90° the interference with even-l states and with the potential and Coulomb scattering vanishes. This would imply that the other resonance is also formed by l=1 protons as an interference effect of such magnitude due to more remote resonances is unlikely. Interference between these two levels might be expected to occur in the Li⁷(p,n)Be⁷ reaction also. However, the predominant interference as indicated by the angular distribution of the neutrons⁸ is between states of opposite parity.

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Dynamics of Neutron Scattering by Molecules^{*}

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The scattering of low-energy neutrons by molecules is investigated by a method which facilitates the treatment of inelastic processes. Dynamical systems characterized by the degrees of freedom of gas molecules are examined in detail. The formalism is applied first to the derivation of the differential cross section for scattering by an ensemble of coupled harmonically oscillating nuclei in thermal equilibrium. Rotator scattering is investigated in the limits of high and low energy, and inelastic corrections to the static approximation are also calculated. The cross section of a thermally excited monatomic gas is presented in a closed form which shows its dependence on the ratio of the mean velocity of thermal motion to the neutron velocity. Results appropriate to large and to small values of the velocity ratio are derived. Quantitative estimates for the validity of the various approximations are given.

1. INTRODUCTION

N recent years, the diffraction of slow neutrons by gases has grown in importance as a research tool, providing a useful complement to other techniques for the determination of nuclear scattering lengths and various molecular properties. But precise theoretical formulas for the differential cross sections have been exhibited, heretofore, only under the restrictive assumption of elastic scattering, or more accurately, the assumption that the energy transferred in a neutronmolecule collision is negligible in comparison with the incident neutron energy. In practice, molecules in thermally excited quantum states may impart to the neutron, and molecules of small inertia may absorb, energy in amounts which bear a significant ratio to the neutron energy. For these reasons, in many cases of experimental interest, the explicit consideration of inelasticity is necessary for a quantitative understanding of the neutron cross sections.

In principle, the cross sections can be computed by an explicit summation over contributions from all the scattering processes that occur. But the large number of molecular states encountered in a gas at thermal equilibrium and the large number of energetically permissible quantum transitions of the gas molecules will, as a rule, render such a procedure impractical. Recourse to more general and implicit methods of performing the required summations must then be sought.

A convenient step in achieving this is the use of operator representations in the description of the internal and external coordinates of the scattering molecules. Expressions for the cross section may then be developed which implicitly sum, in closed form, the contributions of all transitions permitted by the conservation laws. This device is the central one of the present work. We return to it repeatedly, considering in turn interactions with the various molecular degrees of freedom.

The operator techniques to be used are formulated in Sec. 2. In the three succeeding sections, they are illustrated by separate applications to problems involving vibrational, rotational, and translational degrees of freedom. The assembly and generalization of these results for application to the full problem of diffraction by gases is given in the paper which follows.

2. GENERAL FORMULATION

The scattering of neutrons by the nuclei of chemically bound atoms may be accurately treated by means of the familiar Fermi pseudopotential approximation.¹ The short-range potentials acting between neutrons and

^{*} Preliminary accounts of this work were presented at the New York and Washington meetings of the American Physical Society, 1954, Phys. Rev. 94, 790(A) (1954) and Phys. Rev. 95, 605(A) (1954).

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¹ E. Fermi, Ricerca sci. 7, 13 (1936).

nuclei are replaced by point interactions (delta functions). The latter are treated using the formalism of the first Born approximation, a procedure whose accuracy rests upon the adjustment of the interaction strengths to yield correctly the amplitudes for scattering by isolated, fixed nuclei. The interaction between a neutron at \mathbf{r}_n and a nucleus at \mathbf{r}' is then²

$$\mathfrak{V}(\mathbf{r}_n,\mathbf{r}') = (2\pi a/m)\delta(\mathbf{r}_n-\mathbf{r}'), \qquad (2.1)$$

in which m is the neutron mass and a the bound scattering length. The effects of the spin-dependence of the scattering length are treated in detail in the paper which follows; for the present work we assume the scattering independent of spin.

The potential experienced by a neutron traversing a molecule is effectively the sum of the contributions (2.1) due to each of the nuclei $\nu = 1, 2, \dots, N$:

$$V(\mathbf{r}_n) = \sum_{\nu} \mathfrak{V}(\mathbf{r}_n, \mathbf{r}_\nu) = (2\pi/m) \sum_{\nu} a_{\nu} \delta(\mathbf{r}_n - \mathbf{r}_\nu). \quad (2.2)$$

We consider a collision in which the neutron momentum changes from \mathbf{k}_0 to \mathbf{k} , while the molecule simultaneously undergoes a transition between the initial and final states ψ_i and ψ_f , for which the molecular energies are E_i and E_f respectively. The differential cross section in the laboratory system for such a process we denote by $\sigma_{fi}(\theta)$:

$$\sigma_{fi}(\theta) = (m/2\pi)^2 (k/k_0) \\ \times |\langle \psi_f| \int e^{-i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}_n} V(\mathbf{r}_n) d\tau_n |\psi_i\rangle|^2 \quad (2.3)$$
$$= \sum_{\nu,\nu'} a_{\nu} a_{\nu'} (k/k_0) \langle \psi_i | e^{i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}_\nu} |\psi_f\rangle \\ \times \langle \psi_f | e^{-i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}_{\nu'}} |\psi_i\rangle. \quad (2.4)$$

