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SUBJECT: A Review of the Mathematical Formulation of the
Problem of Inelastic Neutron Scattering by
Polycrystalline Material

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FROM: C. A. Preskitt

Abstract

A self-contained development of the formulation of the problem of inelastic neutron scattering by crystals is presented. Although nearly all of the methods and results used in the present discussion have appeared in a variety of previously published papers, additional mathematical details are given here which have been found by the author to aid in understanding the final results.

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I. Introduction

In order to initiate a study of the effect of crystalline binding on the thermal neutron spectrum in crystalline moderators, it has been necessary for the author to familiarize himself with standard formulations of the problem. The treatment by Zemach and Glauber¹ was found to provide the most convenient formalism, but in order to understand the subject as fully as possible and to provide a basis for simplifying or extending the treatment, it was found desirable to proceed from the particle potentials to the scattering cross section in somewhat more detail than could be expected in their paper. The present document thus supplies details not commonly found in the usual publications on the subject and is distributed in this rough form in the hope that these details will be of help to anyone interested in understanding the final results.

A brief quantum mechanical treatment of the harmonic oscillator problem precedes the treatment of the scattering and provides the basis for the description of the modes of the crystal lattice vibrations.

The concluding section obtains the inverse mass expansion of the cross section from the general treatment to show the basis for the heavy crystal approximation.

II. Review of Harmonic Oscillator Problem

In order to obtain an orientation for the formalism to be used later we first study the quantum mechanical solution of the harmonic oscillator problem.

We write the Hamiltonian of the system as

$$H = \frac{1}{2m} (p^2 + m^2 \omega^2 q^2) \quad (2.1)$$

where p and q are operators with the following commutation relations

$$[q_i, p_j] = i\hbar \delta_{ij} \quad (2.2)$$

That is, p and q for the same oscillator do not commute.

The equations of motion can then be written as

$$i\hbar \dot{q} = [q, H] \quad i\hbar \dot{p} = [p, H]$$

so that

$$i\hbar \dot{q} = \frac{1}{2m} \{ [q, p^2] + m^2 \omega^2 [q, q^2] \}$$

But

$$[q, p^2] = -[p^2, q] = -\{p[p, q] + [p, q]p\} = 2i\hbar p$$

and

$$[q, q^2] = 0$$

so that

$$\dot{q} = p/m \quad (2.3)$$

Similarly, we find that

$$\dot{p} = -m\omega^2 q \quad (2.4)$$

Now construct two operators, a and a^* defined as

$$a = \frac{1}{\sqrt{2\hbar m\omega}} (p - im\omega y) \quad (2.5)$$

$$a^* = \frac{1}{\sqrt{2\hbar m\omega}} (p + im\omega y) \quad (2.6)$$

Then by using (2.3) and (2.4) above we get

$$\dot{a} = -i\omega a$$

$$\dot{a}^* = i\omega a^*$$

so that $a(t) = a_0 e^{-i\omega t}$ (2.7)

and $a^*(t) = a_0^* e^{i\omega t}$ (2.8)

The properties of these operators can be seen by the following development:

$$\hbar\omega a^*a = \frac{1}{2m} \{ p^2 + m^2\omega^2 y^2 + im\omega [y, p] \}$$

or $\hbar\omega a^*a = H - \hbar\omega/2$ (2.9)

and in a similar fashion

$$\hbar\omega a a^* = H + \hbar\omega/2 \quad (2.10)$$

from which we see that $[a, a^*] = 1$. Or in general if these operators are assigned to different oscillators

$$[a_i, a_j^*] = \delta_{ij} \quad (2.11)$$

Also $aH - Ha = i\hbar\dot{a} = \hbar\omega a$

and $a^*H - Ha^* = i\hbar\dot{a}^* = -\hbar\omega a^*$

so that $a(H - \hbar\omega) = Ha$ (2.12)

$$a^*(H + \hbar\omega) = Ha^* \quad (2.13)$$

If now E' is an eigenvalue of H , then we can write

$$H\psi(E') = E'\psi(E') \quad (2.14)$$

so that $a(H - \hbar\omega)\psi(E') = Ha\psi(E')$

$$a(E' - \hbar\omega)\psi(E') = Ha\psi(E')$$

Or, $H\{a\psi(E')\} = (E' - \hbar\omega)\{a\psi(E')\}$ (2.15)

That is, if $\psi(E')$ is an eigenvector with eigenvalue E' , then $\{a\psi(E')\}$ is also an eigenvector and has eigenvalue $E' - \hbar\omega$. Similarly

$$a^*(H + \hbar\omega) = Ha^*$$

gives

$$H\{a^*\psi(E')\} = (E' + \hbar\omega)\{a^*\psi(E')\} \quad (2.16)$$

It is elementary to continue the process to get

$$H\{aa\psi(E')\} = (E' - 2\hbar\omega)\{aa\psi(E')\}, \text{ etc.}$$

and

$$H\{a^*a^*\psi(E')\} = (E' + 2\hbar\omega)\{a^*a^*\psi(E')\}$$

so that the operator a is properly termed a destruction operator since it decreases the number of quanta by one, and a^* may be called a creation operator since it increases the number of quanta by one.

The eigenvalues of the problem are seen to go like the following sequence $E = \dots, E' - 2\hbar\omega, E' - \hbar\omega, E', E' + \hbar\omega, E' + 2\hbar\omega, \dots$

Since $\hbar \omega a^* a = H - \hbar \omega/2$ and H is a positive definite quantity, then the lowest eigenvalue occurs when $p = q = 0$. This gives $a = a^* = 0$ and $H = \hbar \omega/2$ as the lowest eigenvalue. The sequence of eigenvalues thus is

$$\frac{\hbar \omega}{2}, \frac{3\hbar \omega}{2}, \frac{5\hbar \omega}{2}, \dots$$

or

$$E_n = (n + \frac{1}{2}) \hbar \omega \quad (2.17)$$

so that $H \psi_n = E_n \psi_n$ is a convenient notation for the eigenvalue equation.

The matrix elements of various combinations of the a and a^* can be found in the following manner using (2.9):

$$H = \hbar \omega a^* a + \frac{\hbar \omega}{2} \quad \text{and} \quad H \psi_n = (n + \frac{1}{2}) \hbar \omega \psi_n$$

so that

$$\hbar \omega (a^* a + \frac{1}{2}) \psi_n = (n + \frac{1}{2}) \hbar \omega \psi_n$$

or

$$a^* a \psi_n = n \psi_n \quad (2.18)$$

with the result that $(\psi_m, a^* a \psi_n) = n(\psi_m, \psi_n) = n \delta_{nm}$.

Thus, $(a^* a)$ is diagonal and has elements

$$(a^* a)_{nn} = n \quad (2.19)$$

and in a similar manner using (2.10) it can be shown that

$$(a a^*)_{nn} = n + 1 \quad (2.20)$$

Since we have already shown by (2.15) and (2.16) that

$$a \psi_{n+1} = \alpha \psi_n, \quad a^* \psi_n = \beta \psi_{n+1}$$

it follows that $\alpha = (\psi_n, a \psi_{n+1})$

and $\beta = (\psi_{n+1}, a^* \psi_n) = (a \psi_{n+1}, \psi_n) = (\psi_n, a \psi_{n+1})^*$

so that $\alpha = \beta^*$.

