0.263	0.916	23.21	0.557
0.0764	52.83	0.261	0.979	24.55	0.558

TABLE 2 (cont)

<u>E(eV)</u>	<u>σ_T</u> (barns)	<u>$\Delta\sigma_T$</u> (barns)	<u>E(eV)</u>	<u>σ_T</u> (barns)	<u>$\sigma\Delta_T$</u> (barns)
1.048	24.82	0.566	1.537	24.46	0.554
1.125	23.20	0.555	1.675	23.19	0.519
1.211	23.90	0.555	1.832	23.63	0.524
1.307	23.42	0.553	2.013	23.43	0.519
1.415	23.92	0.555			

Figure 7.1. Total cross section of $ZrH_{1.85}$.

very low energy (below 0.01 eV), where the multiple scattering in the sample seems to be underestimated.

7.2 ANALYSIS OF SPECTRAL MEASUREMENTS

During this year, there has been little analysis of spectral measurements performed previously under this program, though some analysis is planned for next year. However, of special interest to the reactor design community are the position-dependent measurements made two years ago across the near-critical ^{235}U sphere (APFA-III), and a minor effort has been spent in analyzing these data further.⁽⁵⁶⁾ The lithium glass detector used in the measurements has been calibrated under another program. This calibration has been described in a report⁽⁷⁾ and in an article submitted for publication to Nuclear Instruments and Methods. The calibration was incorporated into the reduction of ^{235}U spectral data and a report on this work is now in preparation. A brief write-up was presented in an informal reactor physics session at the Seattle ANS meeting and is summarized here. The spectral measurements were performed by the time-of-flight techniques at six internal positions in the geometry illustrated in Fig. 7.2.

The following observations arose from these studies:

1. It was shown analytically that the neutron spectrum above 2-3 MeV was essentially equal to the fission spectrum. The observed data (Fig. 7.3) provided an excellent parametric fit to a Maxwellian distribution with a $\bar{T}$ of 1.318 ± 0.005 MeV. This value is in agreement with the Barnard, et al.⁽⁵⁷⁾ value of 1.30 ± 0.01 MeV and Sherwood and King's⁽⁵⁸⁾ observation of 1.30 ± 0.02 MeV. The ENDF/B⁽⁵⁹⁾ recommendation of 1.273 MeV for neutrons below 10 MeV and 1.227 MeV for neutrons above 10 MeV appears to be too low.
2. The use of ENDF/B cross sections in a S_{16} transport theory calculation predicts k_{eff} that is 2.4% too large.

Figure 7.2. Geometry for spectral studies in the ^{235}U sphere
(measurement positions are numbered in the figure).

Figure 7.3. High energy neutron spectrum $N(E)$ from position 3 in APFA III.

3. The neutron spectra from a k calculation are consistently "softer" than the measured data. If the measured spectra at the reactor center are used to calculate fission rate ratios, a reasonable agreement with Hansen's (60) data is obtained, as shown in Table 3.

Table 3
CALCULATED AND MEASURED FISSION RATE RATIOS AT
REACTOR CENTER

Item	Measured ⁽⁶⁰⁾ Ratios	Calculated Ratios	
		With Measured Flux	With Calculated Flux
$^{235}\text{U}/^{238}\text{U}$	6.5	6.58	7.35
$^{237}\text{Np}/^{238}\text{U}$	5.1	5.57	5.96

8. ADDITION OF THE SHORT COLLISION APPROXIMATION TO THE FLANGE CODE

It has been standard practice to use the code GASKET⁽¹⁾ to compute scattering laws up to energy exchanges of 2-3 eV. These scattering laws are then converted by the code FLANGE⁽⁶¹⁾ to quantities of direct interest, like cross sections and Legendre moments. When the original version of this code is used to compute these quantities for energy exchanges higher than the maximum computed by GASKET, the results are wrong (see Fig. 8.1), because FLANGE arbitrarily sets to zero the value of the scattering law outside the scattering matrix supplied by GASKET.