It is convenient to introduce notations for the neutron's gain of momentum and its gain of energy. We write

$$\begin{aligned} \mathbf{\kappa} &= \mathbf{k} - \mathbf{k}_0, \\ \boldsymbol{\epsilon} &= (1/2m) \left(k^2 - k_0^2 \right). \end{aligned} \tag{2.5}$$

We note the restrictions on the energies entering (2.4): The conservation of energy,

$$\epsilon = E_i - E_f, \qquad (2.6)$$

and the requirement that the final neutron energy be non-negative,

$$\epsilon \geqslant -\mathcal{E}_0, \tag{2.7}$$

where $\mathcal{E}_0 = k_0^2/2m$ is the initial neutron energy. Since the various molecular transitions accompanying scattering processes remain unseparated experimentally, the complete differential cross section is obtained by summing the contributions of transitions to all admissable final states of the scattering molecule. The cross section $\sigma_i(\theta)$ so defined depends on the initial state of the molecule; we shall later determine its average $\langle \sigma_i(\theta) \rangle_T$ over a distribution of molecular states in thermal equilibrium at temperature T. The conditions (2.6) and (2.7) may be incorporated into the expression for $\sigma_i(\theta)$ by making use of the delta function and its Fourier representation in carrying out the final state summation³:

$$\sigma_{i}(\theta) = \sum_{f} \int_{-\varepsilon_{0}}^{\infty} d\epsilon \, \delta(E_{i} - E_{f} - \epsilon) \sigma_{fi}(\theta)$$
$$= (2\pi)^{-1} \sum_{f} \int_{-\varepsilon_{0}}^{\infty} d\epsilon \int_{-\infty}^{\infty} dt \, e^{-i\epsilon t} e^{i(E_{i} - E_{f})t} \sigma_{fi}(\theta). \quad (2.8)$$

Introducing the molecular Hamiltonian H, we may, by using the relations $\exp(-iE_f t)\psi_f = \exp(-iHt)\psi_f$, etc., rewrite (2.8) in the form

$$\sigma_{i}(\theta) = (2\pi)^{-1} \sum_{\nu,\nu',f} a_{\nu} a_{\nu'} \int \int (k/k_{0}) e^{-i\epsilon t} \\ \times \langle \psi_{i} | e^{iHt} \exp(i\mathbf{\kappa} \cdot \mathbf{r}_{\nu}) e^{-iHt} | \psi_{f} \rangle \\ \times \langle \psi_{f} | \exp(-i\mathbf{\kappa} \cdot \mathbf{r}_{\nu'}) | \psi_{i} \rangle dt d\epsilon. \quad (2.9)$$

Assuming the final states form a complete set,⁴ we may immediately sum over them.

$$\sigma_{i}(\theta) = (2\pi)^{-1} \sum_{\nu,\nu'} a_{\nu} a_{\nu'} \int \int (k/k_{0}) e^{-i\epsilon t} \\ \times \langle \psi_{i} | e^{iHt} \exp(i\kappa \cdot \mathbf{r}_{\nu}) e^{-iHt} \exp(-i\kappa \cdot \mathbf{r}_{\nu'}) | \psi_{i} \rangle dt d\epsilon.$$
(2.10)

The energy spectrum of the scattered neutrons is contained in the integrand of (2.10). If we define $\sigma_i(\theta, \epsilon)$ as the differential cross section for scattering of neutrons with energy transfer ϵ , we may write

$$\sigma_{i}(\theta,\epsilon) = (k/2\pi k_{0}) \sum_{\nu,\nu'} a_{\nu}a_{\nu'} \int e^{-i\epsilon t} \\ \times \langle \psi_{i} | e^{iHt} \exp(i\mathbf{\kappa} \cdot \mathbf{r}_{\nu}) e^{-iHt} \exp(-i\mathbf{\kappa} \cdot \mathbf{r}_{\nu'}) | \psi_{i} \rangle dt \quad (2.11)$$

together with the integral relation

$$\sigma_i(\theta) = \int_{-\varepsilon_0}^{\infty} \sigma_i(\theta, \epsilon) d\epsilon. \qquad (2.12)$$

The expressions (2.10) and (2.11), representing the cross sections in terms of expectation values in the initial state of the molecule, furnish a convenient starting point for the development of a number of techniques to be used in treating the various molecular degrees of freedom. The Hamiltonians for these coordinates are sufficiently simple to permit the exact or approximate

² A system of units in which \hbar has unit magnitude is employed throughout the present paper and the one that follows.

^a This device was first used in a similar context by W. Lamb, Phys. Rev. 55, 190 (1939). ⁴ When identical nuclei are present in the molecule, the final

⁴ When identical nuclei are present in the molecule, the final states will be restricted by symmetry requirements. The summation may nevertheless be carried out over the complete set since the interactions are symmetrical among the like nuclei, and matrix elements for ψ_f of symmetry character different from ψ_i vanish.

evaluation of the required operator products by elementary methods. In this way we characteristically avoid all explicit use of the molecular wave functions. The expression (2.10) implicitly computes and sums the transition probabilities corresponding to all nonvanishing matrix elements, a property particularly convenient when, as is frequently the case, large numbers of different transitions take place.