Furthermore $a^* a \psi_{n+1} = a^* \alpha \psi_n = \alpha a^* \psi_n = \alpha \beta \psi_{n+1}$

$$\text{or } a^* a \psi_{n+1} = |\alpha|^2 \psi_{n+1} \quad (2.21)$$

Comparing (2.21) with (2.18) shows that

$$|\alpha|^2 = n+1$$

$$\text{so } \alpha = \sqrt{n+1} \quad (2.22)$$

$$\text{Thus } a \psi_{n+1} = \sqrt{n+1} \psi_n \quad (2.23)$$

$$\text{and } a^* \psi_n = \sqrt{n+1} \psi_{n+1} \quad (2.24)$$

We see that a and a^* have non-zero matrix elements only in the immediately off diagonal positions.

$$a = \begin{pmatrix} 0 & \sqrt{1} & 0 & 0 \\ 0 & 0 & \sqrt{2} & 0 \\ 0 & 0 & 0 & \sqrt{3} & \dots \\ 0 & 0 & 0 & 0 & \vdots \end{pmatrix} \quad a^* = \begin{pmatrix} 0 & 0 & 0 & 0 \\ \sqrt{1} & 0 & 0 & 0 \\ 0 & \sqrt{2} & 0 & 0 & \dots \\ 0 & 0 & \sqrt{3} & 0 & \vdots \end{pmatrix} \quad a^* a = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 0 & 0 & 3 & 0 & \dots \\ 0 & 0 & 0 & 4 & \vdots \end{pmatrix}$$

It will also be useful in what follows to have the coordinate operator expressed in terms of the a and a^* . Since

$$a = \frac{1}{\sqrt{2\hbar m \omega}} (p - i m \omega q)$$

and $a^* = \frac{1}{\sqrt{2\hbar m\omega}} (p + im\omega q)$

it follows that

$$a^* - a = \frac{2im\omega}{\sqrt{2\hbar m\omega}} q$$

or $q = i\sqrt{\frac{\hbar}{2m\omega}} (a - a^*)$ (2.25)

and with expressions (2.7) and (2.8) for $a(t)$ and $a^*(t)$ we can write the explicit time dependence of q to be:

$$q(t) = i\left(\frac{\hbar}{2m\omega}\right)^{\frac{1}{2}} (a_0 e^{-i\omega t} - a_0^* e^{i\omega t})$$
 (2.26)

III. Scattering of Neutrons by Polycrystalline Material

In this section expressions will be developed for the cross section for inelastic scattering of neutrons by a crystal. In general the treatment follows that of Zemach and Glauber¹ for scattering by molecules except that considerably more detail is supplied.

The interaction of a neutron with a crystal atom is represented in the Fermi pseudopotential approximation² as*

$$V(\underline{r}_n, \underline{r}_\nu) = \left(\frac{2\pi a_\nu}{m}\right) \delta(\underline{r}_n - \underline{r}_\nu)$$
 (3.1)

where a_ν is the bound scattering length and $\underline{r}_n$ and $\underline{r}_\nu$ are the coordinate vectors of the neutron and the ν^{th} nucleus respectively. The potential which is experienced by the neutron in traversing a crystal with N identical atoms is then

$$V(\underline{r}_n) = \sum_{\nu=1}^N V(\underline{r}_n, \underline{r}_\nu) = \frac{2\pi a}{m} \sum_{\nu=1}^N \delta(\underline{r}_n - \underline{r}_\nu)$$
 (3.2)

* A system of units with $\hbar = K = 1$ is employed throughout.

Using first order perturbation theory the scattering cross section is written as

$$\sigma_{fi}(\theta) = \left(\frac{m}{2\pi}\right)^2 \frac{k}{k_0} \left| \langle \Psi_f | V | \Psi_i \rangle \right|^2 \quad (3.3)$$

where k_0 and k are the initial and final neutron momenta, and Ψ_f and Ψ_i are the total initial and final wave functions of the system of neutron plus crystal. We write $\Psi = \phi \psi$, where ϕ is the neutron wave function and ψ is the crystal wave function, and assume that the Born approximation is valid so that plane waves are used for ϕ . We then get

$$\sigma_{fi}(\theta) = a^2 \frac{k}{k_0} \left| \langle \psi_f | \int e^{-i(\underline{k}-\underline{k}_0) \cdot \underline{r}_n} \sum_v \delta(\underline{r}_n - \underline{r}_v) d\underline{r}_n | \psi_i \rangle \right|^2 \quad (3.4)$$

Due to the presence of the delta function the integration over the neutron coordinates can be performed immediately, the result being

$$\sigma_{fi}(\theta) = a^2 \frac{k}{k_0} \left| \langle \psi_f | \sum_v e^{-i(\underline{k}-\underline{k}_0) \cdot \underline{r}_v} | \psi_i \rangle \right|^2 \quad (3.5)$$

The sum over v brings in terms involving $\underline{r}_v - \underline{r}_v$, when we square the matrix element. In all that follows, these interference terms will be neglected. This procedure is not, in general, valid in calculating the scattering from molecules since only a few such terms will appear. For a crystal, where N is large, neither can we expect to get a proper result for elastic scattering, which is predominantly an interference effect, but the result will still be good for the inelastic scattering (i.e., collisions in which the state of excitation of the crystal is changed). With this approximation (3.5) becomes

$$\sigma_{fi}(\theta) = a^2 \frac{k}{k_0} \sum_v \langle \psi_f | e^{-i\underline{k} \cdot \underline{r}_v} | \psi_i \rangle \langle \psi_i | e^{i\underline{k} \cdot \underline{r}_v} | \psi_f \rangle \quad (3.6)$$

where $\underline{K} = \underline{k} - \underline{k}_0$ is the momentum transfer in the collision.

If E_i and E_f are the initial and final energies of the crystal state, then the states f and i in the above expression are restricted by energy conservation and the requirement that the final neutron energy must be non-negative.

If $\epsilon = \frac{1}{2m}(k^2 - k_0^2)$ is the energy change of the neutron, then

$$\epsilon = E_f - E_i$$

and $\epsilon \geq -k_0^2/2m$.

Since the various crystal states are not distinguished experimentally, that is, only $\sigma(\epsilon, \theta)$ is a measurable quantity, we must sum over all of the admissible final crystal states and average over the distribution of initial states to yield the desired cross section from (3.6). The conservation requirements stated above are automatically included if the sum over final states is given by

$$\sigma_i(\theta) = \sum_f \int_{-\frac{k_0^2}{2m}}^{\infty} d\epsilon \sigma_{fi}(\theta) \delta(E_i - E_f - \epsilon) \quad (3.7)$$

Using the Fourier transform of the delta function in the form

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{-i\alpha t} e^{i\beta t} = \delta(\beta - \alpha)$$

this can be written as

$$\sigma_i(\theta) = \frac{1}{2\pi} \sum_f \int_{-\frac{k_0^2}{2m}}^{\infty} d\epsilon \int_{-\infty}^{\infty} dt e^{-i\epsilon t} e^{i(E_i - E_f)t} \sigma_{fi}(\theta) \quad (3.8)$$

Expression (3.8) can be simplified and the final state sum performed if we note that

$$e^{iE_i t} \psi_f = (1 + iE_i t - \frac{1}{2} E_i^2 t^2 + \dots) \psi_f = (1 + iHt - \frac{1}{2} H H t^2 + \dots) \psi_f$$

$$\text{or } e^{iE_i t} \psi_f = e^{iHt} \psi_f \quad (3.9)$$

where we always understand that an operator in the exponent has meaning only when the exponential is expanded so that the operator may act on a wave function. Using results like (3.9) we then get

$$\begin{aligned} \sigma_{fi}(\theta) = & \frac{a^2}{2\pi} \sum_{\nu} \int dt \int d\epsilon \frac{k}{k_0} e^{-i\epsilon t} \\ & \times \sum_f \langle \psi_i | e^{iHt} e^{i\mathbf{K} \cdot \mathbf{r}_\nu} e^{-iHt} | \psi_f \rangle \langle \psi_f | e^{-i\mathbf{K} \cdot \mathbf{r}_\nu} | \psi_i \rangle \end{aligned} \quad (3.10)$$

This can be summed over the final states since we now have only a matrix product of the form

$$\sum_j A_{ij} B_{jk} = (AB)_{ik}$$

$$\begin{aligned} \text{Thus, } \sum_f \langle \psi_i | e^{iHt} e^{i\mathbf{K} \cdot \mathbf{r}_\nu} e^{-iHt} | \psi_f \rangle \langle \psi_f | e^{-i\mathbf{K} \cdot \mathbf{r}_\nu} | \psi_i \rangle \\ = \langle \psi_i | e^{iHt} e^{i\mathbf{K} \cdot \mathbf{r}_\nu} e^{-iHt} e^{-i\mathbf{K} \cdot \mathbf{r}_\nu} | \psi_i \rangle \end{aligned}$$

so that (3.10) becomes

$$\sigma_i(\theta) = \frac{a^2}{2\pi} \sum_{\nu} \int dt \int d\epsilon \frac{k}{k_0} e^{-i\epsilon t} \langle \psi_i | e^{iHt} e^{i\mathbf{K} \cdot \mathbf{r}_\nu} e^{-iHt} e^{-i\mathbf{K} \cdot \mathbf{r}_\nu} | \psi_i \rangle$$