To eliminate this difficulty FLANGE was modified to compute the scattering law by the short collision time approximation whenever its value is not supplied by GASKET. The validity of the short collision time approximation⁽⁴⁵⁾ in regard to scattering laws has already been discussed. It has been shown previously⁽⁶²⁾ that for large values of β (where β = energy transfer/temperature) the short collision time expression is a good approximation to the exact scattering law in the region around the point $\alpha = \beta$, that is to say the region where the scattering law, $S(\alpha, \beta)$, assumes the largest values. It was further shown that where disagreement exists, the scattering law tends to have negligible values. For these reasons the short collision expression is an acceptable approximation in the region where GASKET calculations have not supplied a value of the scattering law.

The short collision time approximation option in GASKET is used for the continuous modes only, the resulting $S(\alpha, \beta)$ being subsequently convolved with the scattering law corresponding to the discrete modes of

Figure 8.1. Example of double differential cross section of H_2O with and without the s.c.t. approximation.

vibration. The approximate form of the scattering law introduced in the modified FLANGE is calculated completely by short collision and is given by

$$S_{sc}(\alpha, \beta) = e^{\beta/2} \frac{e^{-\frac{T}{4\alpha\bar{T}}(\beta + \alpha)^2}}{\sqrt{4\pi\alpha\frac{\bar{T}}{T}}} \quad (8.1)$$

where T is the actual temperature and $\bar{T}$ is the effective temperature (see Section 6), and α and β are respectively the dimensionless square momentum and energy transfers given by Eq. 2.5. This expression is valid for downscattering only, since it does not satisfy the detailed balance condition. Therefore, a double differential cross section $\sigma(E_i \rightarrow E_f, \mu)$ satisfying detailed balance is obtained in the following way:

$$\sigma(E_i \rightarrow E_f, \mu) = \frac{\sigma_b}{4\pi T} \sqrt{\frac{E_f}{E_i}} e^{-\beta/2} S_{sc}(\alpha, -|\beta|) \quad (8.2)$$

The justification given for using this form, which is the right short collision expression for downscattering ($\beta < 0$) and which satisfies detailed balance because it involves a symmetric scattering law, is that the approximation will be used for large values of $|\beta|$, where the up-scattering is negligible. The only input quantity which is not available to the original version of FLANGE is the effective temperature. Therefore, in the modified version of the code, an additional card has to be inserted after the first card of the regular input.⁽⁶¹⁾ This card carries the effective temperature in eV in format E10.5. The main structure of the code, as well as the output format, has not been altered. The modifications are briefly described here.

All the routines of FLANGE, except one (subroutine DIXN1), compute the required value of the scattering law by interpolating over the α and β mesh provided by GASKET. The interpolation is performed by the subroutine TERP. The short collision time expression has

therefore been introduced in the subroutine TERP as an alternative to the interpolation in the case in which the required value falls outside the range of the GASKET mesh. A reshuffle of exponential factors was required in TERP and all subroutines which use TERP in order to avoid a floating point overflow which arises in the original version of the code when relatively high values of energy are used.

The subroutine DIXN1, which computes the Legendre moments of the kernel by making an exact integration over the variable α , does not require the subroutine TERP; therefore the short collision expression had also to be inserted in DIXN1.

Before the integration, the subroutine DIXN1 performs an interpolation over the variable β . In order to keep unchanged the integration scheme it was found convenient to introduce an additional mesh of thirty α points, logarithmically spaced between $\alpha_{\max}$ provided by GASKET and a value of α which has been arbitrarily set to 3000. This additional set of integration points includes a range in α ample to cover cases of practical interest. The value of the double differential cross section at the points of this extended mesh is obtained either by interpolation over β or by formulas 8.1 and 8.2.

Figures 8.1 to 8.5 show comparisons of quantities calculated with the original and with the modified codes. All the quantities are referred to one hydrogen atom in water and the calculations have been performed using a room temperature Haywood II scattering kernel⁽⁶³⁾ extended to 3 eV.