To simplify the operator products entering (2.11), two devices⁵ will later prove useful. We may introduce the time-dependent position coordinate $\mathbf{r}_{\nu}(t)$ of a nucleus as an operator satisfying the Heisenberg equation of motion:

$$id(\mathbf{r}_{\nu}(t))/dt = \mathbf{r}_{\nu}(t)H - H\mathbf{r}_{\nu}(t). \qquad (2.13)$$

The solution, written in terms of the time-independent operator \mathbf{r}_{ν} , is

$$\mathbf{r}_{\nu}(t) = e^{iHt} \mathbf{r}_{\nu} e^{-iHt}. \qquad (2.14)$$

The expectation value required in (2.11) may then be expressed more compactly as

$$\begin{aligned} \langle \boldsymbol{\psi}_i | e^{iHt} \exp(i \mathbf{\kappa} \cdot \mathbf{r}_{\boldsymbol{\nu}}) e^{-iHt} \exp(-i \mathbf{\kappa} \cdot \mathbf{r}_{\boldsymbol{\nu}'}) | \boldsymbol{\psi}_i \rangle \\ &= \langle \boldsymbol{\psi}_i | \exp[i \mathbf{\kappa} \cdot \mathbf{r}_{\boldsymbol{\nu}}(t)] \exp[-i \mathbf{\kappa} \cdot \mathbf{r}_{\boldsymbol{\nu}'}(0)] | \boldsymbol{\psi}_i \rangle, \end{aligned}$$
(2.15)

where the time-independent coordinate $\mathbf{r}_{\mathbf{r}'}$ has been written in the time-dependent notation as $\mathbf{r}_{\mathbf{r}'}(0)$.

Alternatively we may note that in virtue of the canonical commutation relation between the coordinate \mathbf{r}_{ν} and its conjugate momentum \mathbf{p}_{ν} , the function $\exp(i\boldsymbol{\kappa}\cdot\mathbf{r}_{\nu})$ acts as a translation operator on the momentum:

$$\exp(i\mathbf{\kappa}\cdot\mathbf{r}_{\nu})\mathbf{p}_{\nu}\exp(-i\mathbf{\kappa}\cdot\mathbf{r}_{\nu})=\mathbf{p}_{\nu}-\mathbf{\kappa}.$$
 (2.16)

The same transformation performed on the molecular Hamiltonian H yields a new Hamiltonian, which we denote as $H_{(\nu)}'$. This is

$$H_{(\nu)}' = \exp(i\mathbf{\kappa} \cdot \mathbf{r}_{\nu}) H(\mathbf{p}_{\nu}, \mathbf{r}_{\nu}) \exp(-i\mathbf{\kappa} \cdot \mathbf{r}_{\nu})$$

= $H(\mathbf{p}_{\nu} - \mathbf{\kappa}, \mathbf{r}_{\nu})$ (2.17)

in which only the dependence of H on \mathbf{p}_{ν} and \mathbf{r}_{ν} has been indicated. The expression $H_{(\nu)}'$ may be regarded as the effective Hamiltonian of the molecule after the incident neutron has gained a momentum κ in a collision with the ν th nucleus. The direct scattering terms $(\nu = \nu')$ of (2.11) may now be simplified by noting that they contain the same transformation, applied to an exponential function of the Hamiltonian H,

$$\langle \boldsymbol{\psi}_i | e^{iHt} \exp(i \mathbf{\kappa} \cdot \mathbf{r}_{\nu}) e^{-iHt} \exp(-\mathbf{\kappa} \cdot \mathbf{r}_{\nu}) | \boldsymbol{\psi}_i \rangle = \langle \boldsymbol{\psi}_i | e^{iHt} e^{-iH(\nu)'t} | \boldsymbol{\psi}_i \rangle.$$
 (2.18)

To illustrate the application of the foregoing work we begin by discussing the interaction of the incident neutrons with the vibrational degrees of freedom.

3. SCATTERING BY VIBRATING SYSTEMS

Taking the molecular center of gravity as origin, we represent the equilibrium positions of the vibrating 5 These have also been noted by G. C. Wick, Phys. Rev. 94, 1228 (1954).

nuclei by the vectors \mathbf{b}_{ν} ($\nu = 1, \dots, N$). The displacement of the ν th nucleus from its equilibrium position \mathbf{b}_{ν} at any instant t we denote by $\mathbf{u}_{\nu}(t)$, so that the position vector $\mathbf{r}_{\nu}(t)$ is

$$\mathbf{r}_{\nu}(t) = \mathbf{b}_{\nu} + \mathbf{u}_{\nu}(t). \tag{3.1}$$

In a rotating molecule, the equilibrium positions \mathbf{b}_{ν} are, of course, themselves dependent on time. We shall save the consideration of the \mathbf{b}_{ν} 's as dynamical variables, however, till the next section and the succeeding paper, where the effects of rotation will be treated in some detail. For the present we confine our attention to the vibrational degrees of freedom by assuming the vectors \mathbf{b}_{ν} fixed. Then, by applying (2.11) and (2.15), we may find the scattering by molecules in the vibrational state ψ_i from a knowledge of the expectation values