If we do not wish to integrate out the energy transfer, this is written as

$$\sigma_i(\theta, \epsilon) = \frac{a^2}{2\pi} \frac{k}{k_0} \sum_{\nu} \int_{-\infty}^{\infty} dt e^{-i\epsilon t} \langle \psi_i | e^{iHt} e^{i\mathbf{k} \cdot \mathbf{r}_{\nu}} e^{-iHt} e^{-i\mathbf{k} \cdot \mathbf{r}_{\nu}} | \psi_i \rangle. \quad (3.11)$$

The operator $\mathbf{r}_{\nu}$ in (3.11) is time independent so that

$$i \frac{d}{dt} \left\{ e^{iHt} \mathbf{r} e^{-iHt} \right\} = \left\{ e^{iHt} \mathbf{r} e^{-iHt} \right\} H - H \left\{ e^{iHt} \mathbf{r} e^{-iHt} \right\}$$

but on the other hand we have

$$i \frac{d}{dt} \mathbf{r}(t) = [\mathbf{r}(t), H]$$

so that $\mathbf{r}(t) = e^{iHt} \mathbf{r} e^{-iHt}$ (3.12)

Using (3.12) we can write (3.11) as

$$\sigma_i(\theta, \epsilon) = \frac{a^2}{2\pi} \frac{k}{k_0} \sum_{\nu} \int_{-\infty}^{\infty} dt e^{-i\epsilon t} \langle \psi_i | e^{i\mathbf{k} \cdot \mathbf{r}_{\nu}(t)} e^{-i\mathbf{k} \cdot \mathbf{r}_{\nu}(0)} | \psi_i \rangle. \quad (3.13)$$

The position operator $\mathbf{r}_{\nu}(t)$ can be represented in terms of a displacement vector as

$$\mathbf{r}_{\nu}(t) = \underline{\mathbf{b}}_{\nu} + \underline{\mathbf{u}}_{\nu}(t) \quad (3.14)$$

where $\underline{\mathbf{b}}_{\nu}$ is the equilibrium position of atom ν and $\underline{\mathbf{u}}_{\nu}(t)$ is its displacement at time t . If (3.14) is substituted into (3.13), the $\underline{\mathbf{b}}_{\nu}$ drops out and the expectation value in the expression becomes

$$\langle \psi_i | e^{i\mathbf{k} \cdot \underline{\mathbf{u}}_{\nu}(t)} e^{-i\mathbf{k} \cdot \underline{\mathbf{u}}_{\nu}(0)} | \psi_i \rangle = \chi_{\nu}(t) \quad (3.15)$$

For a crystal of N atoms the initial state wave function ψ_i will not in general be known. Instead one will have a distribution of initial states which is characteristic of an assembly of harmonic oscillators in thermal equilibrium. The matrix element (3.15) must thus be averaged over such a system of initial states. Let us denote this average by the notation $\langle \rangle_T$. Then

$$\langle \chi_v(t) \rangle_T = \langle e^{i\mathbf{k} \cdot \underline{u}_v(t)} e^{-i\mathbf{k} \cdot \underline{u}_v(0)} \rangle_T \quad (3.16)$$

and

$$\sigma(\theta, \epsilon) = \frac{a^2}{2\pi} \frac{k}{h_0} \sum_v \int_{-\infty}^{\infty} dt e^{-i\epsilon t} \langle \chi_v(t) \rangle_T \quad (3.17)$$

In order to calculate the quantity $\langle \chi_v(t) \rangle_T$ we decompose the motions of the crystal atoms into the normal modes for the system. We denote the resulting normal coordinates by g_λ and write the displacement vector $\underline{u}_v$ as

$$\underline{u}_v(t) = \sum_\lambda \underline{c}_v^\lambda g_\lambda(t) \quad (3.18)$$

$\underline{c}_v^\lambda$ is then a unit polarization vector for atom v in mode λ . A fundamental property of these normal modes is their independence, from which it follows that the normal coordinates of different modes will commute. We can thus write

$$e^{i\mathbf{k} \cdot \underline{u}_v(t)} = \prod_\lambda e^{i\mathbf{k} \cdot \underline{c}_v^\lambda g_\lambda(t)}$$

$$\text{so that } \langle \chi_v(t) \rangle_T = \prod_\lambda \langle \chi_v^\lambda(t) \rangle_T \quad (3.19)$$

$$\text{where } \langle \chi_v^\lambda(t) \rangle_T = \langle \exp(i\mathbf{k} \cdot \underline{c}_v^\lambda g_\lambda(t)) \exp(-i\mathbf{k} \cdot \underline{c}_v^\lambda g_\lambda(0)) \rangle_T$$

and the wave function for the system is a product of components, one for each normal mode.

$$\psi_i = \prod_\lambda \psi_i^\lambda$$

The normal coordinates q_λ are the same as those described in Section II for a single oscillator except that we now have N identical oscillators. The appropriate generalization of (2.26) is

$$g_\lambda(t) = i \left(\frac{1}{2NM\omega_\lambda} \right)^{\frac{1}{2}} \left(a_\lambda e^{-i\omega_\lambda t} - a_\lambda^* e^{i\omega_\lambda t} \right) \quad (3.20)$$

It is worth while at this point to digress for a moment to consider in more detail the physical processes described by the quantities $\chi_\nu(t)$. From (3.10) it is clear that we are interested in matrix elements of the form

$$\langle \psi_f | e^{i\mathbf{k} \cdot \mathbf{r}(t)} | \psi_i \rangle = \langle \psi_f | \exp \left[i\mathbf{k} \cdot \sum_\lambda \mathbf{c}^\lambda g_\lambda(t) \right] | \psi_i \rangle \quad (3.21)$$

Where, in terms of the wave functions of the normal modes, we have

$$\psi_f = \prod_\lambda \psi_f^\lambda \quad \psi_i = \prod_\lambda \psi_i^\lambda \quad (3.22)$$

Each ψ_f^λ or ψ_i^λ is characterized by a quantum number n_λ such that (see Sec. II)

$$E_\lambda = (n_\lambda + \frac{1}{2}) \omega_\lambda \quad (k = 1) \quad (3.23)$$

and a complete characterization of the initial or final states, insofar as the energy is concerned, is given by specifying the values of all the n_λ . We thus write the final state vector in the form $\langle n_1, n_2, \dots |$ and the initial state vector as $| n_1^0, n_2^0, \dots \rangle$. The matrix element (3.21) is then taken between these two configurations.

If the exponential is expanded we get terms of the form

$$1 + i\mathbf{k} \cdot \sum_\lambda \mathbf{c}^\lambda g_\lambda(t) - \frac{1}{2} \left\{ \mathbf{k} \cdot \sum_\lambda \mathbf{c}^\lambda g_\lambda(t) \right\}^2 + \dots$$

Using (3.20) and leaving out the $e^{i\omega_\lambda t}$ and certain other multipliers for simplicity (they are not operators relative to the energy eigenfunctions) this expansion results in terms of the following form

$$\begin{aligned} & \langle n_1, n_2, \dots | n_1^0, n_2^0, \dots \rangle + \left(\frac{1}{N}\right)^{\frac{1}{2}} \kappa \sum_{\lambda} c^{\lambda} \langle n_1, n_2, \dots | a_{\lambda} - a_{\lambda}^* | n_1^0, n_2^0, \dots \rangle \quad (3.24) \\ & + \frac{1}{N} \kappa^2 \sum_{\lambda} \sum_{\lambda'} c^{\lambda} c^{\lambda'} \langle n_1, \dots | a_{\lambda} a_{\lambda'} - a_{\lambda} a_{\lambda'}^* - a_{\lambda}^* a_{\lambda'} + a_{\lambda}^* a_{\lambda'}^* | n_1^0, \dots \rangle \\ & + \dots \text{ (terms involving } a_{\lambda} a_{\lambda'}, a_{\lambda} a_{\lambda'}^*, a_{\lambda}^* a_{\lambda'}, a_{\lambda}^* a_{\lambda'}^*, \text{ etc.)} \end{aligned}$$