Figure 8.1 shows the double differential cross section for an incident energy of 4 eV and $\mu = 0.5$, μ being the cosine of the scattering angle. It can be seen that the original FLANGE completely misses the part of the curve below a final energy of 1 eV. Also, it can be seen that slight discontinuity exists at the point where the short collision approximation joins the values calculated by interpolation of the GASKET values. Since the GASKET values in this particular region were also computed by short

Figure 8.2. Example of angular single differential cross section of H_2O with and without the s. c. t. approximation.

Figure 8.3. Example of zeroth Legendre moment of the H₂O kernel with and without the s. c. t. approximation.

Figure 8.4. Example of first Legendre moment of the H_2O kernel with and without the s.c.t. approximation.

Figure 8.5. Total cross section of H₂O computed with and without the s.c.t. approximation.

collision, the reason why the two curves do not join smoothly is that, as mentioned before, only the distributed modes are handled by short collision in GASKET. This residual discontinuity should be of no importance in practical calculations.

Figure 8.2 shows the angular single differential cross section for an incident energy of 4 eV. It is seen that the original FLANGE underestimates the value of the cross section.

Figures 8.3 and 8.4 show the behavior with final energy of the zero-th and first Legendre moments for an incident energy of 4 eV. The difference between the results of the two codes is similar to the one observed in the double differential cross section.

The improvement due to the short collision addition is also visible in the total cross section, Fig. 8.5, where a rapidly dropping tail is replaced by a smooth curve with the correct behavior.

9. ENDF ACTIVITIES

9.1 UC, UO_2 AND CH_2 KERNELS

The thermal inelastic scattering law for U and C bound in UC has been computed in the incoherent approximation using the code GASKET.⁽¹⁾ The weighted frequency spectra input to GASKET were obtained from the central force lattice dynamical model discussed in Section 3. The U and C data were combined to form a kernel for UC with 50 α values and 80 β values covering the energy range to 1 eV. The coherent elastic cross section of UC was computed using the code QSCAT, discussed in Section 4.

Data generated and sent to NNCSC* are for temperatures 296°, 400°, 500°, 600°, 700°, 800°, 1000° and 1200°K. The code SEND, discussed in Section 9.2.2, was used to prepare the data in the ENDF/B format.

Thermal scattering data for UO_2 has also been resubmitted to NNCSC. The data sent previously in the ENDF/A format included incoherent elastic cross sections. With the use of the codes SEND and QSCAT a new data tape was prepared in the ENDF/B format with coherent elastic scattering cross sections.

The thermal inelastic scattering law for H bound in CH_2 has also been computed using GASKET. The weighted frequency spectrum input for GASKET was obtained from the frequencies and amplitude vectors calculated by Sprevak and Koppel.⁽⁵⁾ The elastic cross section for H was computed in the incoherent approximation. The carbon atoms were treated as a free gas of mass 12. The scattering law is given for 80 α values covering the energy range to 1.5 eV for 296°K and 350°K.

---

*National Neutron Cross Section Center

9.2 MISCELLANEOUS ACTIVITIES

9.2.1 Kernel Book⁽²⁹⁾

The documentation of the thermal scattering data submitted to the NNCSC continued this year with completion of the documentation for UO_2 ,⁽²⁸⁾ BeO ,⁽⁶⁴⁾ and CH_2 .⁽⁵⁾

9.2.2 Code SEND

In the past, when thermal scattering data were submitted under this project to the NNCSC at Brookhaven the elastic data were in the ENDF/B format and the inelastic in the ENDF/A format. A new code, SEND, has been written to read inelastic data from BCD tapes, combine this with the elastic data and prepare a new tape compatible with the currently distributed ENDF/B tapes.

Inelastic data generated in the future will still be in the A format but SEND will be used to convert these data to the B format. SEND may also be used to convert previously submitted tapes.