However, the target molecules are in general in thermal equilibrium at a given temperature T rather than in predetermined initial states. The quantity desired for the prediction of experimental cross sections is therefore the mean value of (3.2) computed for a thermal distribution of initial states ψ_i . We denote such average values by the brackets $\langle \rangle_T$, and introduce the abbreviation $\chi_{\nu\nu'}$ for the products of the exponential functions of the displacement coordinates. The quantities to be calculated then take the form

$$\langle \boldsymbol{\chi}_{\boldsymbol{\nu}\boldsymbol{\nu}^{\prime}} \rangle_{T} = \langle \exp[i \boldsymbol{\kappa} \cdot \boldsymbol{u}_{\boldsymbol{\nu}}(t)] \exp[-i \boldsymbol{\kappa} \cdot \boldsymbol{u}_{\boldsymbol{\nu}^{\prime}}(0)] \rangle_{T}.$$
 (3.3)

These expressions may be evaluated exactly without difficulty for harmonically oscillating systems.

The molecular vibrations may be resolved into normal modes by introducing the set of normal coordinates q_{λ} , where $\lambda = 1, 2, \dots, 2N-6$ (or 2N-5 for linear molecules), together with the amplitude vectors $\mathbf{c}_{\mathbf{r}}^{(\lambda)}$ for each particle and each mode:

$$\mathbf{u}_{\nu}(t) = \sum_{\lambda} \mathbf{c}_{\nu}{}^{(\lambda)} q_{\lambda}(t). \qquad (3.4)$$

The dynamical independence of the normal modes makes it possible to separate their effects on the scattering. Since the normal coordinates of different modes commute with one another, exponential functions of $\mathbf{u}_r(t)$ may be factored into products of exponentials, one for each mode,

$$\exp[i\boldsymbol{\kappa}\cdot\boldsymbol{\mathbf{u}}_{\boldsymbol{\nu}}(t)] = \prod_{\lambda} \exp[i\boldsymbol{\kappa}\cdot\boldsymbol{\mathbf{c}}_{\boldsymbol{\nu}}{}^{(\lambda)}q_{\lambda}(t)]. \quad (3.5)$$

A further consequence of the independence of the modes is equipartition of energy among them. The Maxwell-Boltzmann thermal distribution function for the molecule as a whole may likewise be expressed as a product of factors, one for each mode. The mean value (3.3) of the product of two exponentials may accord-

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ingly be separated into a product of mean values:

$$\langle \chi_{\nu\nu'} \rangle_{T} = \prod_{\lambda} \langle \chi_{\nu\nu'}^{(\lambda)} \rangle_{T} = \prod_{\lambda} \langle \exp[i \boldsymbol{\kappa} \cdot \boldsymbol{c}_{\nu}^{(\lambda)} q_{\lambda}(t)] \times \exp[-i \boldsymbol{\kappa} \cdot \boldsymbol{c}_{\nu'}^{(\lambda)} q_{\lambda}(0)] \rangle_{T}.$$
(3.6)

Since the averages to be evaluated take the same form for all modes, we may confine our attention to one of them and omit the index λ . To exhibit the time dependence of a normal coordinate more explicitly, it is convenient to introduce the time-independent operators a and a^{\dagger} which are conventionally used in the quantization of the harmonic oscillator. These obey the commutation relations

$$\begin{bmatrix} a, a^{\dagger} \end{bmatrix} = 1,$$

$$\begin{bmatrix} a, a \end{bmatrix} = \begin{bmatrix} a^{\dagger}, a^{\dagger} \end{bmatrix} = 0.$$
 (3.7)

The normal coordinate q(t), which we shall take to have the same formal properties as the coordinate of a one-dimensional harmonic oscillator of unit mass and angular frequency ω , may then be expressed as

$$q(t) = i(2\omega)^{-\frac{1}{2}}(ae^{-i\omega t} - a^{\dagger}e^{i\omega t}).$$
(3.8)

This defines the normalization of the amplitude vectors $\mathbf{c}_{\nu}^{(\lambda)}$.

Exponential functions of the normal coordinates such as occur in (3.6) may be multiplied by means of the rule

$$e^{A}e^{B} = e^{A+B+\frac{1}{2}[A,B]}, \qquad (3.9)$$

which holds for any operators A and B which commute with their commutator $\lceil A, B \rceil$. Hence, for each of the modes we may write

$$\langle \boldsymbol{\chi}_{\boldsymbol{\nu}\boldsymbol{\nu}'} \rangle_{T} = \langle \exp[i\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}}q(t)] \exp[-i\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}'}q(0)] \rangle_{T} = \langle \exp\{i[\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}}q(t) - \boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}'}q(0)]\} \times \exp\{\frac{1}{2}(\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}})(\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}'})[q(t),q(0)]\} \rangle_{T}.$$
(3.10)

The thermal averages which must be evaluated are thereby reduced to those of single exponential functions whose arguments are displaced oscillator coordinates. The evaluation of such averages may be performed immediately by use of a corollary of Bloch's theorem on the distribution function of an oscillator coordinate⁶: If we let Q be any multiple of an oscillator coordinate or linear combination of them, we have

$$\langle \exp Q \rangle_T = \exp\{\frac{1}{2} \langle Q^2 \rangle_T\}.$$
 (3.11)

A simple, self-contained proof of this relation using the methods of the present section is given in Appendix A. Employing this evaluation of the thermal average in (3.10) and noting that expectation values of $q^2(t)$ are the same as those of $q^2(0)$, we find

$$\langle \boldsymbol{\chi}_{\boldsymbol{\nu}\boldsymbol{\nu}'} \rangle_{T} = \exp\{-\frac{1}{2} [(\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}})^{2} + (\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}'})^{2}] \langle q^{2}(0) \rangle_{T} + (\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}}) \langle \boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}'} \rangle \langle q(l) q(0) \rangle_{T} \}.$$
(3.12)

⁶ F. Bloch, Z. Physik 74, 295 (1932).