In this form the matrix element (3.22) has a particularly simple physical interpretation. This can be seen by looking individually at the matrix elements appearing in (3.24). From (2.18), (2.22), and (2.24) we see that

$$\langle n_1, \dots, n_{\lambda}, \dots | a_{\lambda} | n_1^0, \dots, n_{\lambda}^0, \dots \rangle = \sqrt{n_{\lambda}^0} \delta(n_{\lambda}, n_{\lambda}^0 - 1) \prod_{\lambda'} \delta(n_{\lambda'}, n_{\lambda'}^0) \quad (3.25a)$$

$$\langle n_1, \dots, n_{\lambda}, \dots | a_{\lambda}^* | n_1^0, \dots, n_{\lambda}^0, \dots \rangle = \sqrt{n_{\lambda}^0 + 1} \delta(n_{\lambda}, n_{\lambda}^0 + 1) \prod_{\lambda'} \delta(n_{\lambda'}, n_{\lambda'}^0) \quad (3.25b)$$

$$\langle n_1, n_{\lambda}, \dots | a_{\lambda} a_{\lambda'} | n_1^0, n_{\lambda'}^0, \dots \rangle = \sqrt{n_{\lambda}^0 n_{\lambda'}^0} \delta(n_{\lambda}, n_{\lambda}^0 - 1) \delta(n_{\lambda'}, n_{\lambda'}^0 - 1) \prod_{\lambda''} \delta(n_{\lambda''}, n_{\lambda''}^0) \quad (3.25c)$$

$$\langle n_1, n_{\lambda}, \dots | a_{\lambda} a_{\lambda'}^* | n_1^0, n_{\lambda'}^0, \dots \rangle = \sqrt{n_{\lambda}^0 (n_{\lambda'}^0 + 1)} \delta(n_{\lambda}, n_{\lambda}^0 - 1) \delta(n_{\lambda'}, n_{\lambda'}^0 + 1) \prod_{\lambda''} \delta(n_{\lambda''}, n_{\lambda''}^0) \quad (3.25d)$$

$$\langle n_1, n_{\lambda}, \dots | a_{\lambda}^* a_{\lambda'} | n_1^0, n_{\lambda'}^0, \dots \rangle = \sqrt{(n_{\lambda}^0 + 1) n_{\lambda'}^0} \delta(n_{\lambda}, n_{\lambda}^0 + 1) \delta(n_{\lambda'}, n_{\lambda'}^0 - 1) \prod_{\lambda''} \delta(n_{\lambda''}, n_{\lambda''}^0) \quad (3.25e)$$

$$\langle \dots n_\lambda n_{\lambda'} \dots | a_\lambda^* a_{\lambda'}^* | \dots n_\lambda^0 n_{\lambda'}^0 \dots \rangle = \sqrt{(n_\lambda^0+1)(n_{\lambda'}^0+1)} \delta(n_\lambda, n_\lambda^0+1) \delta(n_{\lambda'}, n_{\lambda'}^0+1) \prod_{\lambda''} \delta(n_\lambda, n_{\lambda''}^0) \quad (3.25f)$$

... etc.

That is, a sum of matrix elements occurs, each of which represents a process in which transitions have occurred such that one or more modes have undergone discrete changes in energy. It is clear that these energy changes are not arbitrary, however, since energy must be conserved. In particular, the six matrix elements in (3.25) correspond respectively to the following six transitions (ϵ = neutron energy change):

$$-\omega_\lambda = \epsilon \quad (3.26a)$$

$$\omega_\lambda = \epsilon \quad (3.26b)$$

$$-\omega_\lambda - \omega_{\lambda'} = \epsilon \quad (3.26c)$$

$$-\omega_\lambda + \omega_{\lambda'} = \epsilon \quad (3.26d)$$

$$\omega_\lambda - \omega_{\lambda'} = \epsilon \quad (3.26e)$$

$$\omega_\lambda + \omega_{\lambda'} = \epsilon \quad (3.26f)$$

Higher order terms in (3.24) correspond to transitions in which

$$-\omega_\lambda - \omega_{\lambda'} - \omega_{\lambda''} = \epsilon, \quad -\omega_\lambda + \omega_{\lambda'} - \omega_{\lambda''} = \epsilon, \text{ etc.}$$

The transitions thus group naturally into processes involving emission or absorption of either one, two, three, etc., quanta (phonons) of the crystal lattice vibration.

It is important to note the relative magnitude of these processes and this is easily seen from (3.24). Clearly the matrix elements for the emission or absorption of combinations of η phonons decrease in magnitude for increasing η like $(\frac{1}{N})^{+\eta/2}$. In addition, for a given η , the probability that the matrix element for the transition will contain different λ is greater by a factor of at least N than that for which all λ

are the same. This follows from (3.24) since the number of modes (different λ) is of order N . These results will be of use later when closed forms for the transition probabilities are written down.

Returning now to the original line of development we write (3.19)

(using $e^A e^B = e^{(A+B+\frac{1}{2}[A,B])}$) as

$$\langle \chi_{\lambda}^{\lambda}(t) \rangle_T = \langle \exp \{ i [\kappa \cdot \underline{c}_{\lambda}^{\lambda} g_{\lambda}(t) - \kappa \cdot \underline{c}_{\lambda}^{\lambda} g_{\lambda}(0)] + \frac{1}{2} (\kappa \cdot \underline{c}_{\lambda}^{\lambda})^2 [g_{\lambda}(t), g_{\lambda}(0)] \} \rangle_T$$

but $[q_{\lambda}(t), q_{\lambda}(0)] = 0$ so we get

$$\langle \chi_{\lambda}^{\lambda}(t) \rangle_T = \langle \exp \{ i [\kappa \cdot \underline{c}_{\lambda}^{\lambda} (g_{\lambda}(t) - g_{\lambda}(0))] \} \rangle_T \quad (3.27)$$

It is shown in Ref. 1 that if Q is any multiple of an oscillator coordinate or a linear combination of them, we can write

$$\langle \exp Q \rangle_T = \exp \left\{ \frac{1}{2} \langle Q^2 \rangle_T \right\}$$

Using this result on (3.27) we get

$$\langle \chi_{\lambda}^{\lambda}(t) \rangle_T = \exp \left\{ \frac{1}{2} \langle - (\kappa \cdot \underline{c}_{\lambda}^{\lambda})^2 [g_{\lambda}^2(t) + g_{\lambda}^2(0) - g_{\lambda}(t) g_{\lambda}(0) - g_{\lambda}(0) g_{\lambda}(t)] \rangle_T \right\} \quad (3.28)$$

This can be simplified with the following relationships:

$$g_{\lambda}^2(t) = \left\{ e^{iHt} g_{\lambda} e^{-iHt} \right\} \left\{ e^{iHt} g_{\lambda} e^{-iHt} \right\} = e^{iHt} g_{\lambda} g_{\lambda} e^{-iHt}$$

$$\langle \psi_{\epsilon}^{\lambda} | g_{\lambda}^2(t) | \psi_{\epsilon}^{\lambda} \rangle = \langle \psi_{\epsilon}^{\lambda} | e^{iHt} g_{\lambda} g_{\lambda} e^{-iHt} | \psi_{\epsilon}^{\lambda} \rangle = \langle \psi_{\epsilon}^{\lambda} | e^{iE_{\epsilon}t} g_{\lambda} g_{\lambda} e^{-iE_{\epsilon}t} | \psi_{\epsilon}^{\lambda} \rangle$$

or

$$\langle \psi_{\epsilon}^{\lambda} | g_{\lambda}^2(t) | \psi_{\epsilon}^{\lambda} \rangle = \langle \psi_{\epsilon}^{\lambda} | g_{\lambda} g_{\lambda} | \psi_{\epsilon}^{\lambda} \rangle = \langle \psi_{\epsilon}^{\lambda} | g_{\lambda}^2(0) | \psi_{\epsilon}^{\lambda} \rangle$$

It is also clear that $[q_\lambda(t), q_\lambda(0)] = 0$ so that (3.28) becomes

$$\langle \chi_\lambda^\dagger(t) \rangle_T = \exp \left\{ -(\kappa \cdot \underline{c}_\lambda^\dagger)^2 \left[\langle g_\lambda^2(0) \rangle_T - \langle g_\lambda(t) g_\lambda(0) \rangle_T \right] \right\} \quad (3.29)$$

This quantity can be evaluated if we now substitute (3.20) for the $q_\lambda(t)$ in (3.29). For the products $\{q_\lambda(t), q_\lambda(0)\}$ we get expectation values of the form

$$\begin{aligned} \langle \psi_i^\dagger | g_\lambda(t) g_\lambda(0) | \psi_i^\dagger \rangle = & -\frac{1}{2NM\omega_\lambda} \langle \psi_i^\dagger | a_\lambda a_\lambda e^{-i\omega_\lambda t} \\ & + a_\lambda^* a_\lambda^* e^{i\omega_\lambda t} - a_\lambda a_\lambda^* e^{-i\omega_\lambda t} - a_\lambda^* a_\lambda e^{i\omega_\lambda t} | \psi_i^\dagger \rangle \end{aligned}$$