9.2.3 Code Dissemination

Updated versions of the codes FLANGE I and GASKET (see Section 8) have been sent to the Argonne Code Center. The modification to FLANGE allows for calculation of scattering cross sections in the short collision approximation at energy transfers for which GASKET calculations have not been performed. This modification is discussed in Section 8.1. The updated version of GASKET includes the capability of describing diffusive modes.⁽⁶⁵⁾

REFERENCES

1. J. U. Koppel, J. R. Triplett and Y. D. Naliboff, "GASKET, A Unified Code for Thermal Neutron Scattering," USAEC Report GA-7480, General Dynamics Corporation, General Atomic Division (1966).
2. The following papers were presented at the 25th Annual Meeting of the Physical Society of Argentina (Asociación Física Argentina) in La Plata, Argentina, July 1969

G. M. Borgonovi and D. Sprevak - Dispersion Coherente de neutrones en Berilio. (Coherent Scattering of Neutrons in Beryllium.)

D. Sprevak and J. U. Koppel - Dispersion de neutrones por difenilo. (Scattering of neutrons by diphenyl.)

D. Sprevak and J. U. Koppel - Dispersion de neutrones por bencens. (Scattering of neutrons by benzene.)

D. Sprevak and J. U. Koppel - Dispersion de neutrones por polietileno. (Scattering of neutrons by polyethylene.)
3. E. L. Slaggie, "Thermal Expansion and Phonon Frequency Shifts in Non-Primitive Lattices," Bulletin of the American Physical Society, 14 498 (1969).
4. J. M. Neill and J. C. Young, "Thermal Neutron Spectra in Slab and Cylindrical Geometries," Nukleonik 12 (1968) 18.
5. D. Sprevak and J. U. Koppel, "Neutron Scattering by Polyethylene," Nukleonik 12, 87, January 1969.
6. E. L. Slaggie, "Central Force Lattice Dynamical Model for Uranium Carbide," USAEC Report GA-8675, Gulf General Atomic Incorporated, November 14, 1968.

7. J. M. Neill, D. Huffman, C. A. Preskitt, and J. C. Young, "Calibration and Use of a 5-in. Diameter Lithium Glass Detector," USAEC Report GA-9048, Gulf General Atomic Incorporated, December 1968.
8. E. L. Slaggie, "Formal Expansions for the Potential Energy of a Distorted Crystal in Terms of Strain Parameters and Components of a Static Electric Field," USAEC Report GA-9162, Gulf General Atomic Incorporated, January 6, 1969.
9. E. L. Slaggie, "Thermal Expansion and Phonon Frequency Shifts," USAEC Report GA-9417, Gulf General Atomic Incorporated (1969).
10. G. M. Borgonovi, "Coherent Scattering Law for Polycrystalline Beryllium," USAEC Report GA-9364, Gulf General Atomic Incorporated, May 16, 1969.
11. G. M. Borgonovi and D. Sprevak, "Calculation of Coherent Neutron Scattering from Polycrystalline Beryllium," USAEC Report GA-9705, Gulf General Atomic Incorporated, September 1969.
12. P. A. Egelstaff, "The Inelastic Scattering of Cold Neutrons by Crystals," AERE N/R 1164, 1958.
13. L. J. Slutsky and C. W. Garland, "Lattice Dynamics of Hexagonal Close-Packed Metals," J. Chem. Phy. 26, 878 (1947); ibid 28, 331 (1958).
14. I. Waller and P. O. Fröman, "On Neutron Diffraction Phenomena According to the Kinematical Theory I," Arkiv for Fysik, 4, 183, 331, (1958).
15. J. R. Beyster, "Neutron Scattering From Light Water," Nucl. Sci. Eng. 31, 254 (1968).
16. D. Sprevak, G. M. Borgonovi, J. M. Neill and G. W. Carriveau, "Neutron Thermalization in Benzene," Nukleonik, 11, 233 (1968).
17. G. W. Carriveau, "Single Differential Cross Section of Zirconium Hydride," USAEC Report GA-8345, General Dynamics Corp., General Atomic Division, June 1967.
18. G. M. Borgonovi, G. W. Carriveau, "Neutron Scattering from MgO," Phys. Rev. 174, 953 (1968).