The averages of the expressions bilinear in the coordinates which occur in (3.12) may be found directly from the expansion (3.8) and the commutation rules (3.7). These yield

$$\langle q(t)q(0)\rangle_T = (2\omega)^{-1} \{ \langle aa^{\dagger}\rangle_T e^{-i\omega t} + \langle a^{\dagger}a\rangle_T e^{i\omega t} \} = (2\omega)^{-1} \{ \langle \langle n\rangle_T + 1 \rangle e^{-i\omega t} + \langle n\rangle_T e^{i\omega t} \}$$
(3.13)

in which we have introduced the thermally averaged excitation quantum member $\langle n \rangle_T$. With the notation⁷

$$z = \exp(\omega/T), \qquad (3.14)$$

$$\langle n \rangle_T$$
 is given by $\langle n \rangle_T = (z-1)^{-1},$ (3.15)

and (3.13) becomes

$$\langle q(t)q(0)\rangle_T = [2\omega(z-1)]^{-1}(ze^{-i\omega t}+e^{i\omega t}).$$
 (3.16)

The thermal average (3.12) is therefore given by

$$\begin{array}{l} \langle \chi_{\nu\nu'} \rangle_T = \exp\{-\frac{1}{2} [(\mathbf{\kappa} \cdot \mathbf{c}_{\nu})^2 + (\mathbf{\kappa} \cdot \mathbf{c}_{\nu'})^2] \\ \times (z+1)/2\omega(z-1)\} \exp\{(\mathbf{\kappa} \cdot \mathbf{c}_{\nu})(\mathbf{\kappa} \cdot \mathbf{c}_{\nu'})z^{\frac{1}{2}} \\ \times (z^{\frac{1}{2}}e^{-i\omega t} + z^{-\frac{1}{2}}e^{i\omega t})/2\omega(z-1)\}. \end{array}$$
(3.17)

As we have shown in the previous section, the distribution of energy transferred by the neutron to the scattering system is given by a Fourier integral over time of the thermal average (3.3). The latter, by means of (3.6), may be expressed as a product of factors of the form (3.17), one for each mode. Since the scattering system is equivalent to an assembly of harmonic oscillators, energy transfers must be confined to the discrete values $\epsilon = \sum n_{\lambda} \omega_{\lambda}$, where the ω_{λ} are the normal mode angular frequencies and the n_{λ} are integers. This restriction of the energy transfers may be seen to follow, in the present context, from the fact that the expressions (3.17) for the different modes are periodic in time with the fundamental frequencies ω_{λ} . Fourier integrals of these expressions are easily evaluated by expanding them first as complex Fourier series. As we shall see, the coefficients of the series may then be identified with the probabilities that the different quantum transitions take place.

To expand (3.17) as a Fourier series, we need only note that the second of its two exponential factors already has the form of the generating function for the modified Bessel functions⁸ I_n :

$$e^{\frac{1}{2}x(y+1/y)} = \sum_{n=-\infty}^{\infty} y^n I_n(x).$$
 (3.18)

Carrying out the expansion as indicated, and eliminating z via (3.14) we find for (3.17)

$$\langle \boldsymbol{\chi}_{\boldsymbol{\nu}\boldsymbol{\nu}'} \rangle_{T} = \exp\{-\left[(\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}})^{2} + (\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}'})^{2}\right](4\omega)^{-1} \coth(\omega/2T)\} \\ \times \sum_{n=-\infty}^{\infty} e^{in\omega t} e^{-(n\omega/2T)} I_{n} \left(\frac{(\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}})(\boldsymbol{\kappa} \cdot \boldsymbol{c}_{\boldsymbol{\nu}'})}{2\omega \sinh(\omega/2T)}\right).$$
(3.19)

⁷ By measuring the temperature in units of energy, we eliminate explicit occurrence of the Boltzmann constant. ⁸ G. N. Watson, Bessel Functions (The Macmillan Company,

New York, 1948).