The operators (a_λ, a_λ) and $(a_\lambda^*, a_\lambda^*)$ have no non-zero diagonal elements as can be seen from (2.23) and (2.24) so that we get for the previous expression

$$\langle g_\lambda(t) g_\lambda(0) \rangle_T = \frac{1}{2NM\omega_\lambda} \left\{ \langle a_\lambda a_\lambda^* \rangle_T e^{-i\omega_\lambda t} + \langle a_\lambda^* a_\lambda \rangle_T e^{i\omega_\lambda t} \right\} \quad (3.30)$$

Referring to (2.19) and (2.20) we see that

$$\langle \psi_i^\dagger | a_\lambda a_\lambda^* | \psi_i^\dagger \rangle = n_\lambda + 1 \quad \langle \psi_i^\dagger | a_\lambda^* a_\lambda | \psi_i^\dagger \rangle = n_\lambda$$

$$\text{so that} \quad \langle a_\lambda a_\lambda^* \rangle_T = \langle n_\lambda \rangle_T + 1 \quad (3.31a)$$

$$\text{and} \quad \langle a_\lambda^* a_\lambda \rangle_T = \langle n_\lambda \rangle_T \quad (3.31b)$$

For a system of Bose-Einstein oscillators in thermal equilibrium at temperature T we have

$$\langle n \rangle_T = \frac{1}{e^{\hbar\omega/kT} - 1}$$

for the average excitation quantum number. We take this to be the thermally averaged quantum number for each mode of the crystal and write

$$\langle n_\lambda \rangle_T = \frac{1}{e^{\omega_\lambda/T} - 1} \quad (\hbar = k = 1) \quad (3.32)$$

Using this with (3.31) the expression (3.30) reduces to

$$\langle g_\lambda(t) g_\lambda(0) \rangle_T = \frac{z_\lambda e^{-i\omega_\lambda t} + e^{i\omega_\lambda t}}{2\omega_\lambda(z_\lambda - 1)NM}$$

where

$$z_\lambda = \exp(\omega_\lambda/T)$$

and the quantity $\langle \chi_\nu^\lambda(t) \rangle_T$ becomes

$$\langle \chi_\nu^\lambda(t) \rangle_T = \exp \left\{ \frac{-(\mathbf{k} \cdot \mathbf{e}_\nu^\lambda)^2}{NM} \left[\frac{z_\lambda + 1}{2\omega_\lambda(z_\lambda - 1)} - z_\lambda^{\pm \frac{1}{2}} \frac{z_\lambda^{\pm \frac{1}{2}} e^{-i\omega_\lambda t} + z_\lambda^{\mp \frac{1}{2}} e^{i\omega_\lambda t}}{2\omega_\lambda(z_\lambda - 1)} \right] \right\} \quad (3.31)$$

If we now use the easily verified relationships

$$\frac{z + 1}{z - 1} = \coth \left(\frac{\omega}{2T} \right)$$

and $z - 1 = 2z^{\frac{1}{2}} \sinh \left(\frac{\omega}{2T} \right)$

we can write (3.31) as

$$\langle \chi_\nu^\lambda(t) \rangle_T = \exp \left\{ -\frac{(\mathbf{k} \cdot \mathbf{e}_\nu^\lambda)^2}{NM} \left[\frac{\coth(\frac{\omega}{2T})}{2\omega_\lambda} - \frac{z_\lambda^{\pm \frac{1}{2}} e^{-i\omega_\lambda t} + z_\lambda^{\mp \frac{1}{2}} e^{i\omega_\lambda t}}{4\omega_\lambda \sinh(\frac{\omega_\lambda}{2T})} \right] \right\} \quad (3.32)$$

If we write

$$\chi_{\lambda}^{\nu} = \frac{(\kappa \cdot \epsilon_{\lambda}^{\nu})^2}{2NM\omega_{\lambda} \sinh(\frac{\omega_{\lambda}}{2T})} \quad y = z_{\lambda}^{-\frac{1}{2}} e^{i\omega_{\lambda}t}$$

then (3.32) becomes

$$\langle \chi_{\lambda}^{\nu}(t) \rangle_T = \exp \left\{ -\frac{(\kappa \cdot \epsilon_{\lambda}^{\nu})^2}{NM} \frac{\coth(\frac{\omega_{\lambda}}{2T})}{2\omega_{\lambda}} \right\} \exp \left\{ \frac{1}{2} x (y + \frac{1}{y}) \right\}$$

The second exponential is the generating function of the Bessel functions I_n . That is,

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

so that $\langle \chi_{\lambda}^{\nu}(t) \rangle_T$ becomes finally

$$\langle \chi_{\lambda}^{\nu}(t) \rangle_T = \exp \left\{ -\frac{(\kappa \cdot \epsilon_{\lambda}^{\nu})^2}{NM} \frac{\coth(\frac{\omega_{\lambda}}{2T})}{2\omega_{\lambda}} \right\} \sum_{n=-\infty}^{\infty} e^{in\omega_{\lambda}t} e^{-\frac{n\omega_{\lambda}}{2T}} I_n \left[\frac{(\kappa \cdot \epsilon_{\lambda}^{\nu})^2}{2NM\omega_{\lambda} \sinh(\frac{\omega_{\lambda}}{2T})} \right] \quad (3.33)$$

Using (3.19) and (3.17) the cross section can thus be written as

$$\sigma(\theta, \epsilon) = \frac{a^2}{2\pi} \frac{h}{h_0} \sum_{\lambda} \exp \left\{ -\frac{(\kappa \cdot \epsilon_{\lambda}^{\nu})^2}{NM} \frac{\coth(\frac{\omega_{\lambda}}{2T})}{2\omega_{\lambda}} \right\} \int_{-\infty}^{\infty} dt e^{-i\epsilon t} \frac{1}{\pi} \sum_{n=-\infty}^{\infty} e^{in\omega_{\lambda}t} e^{-\frac{n\omega_{\lambda}}{2T}} I_n(x_{\lambda}^{\nu}) \quad (3.34)$$

where the first exponential has been removed from the product over λ by summing over λ in the exponent. Since it is time independent it is also removed from the integral.

The time integral in (3.34) can now be seen to yield a result which is easily interpreted and simplified in terms of the discrete phonon

absorption and emission processes described earlier. This is most easily seen by looking at the terms which will result when the product over λ is performed. These will be the following:

$$\begin{aligned}
 & \sum_i e^{-\frac{\omega_i}{2T}} e^{i\omega_i t} I_1(x_i^v) + \sum_i e^{\frac{\omega_i}{2T}} e^{-i\omega_i t} I_1(x_i^v) \\
 & + \sum_{j>k} e^{-(\omega_j+\omega_k)/2T} e^{i(\omega_j+\omega_k)t} I_1(x_j^v) I_1(x_k^v) + \dots \\
 & + \sum_{l>m>n} e^{-(\omega_l+\omega_m+\omega_n)/2T} e^{i(\omega_l+\omega_m+\omega_n)t} I_1(x_l^v) I_1(x_m^v) I_1(x_n^v) + \dots \\
 & + \sum_{i>j} e^{-(\omega_i-\omega_j)/2T} e^{i(\omega_i-\omega_j)t} I_1(x_i^v) I_1(x_j^v) + \dots \\
 & + \sum_k e^{-2\omega_k/2T} e^{i2\omega_k t} I_2(x_k^v) + \sum_k e^{2\omega_k/2T} e^{-i2\omega_k t} I_2(x_k^v) + \dots \\
 & + \sum_{i>j} e^{-(\omega_i+2\omega_j)/2T} e^{i(\omega_i+2\omega_j)t} I_1(x_i^v) I_2(x_j^v) + \dots
 \end{aligned}$$

The general form should now be clear. That is, one has terms in all orders like those shown above.