19. J. A. Young and J. U. Koppel, "Coherent Inelastic Scattering from Polycrystalline Beryllium," USAEC Report GA-6238, General Dynamics Corporation, General Atomic Division, March, 12, 1965.
20. N. Kroo, G. Borgonovi, K. Sköld and K. E. Larsson, "Inelastic Scattering of Neutrons from Condensed Argon," Swedish Atomic Energy Commission Report No. RFX-326, 1964.
21. F. W. deWette and Aneesur Rahman, "Inelastic Scattering of Neutrons by Polycrystals," Phys. Rev. 176, 784 (1968).
22. J. C. Warner (Ed.), Metallurgy of Uranium and Its Alloys, USAEC (1955).
23. S. N. Purohit, et al., "Inelastic Neutron Scattering in Metal Hydrides, UC and UO₂, and Applications of the Scattering Law," published in "Neutron Thermalization and Reactor Spectra," Proceedings, IAEA Conference, Ann Arbor, Michigan, July 1967.
24. J. U. Koppel, "Neutron Scattering by Hydrogenous Moderators," USAEC Report GA-7055, General Dynamics Corporation, General Atomic Division (1966).
25. E. L. Slaggie, "Central Force Lattice Dynamical Model for Zirconium Hydride," J. Phys. Chem. Solids, 29, 923 (1968).
26. E. L. Slaggie, "Central Force Lattice Dynamical Model for Uranium Carbide," USAEC Report GA-8675, Gulf General Atomic Incorporated, November 14, 1968.
27. W. M. Lomer and G. G. Low, in Thermal Neutron Scattering, edited by P. A. Egelstaff (Academic Press, 1965) p. 19.
28. J. A. Young, "Neutron Scattering from Uranium Dioxide," USAEC Report GA-8760, Gulf General Atomic Incorporated (1968). (Published in Nukleonik, 12, 205 (1969).
29. J. U. Koppel and D. H. Houston, "Reference Manual for ENDF Thermal Neutron Scattering Data," USAEC Report GA-8774, Gulf General Atomic Incorporated, December 16, 1968.
30. E. L. Slaggie, "Central Force Lattice Dynamical Model for Zirconium Hydride," General Atomic Report GA-8132, July 29, 1967.
31. C. P. Kempter, R. O. Elliott, and K. A. Gschneider, Jr., "Thermal Expansion of Delta and Epsilon Zirconium Hydrides," J. Chem. Phys. 33, 837 (1960).

32. A. A. Maradudin and A. E. Fein, "Scattering of Neutrons by an Anharmonic Crystal," Phys. Rev. 129, 2589 (1962).
33. R. A. Cowley, "Lattice Dynamics of an Anharmonic Crystal," Advances in Physics 12, 421 (1963).
34. K. E. Larsson, U. Dahlborg, S. Holmryd, "A Study of Some Temperature Effects on the Phonons and Aluminum by Use of Cold Neutrons," Arkiv. Fysik 17, 369 (1960).
35. B. N. Brockhouse, T. Arase, G. Caglioti, M. Sakamoto, R. N. Sinclair and A. D. B. Woods, Inelastic Scattering of Neutrons in Solids and Liquids (IAEA, Vienna, 1961), p. 531.
36. D. H. Perkel, "A Family of Thermalization Codes," General Atomic Report GAMD-754 (Rev.), July 1, 1959.
37. B. Kopelman, Materials for Nuclear Reactors, (McGraw-Hill, New York, 1959), p. 370.
38. A. Yoshimori and Y. Kitano, "Theory of the Lattice Vibration of Graphite," J. Phys. Soc. of Japan 11, 352 (1956).
39. J. A. Young and J. U. Koppel, "Phonon Spectrum of Graphite," J. Chem. Phys. 42, 357 (1965).
40. H. Takahashi, preprint. (To be published in Nucl. Sci. and Eng.)
41. M. Born and K. Huang, Dynamical Theory of Crystal Lattices, (Oxford, 1954), p. 245 ff.
42. H. Bethe and G. Placzek, "Resonance Effects in Nuclear Processes," Phys. Rev. 51, 540 (1937).
43. W. E. Lamb, "Capture of Neutrons by Atoms in a Crystal," Phys. Rev. 55, 190 (1939).
44. L. Dresner, Resonance Absorption in Nuclear Reactors, Pergamon Press (1960).
45. M. S. Nelkin and D. E. Parks, "Effects of Chemical Binding on Nuclear Recoil," Phys. Rev. 119, 1060 (1960).
46. See for example C. R. Adkins, P. J. Persiani, R. N. Hwang and J. J. Kaganove, "The Chemical Binding Effects on the Resonance Line Shapes of ^{238}U in a UO_2 Lattice," Proceedings of the Conf. on Neutron Cross Section Technology, March 22-24, 1966, Washington, D. C. (CONF 660303).