To find the distribution of scattered particles we substitute this expansion, performed for each of the modes, in the appropriately specialized form of Eq. (2.11),

$$\langle \sigma(\theta, \epsilon) \rangle_{T} = \frac{k}{2\pi k_{0}} \sum_{\nu, \nu'} a_{\nu} a_{\nu'} \int e^{-i\epsilon t} \\ \times \exp[i\kappa \cdot (\mathbf{b}_{\nu} - \mathbf{b}_{\nu'})] \prod_{\lambda} \langle \chi_{\nu\nu'}{}^{(\lambda)} \rangle_{T} dt.$$
(3.20)

The individual Fourier components of the product $\prod \langle \chi_{\nu\nu'}^{(\lambda)} \rangle_T$ in (3.20) will contribute delta functions of the energy transfer ϵ . From the coefficients of these we may identify the partial cross sections for any given excitation processes. In particular, for a scattering characterized by the energy transfer $\epsilon = \sum_{\lambda} n_{\lambda} \omega_{\lambda}$, we find the partial differential cross section⁹

$$\langle \sigma_{[n_{\lambda}]}(\theta) \rangle_{T} = (k/k_{0}) \sum_{\nu, \nu'} a_{\nu} a_{\nu'} \\ \times \exp[i \kappa \cdot (\mathbf{b}_{\nu} - \mathbf{b}_{\nu'})] W_{\nu\nu'} \quad (3.21)$$

where

$$W_{\nu\nu'} = \prod_{\lambda} \exp\{-\left[(\mathbf{\kappa} \cdot \mathbf{c}_{\nu}^{(\lambda)})^{2} + (\mathbf{\kappa} \cdot \mathbf{c}_{\nu'}^{(\lambda)})^{2}\right] \\ \times (4\omega_{\lambda})^{-1} \coth(\omega_{\lambda}/2T)\} \exp(-n_{\lambda}\omega_{\lambda}/2T) \\ \times I_{n_{\lambda}} \left(\frac{(\mathbf{\kappa} \cdot \mathbf{c}_{\nu}^{(\lambda)})(\mathbf{\kappa} \cdot \mathbf{c}_{\nu'}^{(\lambda)})}{2\omega_{\lambda} \sinh(\omega_{\lambda}/2T)}\right). \quad (3.22)$$

For many of the cases occurring in practice the argument of the modified Bessel function is sufficiently small to permit approximation of the function by the first term of its power series expansion:

$$I_n(x) \sim (\frac{1}{2}x)^{|n|} (|n|!)^{-1}.$$
 (3.23)

4. SCATTERING BY ROTATORS

The scattering of slow neutrons by a gas may induce large numbers of different rotational transitions among the molecules. In this situation, which prevails characteristically for neutron energies larger than the rotational level spacings, the labor of computing the cross sections by direct summation of the transition probabilities becomes prohibitive. The present section is devoted to the development of convenient approximation methods for finding the cross sections and energy distributions of neutrons scattered by rotators.

Many features of the problem may be understood by treating the simplest model of a scattering system possessing only rotational degrees of freedom. We consider the collisions of neutrons with a single point scatterer constrained to move on the surface of a sphere. Let **b** denote the position coordinate of the scatterer, \mathfrak{M} its mass, *a* its scattering length, and $b = |\mathbf{b}|$ the radius of the sphere. The Hamiltonian for the rotator is given by

$$H = L^2/(2\mathfrak{M}b^2) = (\mathbf{b} \times \mathbf{p})^2/(2\mathfrak{M}b^2), \qquad (4.1)$$

where L is the angular momentum of the scatterer. This model proves of considerable practical use in treating the scattering by spherical top or linear molecules. Molecules of both these types with moment of inertia I rotate with energy $L^2/(2I)$. The direct scattering by a nucleus at distance *b* from the center of mass of either of these is described by the model if an effective "rotational mass" $\mathfrak{M} = I/b^2$ is given the nucleus.

A number of approaches will be developed for treating the rotator. For sufficiently high neutron energies the scattering may be described in an essentially classical way. Using the techniques of Sec. 2, the classical limit may be taken as the starting point for an approximation method which computes successive quantummechanical corrections to the cross sections and energy distributions. Alternatively, for the frequently occurring cases in which $(m/\mathfrak{M})\ll 1$ an expansion of the general expression for the cross section in powers of the mass ratio may be performed. These methods are developed in turn and compared. The interference effects which arise when the rotator consists of more than one scatterer are treated at a later point. In Appendix C another approximation method is introduced which treats neutron scattering in the very low energy regions where the other methods are unsuitable.

For the rotator, the Hamiltonian after collision, defined in the sense of (2.17), is

$$H' = [\mathbf{b} \times (\mathbf{p} - \mathbf{\kappa})]^2 / (2\mathfrak{M}b^2)$$

= [L²+\mathbf{\kappa} \cdot (\beta \times \beta - \beta \times \beta) + (\beta \times \mathbf{\kappa})^2] / (2\mathfrak{M}b^2). (4.2)

The scattering by rotators with angular momentum lmay be described in terms of an expectation value of the form (2.18) averaged over all orientations of the axis of rotation. Writing this as $\langle \chi \rangle_l$, we have

$$\langle \chi \rangle_{l} = \frac{1}{2l+1} \sum_{m=-l}^{l} \langle Y_{l}^{m} | e^{iHt} e^{-iH't} | Y_{l}^{m} \rangle \quad (4.3)$$

$$= \langle e^{iHt} e^{-iH't} \rangle_l, \tag{4.4}$$

where the wave functions $Y_l^m = Y_l^m(\vartheta, \varphi)$ are normalized spherical harmonics.