Or, more generally, the terms will be of the form

$$\sum_{i,j,k,\dots} e^{\pm(l\omega_i \pm m\omega_j \pm n\omega_k \pm \dots)/2T} e^{\mp i(l\omega_i \pm m\omega_j \pm n\omega_k \pm \dots)t} I_l(x_i^{\sim}) I_m(x_j^{\sim}) I_n(x_k^{\sim}) \dots \quad (3.35)$$

Considerable simplification is realized if the restricted sums in the above expressions are changed to the unrestricted forms. The general expression (3.35) will then have the form

$$\frac{(l!)(m!)(n!) \dots}{(l+m+n+\dots)!} \sum_{i,j,k,\dots} e^{\pm(l\omega_i \pm m\omega_j \pm n\omega_k \pm \dots)/2T} \times e^{\mp i(l\omega_i \pm m\omega_j \pm n\omega_k \pm \dots)t} I_l(x_i^{\sim}) I_m(x_j^{\sim}) I_n(x_k^{\sim}) \dots \quad (3.36)$$

where the $i, j, k, \dots$ now vary independently over their whole range and the factorial factors divide out the resulting redundant combinations. If we recall that the argument of the Bessel function is

$$x_{\lambda}^{\sim} = \frac{(K \cdot C_{\lambda}^{\sim})^2}{2NM\omega_{\lambda} \sinh\left(\frac{\omega_{\lambda}}{2T}\right)}$$

which will be small for the cases of interest, then it is sufficiently accurate to write

$$I_n(x) = \sum_{r=0}^{\infty} \frac{(\frac{1}{2}x)^{n+r}}{r!(n+r)!} \approx \frac{(\frac{1}{2}x)^n}{n!} \quad (3.37a)$$

from which it follows that

$$I_n(x) \approx \frac{[I_1(x)]^n}{n!} \quad (3.37b)$$

Using (3.37) to simplify (3.36) we get

$$\frac{1}{(\ell+m+n+\dots)!} \sum_{i,j,\dots} e^{\pm(\ell\omega_i \pm m\omega_j \pm \dots)/2\tau} \quad (3.38)$$

$$\times e^{\mp i(\ell\omega_i \pm m\omega_j \pm \dots)t} [I_i(x_i^\sim)]^\ell [I_j(x_j^\sim)]^m \dots$$

The product over λ in (3.34) will thus consist of a sum of terms of this form with all combinations of the $\ell, m, \dots$.

The integral over t in (3.34) can now be easily evaluated since it results in delta functions of the neutron energy change ϵ . That is, although the $\ell, m, \dots$ appear in arbitrary combinations in (3.38) its contribution to the integral will be zero unless

$$\epsilon = \pm \ell\omega_i \pm m\omega_j \pm \dots$$

This is simply the energy conservation condition for the multiphonon processes considered earlier, and identifies the general term (3.38) as arising from an interaction involving a total of $\ell + m + \dots$ phonons. The time integral in (3.34) thus simplifies to

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{-i\epsilon t} \prod_{\lambda} \sum_{n=-\infty}^{\infty} e^{in\omega_{\lambda}t} e^{-\frac{n\omega_{\lambda}}{2\tau}} I_n(x_{\lambda}^\sim) \quad (3.39a)$$

$$= e^{-\frac{\epsilon}{2\tau}} \left\{ \sum_i I_i(x_i^\sim) + \frac{1}{2!} \sum_{j,k} I_j(x_j^\sim) I_k(x_k^\sim) + \frac{1}{3!} \sum_{\ell,m,n} I_{\ell}(x_{\ell}^\sim) I_m(x_m^\sim) I_n(x_n^\sim) \dots \right\}$$

with the auxiliary conditions that

$$\omega_i = \epsilon, \quad \omega_j + \omega_k = \epsilon, \quad \omega_{\ell} + \omega_m + \omega_n = \epsilon, \quad \text{etc.} \quad (3.39b)$$

The n^{th} term of (3.39a) corresponds to a process involving combinations of absorption and emission of n phonons of the crystal lattice vibration.

A more compact notation for (3.39) is

$$\begin{aligned} & \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{-i\epsilon t} \prod_{\lambda} \sum_{n=-\infty}^{\infty} e^{in\omega_{\lambda}t} e^{-\frac{n\omega_{\lambda}}{2T}} I_n(x_{\lambda}^{\nu}) \\ &= e^{-\epsilon/2T} \sum_{n=1}^{\infty} \frac{1}{n!} \left\{ \sum_{\substack{\pm\omega_1 \\ \vdots \\ \pm\omega_n}} I_1(x_1^{\nu}) I_1(x_2^{\nu}) \cdots I_1(x_n^{\nu}) \right\} \end{aligned} \quad (3.40)$$

In (3.40) the ω_i is always positive and a summation over $\pm\omega_i$ means to perform the sum for both signs of ω_i .

Using (3.37a) the complete expression for the scattering cross section in (3.34) becomes

$$\begin{aligned} \sigma(\theta, \epsilon) &= a^2 \frac{k}{k_0} \sum_{\nu} \exp \left\{ \sum_{\lambda} - \frac{(K \cdot \underline{c}_{\lambda}^{\nu})^2}{NM} \frac{\coth(\frac{\omega_{\lambda}}{2T})}{2\omega_{\lambda}} \right\} e^{-\epsilon/2T} \\ &\times \sum_{n=1}^{\infty} \frac{1}{n!} \left\{ \sum_{\substack{\pm\omega_1 \\ \vdots \\ \pm\omega_n}} \left(\frac{1}{2} x_1^{\nu} \right) \left(\frac{1}{2} x_2^{\nu} \right) \cdots \left(\frac{1}{2} x_n^{\nu} \right) \right\} \end{aligned} \quad (3.41)$$

together with the energy conservation condition on the allowed combination of the ω_i .

In order to be useful this expression must be averaged over the atoms of the crystal (average over ν) and over crystal orientations for each mode (average over the directions $\underline{c}_{\nu}^{\lambda}$). These operations yield an average cross section per atom equal to

$$\begin{aligned} \bar{\sigma}(\theta, \epsilon) &= a^2 \frac{k}{k_0} \exp \left\{ \sum_{\lambda} \frac{K^2 \coth(\frac{\omega_{\lambda}}{2T})}{6NM \omega_{\lambda}} \right\} e^{-\epsilon/2T} \\ &\times \sum_{n=1}^{\infty} \frac{1}{n!} \left(\frac{K^2}{2} \right)^n \left\{ \sum_{\substack{\pm\omega_1 \\ \vdots \\ \pm\omega_n}} z_1 \cdots z_n \right\} \end{aligned} \quad (3.42)$$

where

$$z_j = \left[3NM \omega_j \sinh \left(\frac{\omega_j}{2T} \right) \right]^{-1}$$

The assumption is now made that the normal modes of the crystal lattice vibration form a continuum, rather than the discrete set upon which the sums in (3.42) are taken. If $\rho(\omega) d\omega$ is the normalized number of modes between ω and $\omega + d\omega$, then since the total number of modes is $3N - 6 \approx 3N$ the sums over the modes are replaced by integrals in the following manner.

$$\sum_{\lambda} f(\omega_{\lambda}) \longrightarrow 3N \int_0^{\infty} f(\omega) \rho(\omega) d\omega$$

$$\sum_{\pm \omega_{\lambda}} f(\omega_{\lambda}) \longrightarrow 3N \int_{-\infty}^{\infty} f(\omega) \rho(\omega) d\omega$$

where $\int_0^{\infty} \rho(\omega) d\omega = 1$, $\rho(\omega) = \rho(-\omega)$.

It will be convenient to define the following quantities

$$\alpha = \frac{1}{M} \int_0^{\infty} \frac{\rho(\omega)}{\omega} \coth \left(\frac{\omega}{2T} \right) d\omega \quad (3.43)$$

$$\beta = \frac{1}{M} \int_0^{\infty} \frac{\rho(\omega) d\omega}{\omega \sinh \left(\frac{\omega}{2T} \right)} \quad (3.44)$$

$$g(\omega, T) = \frac{\rho(\omega)}{2\beta M \omega \sinh \left(\frac{\omega}{2T} \right)} \quad (3.45)$$

with the understanding that

$$g(\omega, \tau) = g(-\omega, \tau) \quad (3.46)$$

This latter condition simply defines $\rho(\omega)$ for negative values of ω .