47. Brother A. Bernabei, "The Effects of Crystalline Binding on the Doppler Broadening of a Neutron Resonance," Brookhaven National Laboratory Report BNL-860 (T-344), June 1964.
48. Alf Sjölander, "Multi-Phonon Processes in Slow Neutron Scattering," Arkiv. Fysik 14, 315 (1958).
49. A. A. Maradudin and S. H. Vosko, "Symmetry Properties of the Normal Vibrations of a Crystal," Rev. Modern Phys. 40, 1 (1968).
50. P. A. Egelstaff and P. Schofield, "On the Evaluation of the Thermal Neutron Scattering Law", Nucl. Sci. Eng. 12, 260 (1962).
51. H. E. Jackson and J. E. Lynn, "Resonant Absorption of Neutrons by Crystals," Phys. Rev. 127, 461 (1962).
52. D. Sprevak, G. M. Borgonovi, J. M. Neill, and G. W. Carriveau, "Neutron Thermalization in Benzene," Nukleonik, 11, 233 (1968).
53. G. M. Borgonovi and G. W. Carriveau, "Total Cross Section of Uranium Dicarbide," USAEC Report GA-8782, Gulf General Atomic Incorporated, July 18, (1968).
54. J. L. Russell, Jr., J. M. Neill and J. R. Brown, "Total Cross Section Measurements of H₂O," USAEC Report GA-7581, General Dynamics Corporation, General Atomic Division, December 1966.
55. J. M. Neill, et al., Proceedings of the Conference on Neutron Cross Sections and Technology, Vol. II, p. 1183, Washington, D. C., March 1968.
56. J. M. Neill, K. L. Crosbie and J. L. Russell, Jr., "Neutron Spectrum Measurements Across a Bare ²³⁵U Metal Sphere," USAEC Report GA-9284, Gulf General Atomic Incorporated, May 1969.
57. E. Barnard, et al., "Time-of-Flight Measurements of Neutron Spectra from the Fission of ²³⁵U, ²³⁸U, and ²³⁹Pu," Nucl. Phys. 71, 228 (1965).
58. G. G. Sherwood and J. S. King, "²³⁵U Fission Neutron Spectrum 6-20 MeV," Trans. Amer. Nucl. Soc. 10, 555 (1967).
59. C. R. Lubitz and P. P. Syzdlik, "²³⁵U Cross Sections for the ENDF/B," KAPL, December 1966.

60. G. E. Hansen, "Properties of Elementary Fast Critical Assemblies," Proceedings 2nd International Conference on Peaceful Uses of Atomic Energy, Geneva, 12, 84-88 (1958).
61. FLANGE, Appendix E to USAEC Report GA-6824, "Integral Neutron Thermalization Annual Summary Report, October 1, 1964 through September 30, 1965." General Dynamics Corporation, General Atomic Division (1965).
62. J. R. Beyster, et al., "Integral Neutron Thermalization, Annual Summary Report, October 1, 1967 through September 30, 1968," USAEC Report GA-9036, Gulf General Atomic Incorporated, October 30, 1968.
63. J. U. Koppel, "Neutron Scattering by Hydrogenous Moderators," USAEC Report GA-7055, General Dynamics Corporation, General Atomic Division, March 1965.
64. G. M. Borgonovi, "Lattice Dynamics and Neutron Scattering of BeO," USAEC Report GA-8758, Gulf General Atomic Incorporated, (1968).
65. J. R. Beyster, et al., "Integral Neutron Thermalization, Annual Summary Report, Oct. 4, 1966 through Sept. 30, 1967," USAEC Report GA-8280, General Dynamics Corporation, General Atomic Division (1967).