A classical description of the scattering is obtained by considering the limit of large values of $k_0 b$, or effectively, by neglecting the contributions of the operator \mathbf{L} in H' and H. Then (4.4) reduces to the form

$$\langle \boldsymbol{\chi} \rangle_l = \langle \exp[-it(\mathbf{b} \times \mathbf{\kappa})^2/(2\mathfrak{M}b^2) \rangle_l.$$

Since this form no longer contains operators which fail to commute with the coordinate **b**, the average over orientations (4.3) may be performed classically. If xis the cosine of the angle between κ and **b**, we have

$$\langle \chi \rangle_l = \frac{1}{2} \int_{-1}^{+1} \exp[-it\kappa^2(1-x^2)/2\mathfrak{M}] dx.$$

⁹ Equivalent results are quoted by N. K. Pope, Can. J. Phys. 30, 597 (1952). The present procedure of treating all the inelastic processes at once and performing the thermal averaging before computing the individual transition probabilities considerably simplifies the derivation.

Substituting this in the expression (2.11) for the cross so that the differential cross section becomes¹⁰ section and integrating over time yields

$$\sigma_{cl}(\theta,\epsilon) = \frac{1}{2}a^2(k/k_0)\int \delta(\epsilon + \kappa^2(1-x^2)/2\mathfrak{M})dx$$

Performing the integral of the delta function, we find

$$\sigma_{cl}(\theta,\epsilon) = a^2 \frac{k}{k_0} \frac{\mathfrak{M}}{\kappa(\kappa^2 + 2\mathfrak{M}\epsilon)^{\frac{1}{2}}} \quad \text{for} \quad -\frac{\kappa^2}{2\mathfrak{M}} \leq \epsilon \leq 0,$$

=0, otherwise. (4.5)

In terms of the neutron momenta, the inequality in (4.5) becomes

$$k_0 \ge k \ge k_0 [m \cos\theta + (\mathfrak{M}^2 - m^2 \sin^2\theta)^{\frac{1}{2}}](\mathfrak{M} + m)^{-1},$$

which states the bounds of the outgoing neutron spectrum. The differential cross section $\sigma_{cl}(\theta) = \int \sigma_{cl}(\theta, \epsilon) d\epsilon$, which may be found by a numerical integration, is more easily determined, in most cases, from an expansion in powers of the mass ratio m/\mathfrak{M} :

$$\sigma_{cl}(\theta)/a^{2} = 1 - (m/\mathfrak{M})[(4/3) - (4/3) \cos\theta] + (m/\mathfrak{M})^{2}(1 - \cos\theta)[(4/3) - \frac{4}{5} \cos\theta] - (48/35) (m/\mathfrak{M})^{3}(1 - \cos\theta)^{2} + (16/315) (m/\mathfrak{M})^{4}(1 - \cos\theta)^{2} \times (27 - 10 \cos\theta - 5 \cos^{2}\theta) - \cdots$$

These terms occur in the calculation of the cross section of methane, described in the following paper.

A cross section $\sigma^{(1)}(\theta,\epsilon)$ taking into account the first quantum corrections to $\sigma_{cl}(\theta,\epsilon)$, i.e., corrections of order $(\kappa b)^{-2}$, may also be calculated. We record here only the result; the derivation is given in Appendix B:

$$\sigma^{(1)}(\theta,\epsilon) = \sigma_{cl}(\theta,\epsilon) \left[1 + \frac{1}{8} (\kappa b)^{-2} Y(y) \right],$$

where

$$y = \kappa^2 (\kappa^2 + 2\mathfrak{M}\epsilon)^{-1},$$

$$Y(y) = y + 4y^2 - 5y^3 + l(l+1)(6y^2 + 2y).$$

For a development of the general expression for the differential cross in powers of the mass ratio (m/\mathfrak{M}) , we return to (4.4).

Consider a Taylor expansion,

$$\langle \chi \rangle_l = \sum_{n=0}^{\infty} c_n \left(\frac{it}{\mathfrak{M}}\right)^n,$$
 (4.6)

in which the coefficients c_n are functions of κ and l. Then $\sigma_l(\theta, \epsilon)$ can be expressed formally in terms of derivatives of the δ function as follows:

$$\sigma_{l}(\theta,\epsilon) = \frac{a^{2}}{2\pi} \frac{k}{k_{0}} \sum_{n} c_{n} \left(\frac{-1}{\mathfrak{M}} \frac{d}{d\epsilon}\right)^{n} \int_{-\infty}^{\infty} e^{-i\epsilon t} dt$$
$$= a^{2} (k/k_{0}) \sum_{n} (-\mathfrak{M})^{-n} c_{n} \delta^{(n)}(\epsilon).$$

Integration over the variable ϵ may now be performed

$$\sigma_{l}(\theta) = a^{2}k_{0}^{-1} \sum \mathfrak{M}^{-n} (d/d\epsilon)^{n} (kc_{n})_{\epsilon=0}$$

= $a^{2}k_{0}^{-1} \sum (m/\mathfrak{M})^{n} (k^{-1}d/dk)^{n} (kc_{n})_{k=k_{0}}.$ (4.7)

Thus the expansion of the expectation value in powers of the time expresses the differential cross section as a power series in (m/\mathfrak{M}) . This ratio measures the inelasticity of the scattering. A small amount of inelasticity means a slowly varying time-dependence of the expectation value. This, in turn, implies small coefficients of t in the time expansion and rapid decrease of the successive terms. The zeroth-order term of the expansion is the well-known static approximation which neglects inelasticity entirely.