In terms of the quantities (3.43) through (3.45) the cross section in (3.42) then becomes

$$\sigma_v(\theta, \epsilon) = \frac{k^2}{k_0} e^{-\frac{\epsilon}{2}\tau} e^{-\frac{\kappa^2}{2}\alpha} \sum_{n=1}^{\infty} \frac{1}{n!} \left(\frac{\kappa^2}{2} \beta \right)^n \int_{-\infty}^{\infty} \dots \int_{-\infty}^{\infty} d\omega_1 \dots d\omega_n g(\omega_1, \tau) \dots g(\omega_n, \tau) \quad (3.47)$$

with the auxiliary condition that

$$\sum_{i=1}^n \omega_i = \epsilon \quad (3.47a)$$

In this expression for the cross section the n^{th} term corresponds to an inelastic scattering of the neutron in which n phonons of the lattice vibration have been absorbed or emitted in combinations such that energy is conserved. Equation (3.47) is given in Ref. 5.

In expression (3.41) and the subsequent considerations which resulted in (3.47) and (3.47a) no mention has been made of the possibility of any anisotropy of the crystal lattice. Functions of the form

$$\sum_{\lambda} (\kappa \cdot \underline{e}_{\lambda})^2 f(\omega_{\lambda})$$

have been transformed to integrals without regard to whether the density function $\rho(\omega)$ may be different for different groups of modes. For example, the restoring forces on individual carbon atoms in the lattice of a graphite crystal are considerably greater for displacements within the crystal planes than for those at right angles to the crystal planes. If the crystal modes corresponding to these two cases are called the xy

and z modes respectively, then the transformation to integrals can be written as

$$\sum_{\lambda} (\mathbf{K} \cdot \mathbf{e}_{\lambda}^{\lambda})^2 f(\omega_{\lambda}) \rightarrow N_z \int_0^{\infty} K_z^2 f(\omega) \rho_z(\omega) d\omega + N_{xy} \int_0^{\infty} K_{xy}^2 f(\omega) \rho_{xy}(\omega) d\omega \quad (3.48)$$

Explicit account has been taken here of the fact that $\mathbf{e}_{\lambda}^{\lambda}$ will be a unit vector in either the z direction or else in the xy plane. When expressions such as (3.48) are averaged over orientations of the crystal, considerable simplification occurs if the crystal is considered to be randomly oriented. In that case the average values of the x, y, and z components of K^2 are simply $\frac{1}{3} K^2$ and (3.48) becomes

$$\sum_{\lambda} (\mathbf{K} \cdot \mathbf{e}_{\lambda}^{\lambda})^2 f(\omega_{\lambda}) \rightarrow \frac{1}{3} K^2 \int_0^{\infty} f(\omega) [N_z \rho_z(\omega) + N_{xy} \rho_{xy}(\omega)] d\omega \quad (3.48a)$$

The crystal is thus assumed to be isotropic insofar as angle integrations are concerned, but the essential effects of the anisotropy of the lattice are retained through the density function. An isotropic Debye crystal, for example, has a density function $\rho_x = \rho_y = \rho_z = \rho(\omega) \propto \omega^2$. This quadratic dependence has been shown³ to be directly related to the three-dimensional character of the crystal. An accurate description of graphite, on the other hand, has been shown to indicate a two-dimensional structure^{3,4} except for very low temperatures, with the consequence that the density function has the form $\rho(\omega) \propto \omega$. This consequence of the anisotropy is, of course, retained in (3.48a). No further mention need be made of anisotropic crystals since the density function will be considered to be the total density function in all cases.

The integrals in (3.47) become increasingly difficult to evaluate as n increases. The integrals for processes up to $n = 3$ are, for example, as follows.

$$\text{For } n=1 \quad \int_{-\infty}^{\infty} g(\omega, T) d\omega = \int_{-\infty}^{\infty} g(\omega, T) \delta(\epsilon - \omega) d\omega$$

$$\text{or } \int_{-\infty}^{\infty} g(\omega, T) d\omega = g(\epsilon, T) \quad (3.49)$$

For $n = 2$ if we let ω_1 vary continuously over its range, then $\omega_2 = \epsilon - \omega_1$, so that

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} g(\omega_1, T) g(\omega_2, T) d\omega_1 d\omega_2 = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} g(\omega_1, T) g(\omega_2, T) \delta(\epsilon - \omega_1 - \omega_2) d\omega_1 d\omega_2$$

$$\text{or } \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} g(\omega_1, T) g(\omega_2, T) d\omega_1 d\omega_2 = \int_{-\infty}^{\infty} g(\omega, T) g(\epsilon - \omega, T) d\omega \quad (3.50)$$

For $n = 3$ both ω_1 and ω_2 may vary continuously and ω_3 is then restricted by the condition $\omega_3 = \epsilon - \omega_1 - \omega_2$, and we get

$$\begin{aligned} & \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} g(\omega_1, T) g(\omega_2, T) g(\omega_3, T) d\omega_1 d\omega_2 d\omega_3 \\ &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} g(\omega_1, T) g(\omega_2, T) g(\epsilon - \omega_1 - \omega_2, T) d\omega_1 d\omega_2 \end{aligned} \quad (3.51)$$

If $n > 3$ these integrals become more complicated than can conveniently be handled even numerically and it is desirable to approximate them. A method based on statistics has been suggested by Schofield and Hassitt⁵ and will be used here. This method is based on the central limit theorem of statistics⁶ which states the following for quite general conditions. If $\lambda_1, \lambda_2, \lambda_3, \dots, \lambda_n$ are n independent variables with probability density $f(\lambda_1), f(\lambda_2), \dots, f(\lambda_n)$, then as n approaches infinity the probability density of the variable λ , where $\lambda = \lambda_1 + \lambda_2 + \dots + \lambda_n$, approaches the probability density of the normal probability distribution. Since the λ_i are independent we have

$$f(\lambda) = \int_{-\infty}^{\infty} \dots \int_{-\infty}^{\infty} f(\lambda_1) f(\lambda_2) \dots f(\lambda_n) d\lambda_1 d\lambda_2 \dots d\lambda_n$$

so that the central limit theorem states that

$$\lim_{n \rightarrow \infty} \int_{-\infty}^{\infty} \dots \int_{-\infty}^{\infty} f(\lambda_1) \dots f(\lambda_n) d\lambda_1 \dots d\lambda_n = \frac{1}{\sqrt{2\pi}S} \exp\left\{-\frac{(\lambda - m)^2}{2S^2}\right\} \quad (3.52)$$

where $\lambda = \lambda_1 + \lambda_2 + \dots + \lambda_n$, m is the mean value of λ , and S is the standard deviation of the distribution $f(\lambda)$.

Considering $g(\omega, T)$ as a probability density of the variable ω , and since ω has zero mean, application of the central limit theorem gives for large n

$$\int_{-\infty}^{\infty} \dots \int_{-\infty}^{\infty} d\omega_1 \dots d\omega_n g(\omega_1, T) \dots g(\omega_n, T) = \frac{1}{\sqrt{2\pi n}S_0^2} \exp\left\{-\frac{\epsilon^2}{2nS_0^2}\right\} \quad (3.53)$$

Since all of the ω_i have the same frequency function the condition $S^2 = nS_0^2$ (S_0 = standard deviation of $g(\omega_1, T)$) has been used. The

standard deviation, S_0 , must be chosen to be consistent with the definitions of α and β if $g(\omega, T)$ is a Gaussian. With (3.43) through (3.46) it is easy to show that

$$\alpha = \beta \int_{-\infty}^{\infty} g(\omega, T) \exp\left(\frac{\omega}{2T}\right) d\omega. \quad (3.54)$$

from which it follows⁵ that

$$S_0^2 = \frac{2T}{M\alpha} \quad (3.55)$$

and
$$\beta = \alpha \exp\left\{-\frac{1}{4MT\alpha}\right\} \quad (3.56)$$

Expression (3.53) can therefore be written as

$$\int_{-\infty}^{\infty} \dots \int_{-\infty}^{\infty} d\omega_1 \dots d\omega_n g(\omega_1, T) \dots g(\omega_n, T) = \left(\frac{M\alpha}{4\pi nT}\right)^{\frac{1}{2}} \exp\left\{-\frac{M\alpha \epsilon^2}{2nT}\right\} \quad (3.57)$$

and the cross section for an n phonon process for large n is then

$$\sigma_n(\epsilon, \theta) = a^2 \frac{k}{k_0} e^{-\frac{\epsilon}{2T}} \frac{1}{n!} \left(\frac{k^2}{2}\alpha\right)^n \exp\left\{-\frac{n}{4MT\alpha}\right\} \left(\frac{M\alpha}{4\pi nT}\right)^{\frac{1}{2}} \exp\left\{-\frac{M\alpha \epsilon^2}{4nT}\right\} e^{-\frac{k^2}{2}\alpha}$$

which, upon rearrangement of terms becomes

$$\sigma_n(\epsilon, \theta) = a^2 \frac{k}{k_0} \frac{1}{n!} \left(\frac{k^2}{2}\alpha\right)^n \exp\left\{-\frac{k^2}{2}\alpha\right\} \left(\frac{M\alpha}{4\pi nT}\right)^{\frac{1}{2}} \exp\left\{-\frac{M\alpha}{4nT} \left(\epsilon + \frac{n}{M\alpha}\right)^2\right\} \quad (3.58)$$

The complete inelastic scattering cross section is then written as

$$\sigma(\epsilon, \theta) = \sum_{n=1}^{\infty} \sigma_n(\epsilon, \theta) \quad (3.59)$$

where $\sigma_n(\epsilon, \theta)$ is the n^{th} term of (3.47) for small n or (3.58) for large n . Or, as a practical matter, (3.58) can be assumed to hold if it is not possible to evaluate the integrals in (3.47), although this certainly will not be accurate for $n \sim 1$ or 2 .

The total inelastic scattering cross section is obtained from (3.58) by integrating over solid angles. We can write the partial cross section for an n phonon event as

$$\sigma_n(\epsilon, \theta) = A e^{-\frac{K^2}{2} \alpha} \left(\frac{K^2}{2} \right)^n$$

$$\text{where } A = a^2 \frac{\hbar}{h_0} e^{-\epsilon/2T} \frac{1}{n!} (\beta)^n \int_{-\infty}^{\infty} \dots \int_{-\infty}^{\infty} d\omega_1 \dots d\omega_n g(\omega_1, T) \dots g(\omega_n, T)$$

$$\text{so that } \sigma(\epsilon) = \int_{-1}^1 \sigma(\epsilon, \theta) 2\pi d\mu = 2\pi A \int_{-1}^1 \left(\frac{K^2}{2} \right)^n e^{-\frac{K^2}{2} \alpha} d\mu \quad (\mu = \cos \theta) \quad (3.60)$$

Since, however, $\frac{K^2}{2}$ is defined as

$$\frac{K^2}{2} = E_1 + E_2 - 2\mu(E_1 E_2)^{\frac{1}{2}} = x$$

we see that $dx = -2(E_1 E_2)^{1/2} d\mu$. Expression (3.60) then becomes

$$\sigma(\epsilon) = \frac{\pi A}{(E_1 E_2)^{\frac{1}{2}}} \int_p^q x^n e^{-\alpha x} dx \quad (3.61)$$

$$\text{where } p = E_1 + E_2 - 2(E_1 E_2)^{\frac{1}{2}},$$

$$\text{and } q = E_1 + E_2 + 2(E_1 E_2)^{\frac{1}{2}}.$$

The integral in (3.61) is elementary, the result being

$$\int_p^g x^n e^{-\alpha x} dx = -\frac{n!}{(\alpha)^{n+1}} e^{-\alpha x} \left[1 + x\alpha + \frac{(x\alpha)^2}{2!} + \dots + \frac{(x\alpha)^n}{n!} \right] \Big|_p^g$$

Thus, if we write

$$S_n(x) = 1 + \alpha x + \frac{(\alpha x)^2}{2!} + \dots + \frac{(\alpha x)^n}{n!}$$

then the cross section in expression (3.60) becomes

$$\sigma(\epsilon) = \pi a^2 \frac{e^{-\epsilon/2T}}{\alpha E_1} \sum_{n=1}^{\infty} \left(\frac{\beta}{\alpha}\right)^n \left[e^{-\alpha p} S_n(p) - e^{-\alpha g} S_n(g) \right] \quad (3.61)$$

$$\times \int_{-\infty}^{\infty} \dots \int_{-\infty}^{\infty} d\omega_1 \dots d\omega_n g(\omega_1, T) \dots g(\omega_n, T)$$

This same expression for the cross section is given by Scholfield and Hassitt.⁵

IV. Inverse Mass Expansion of the Cross Section

The starting point for this discussion is equation (3.29), which after averaging over crystal orientations becomes

$$\langle \chi_{\lambda}^{\lambda}(t) \rangle_T = \exp \left\{ \frac{k^2}{3} \left[\langle g_{\lambda}(t) g_{\lambda}(0) \rangle_T - \langle g_{\lambda}^2(0) \rangle_T \right] \right\} \quad (4.1)$$

where, as shown in (3.30) and (3.31), we can write

$$\langle g_{\lambda}(t) g_{\lambda}(0) \rangle_T = \frac{1}{2NM\omega_{\lambda}} \left\{ (\langle n_{\lambda} \rangle_T + 1) e^{-i\omega_{\lambda}t} + \langle n_{\lambda} \rangle_T e^{i\omega_{\lambda}t} \right\} \quad (4.2)$$

In terms of these quantities the cross section is written as

$$\sigma(\epsilon, \theta) = \frac{a^2}{2\pi} \frac{k}{k_0} \int_{-\infty}^{\infty} dt e^{-i\epsilon t} \prod_{\lambda} \langle \chi_{\lambda}^{\lambda}(t) \rangle_T$$

The infinite product is simply performed by summing in the exponent as before. When this sum is converted to an integral over the mode frequencies, (4.1) becomes

$$\prod_{\lambda} \langle \chi_{\lambda}^{\lambda}(t) \rangle_T = \exp \left\{ \frac{\kappa^2}{3} (3N) \int_0^{\infty} d\omega \rho(\omega) \left[\langle g_{\lambda}(t) g_{\lambda}(0) \rangle_T - \langle g_{\lambda}^2(0) \rangle_T \right] \right\} \quad (4.3)$$

To illustrate the method, the following development will be for an ordinary Debye crystal, although the application to any other form for $\rho(\omega)$ is straightforward.

For a Debye crystal we write

$$\int_0^{\infty} N(\omega) d\omega = 3N \int_0^{\omega_D} \rho(\omega) d\omega = 3N \int_0^{\omega_D} 3 \frac{\omega^2}{\theta^3} d\omega$$

That is, the density of modes is written as

$$\rho(\omega) = \frac{3}{\theta^3} \omega^2 \quad (4.4)$$

The integral of the first term in (4.3) then becomes

$$3N \int_0^\infty \langle g_\lambda(t) g_\lambda(0) \rangle_T \rho(\omega) d\omega = \frac{9}{2M\theta^3} \int_0^\theta [(\langle n_T + 1 \rangle e^{-i\omega t} + \langle n_T \rangle e^{i\omega t})] \omega d\omega$$

Or,

$$3N \int_0^\infty \langle g_\lambda(t) g_\lambda(0) \rangle_T \rho(\omega) d\omega = \frac{3}{2M} g(t) \quad (4.5a)$$

where

$$g(t) = \frac{3}{\theta^3} \int_0^\theta [(\langle n_T + 1 \rangle e^{-i\omega t} + \langle n_T \rangle e^{i\omega t})] \omega d\omega \quad (4.5b)$$

In terms of (4.5) the product (4.3) can be expressed as

$$\prod_\lambda \langle \chi_\lambda^\dagger(t) \rangle = \exp \left\{ \frac{K^2}{2M} [g(t) - g(0)] \right\}$$

and the cross section then becomes

$$\sigma(\theta, \epsilon) = \frac{a^2}{2\pi} \frac{\hbar}{\hbar_0} \int_{-\infty}^\infty dt e^{-i\epsilon t} \exp \left\{ \frac{K^2}{2M} [g(t) - g(0)] \right\} \quad (4.6)$$

This is the same as the expression given by Nelkin⁷ for the cross section. The mass expansion consists of expanding the exponential in (4.6) in powers of

$$\frac{K^2}{2M} [g(t) - g(0)].$$

The individual terms of the expansion will have no physical interpretation as did those of the phonon expansion, but for a heavy moderator considerable simplification results since only one term may be significant. That is, we write for a heavy crystalline moderator

$$\exp\left\{\frac{K^2}{2M} [g(t) - g(0)]\right\} \approx 1 + \frac{K^2}{2M} [g(t) - g(0)]$$

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