We must now develop a technique for evaluating the coefficients c_n for the case of a point scatterer constrained to move on a sphere. For states in which the scatterer has angular momentum l, the expectation value (4.4) may be written as

$$\langle \chi \rangle_{l} = (2l+1)^{-1} \sum_{m} \langle Y_{l}^{m} | e^{-it(H'-E_{l})} | Y_{l}^{m} \rangle \quad (4.8)$$

where $E_l = l(l+1)/(2\mathfrak{M}b^2)$ is the energy of the rotational state. Making use of the expression (4.2) for H', we may write

$$H' - E_l = (H - E_l) - (i\kappa/\mathfrak{M}b)\beta + (\kappa^2/2\mathfrak{M})\gamma, \quad (4.9)$$

where the operators β and γ are defined as

$$\beta = \frac{1}{2}i\mathbf{\kappa} \cdot (\mathbf{b} \times \mathbf{L} - \mathbf{L} \times \mathbf{b})/\kappa b$$

= $\mathbf{\kappa} \cdot (\mathbf{b} + i\mathbf{b} \times \mathbf{L})/\kappa b$, (4.10)
 $\gamma = (\mathbf{\kappa} \times \mathbf{b})^2/(\kappa^2 b^2)$,

and the commutation relation between **b** and **L** has been used in deriving the second expression for β . Explicit representations of these operators may be constructed by choosing a system of polar coordinates¹¹ (ϑ, φ) with its polar axis in the direction of κ . Then we have the angular momentum components:

$$\begin{split} L_x &= i \sin \varphi (\partial/\partial \vartheta) + i \cot \theta \cos \varphi (\partial/\partial \varphi), \\ L_y &= -i \cos \varphi (\partial/\partial \vartheta) + i \cot \theta \sin \varphi (\partial/\partial \varphi), \\ L_z &= -i (\partial/\partial \varphi), \end{split}$$

in terms of which we may evaluate H, β , and γ . Letting $x = \cos\vartheta$ we find

$$H = \frac{L^2}{2\mathfrak{M}b^2} = \frac{-1}{2\mathfrak{M}b^2} \left\{ \frac{1}{\sin\vartheta} \frac{\partial}{\partial\vartheta} \left(\sin\vartheta \frac{\partial}{\partial\vartheta} \right) - \frac{m^2}{\sin^2\vartheta} \right\}$$
$$= \frac{-1}{2\mathfrak{M}b^2} \left\{ \frac{\partial}{\partial x} (1 - x^2) \frac{\partial}{\partial x} - \frac{m^2}{1 - x^2} \right\}, \quad (4.11)$$

$$\beta = \sin\vartheta \left(\frac{\partial}{\partial \vartheta} \right) + \cos\vartheta = (x^2 - 1) \left(\frac{\partial}{\partial x} \right) + x, \qquad (4.12)$$

$$\gamma = \sin^2 \vartheta = 1 - x^2. \tag{4.13}$$

¹⁰ A similar procedure was employed by Wick (reference 5) to derive the approximation of Plazek for total neutron cross sections, G. Placzek, Phys. Rev. 86, 377 (1952). ¹¹ The azimuthal angle φ is measured from the x axis.

Since, owing to the choice of coordinate system, these operators are independent of φ , the spherical harmonics Y_{l}^{m} occurring as wave functions in (4.8) may be replaced by the associated Legendre functions $P_l^m(x)$, multiplied by suitable normalization constants $N_{l, m^{\frac{1}{2}}}$.

We now define the "quantum number" operator \mathfrak{N} and the "raising" and "lowering" operators \mathfrak{R} and \mathfrak{L} by

$$\begin{aligned} \mathfrak{N}P_{l}^{m}(x) &= lP_{l}^{m}(x), \\ \mathfrak{R}P_{l}^{m}(x) &= P_{l+1}^{m}(x), \\ \mathfrak{L}P_{l}^{m}(x) &= P_{l-1}^{m}(x), \\ &= 0 \quad \text{if} \quad |m| = l. \end{aligned}$$

By means of the recursion relations

$$(x^{2}-1)(d/dx)P_{l}^{m}(x) - (l-m+1)P_{l+1}^{m}(x) + (l+1)xP_{l}^{m}(x) = 0$$

and

$$(2l+1)xP_{l^{m}}(x) - (l-m+1)P_{l+1}{}^{m}(x) - (l+m)P_{l-1}{}^{m}(x) = 0$$

we infer

we inter

$$\begin{split} H &= \mathfrak{N}(\mathfrak{N}+1)/(2\mathfrak{M}b^2), \\ \beta &= \mathfrak{N}\frac{(\mathfrak{N}+1)(\mathfrak{N}+1-m)}{2\mathfrak{N}+1} - \mathfrak{L}\frac{\mathfrak{N}(\mathfrak{N}+m)}{2\mathfrak{N}+1}, \\ \gamma &= 2(\mathfrak{N}^2 + \mathfrak{N} - 1 + m^2)/(2\mathfrak{N} - 1)(2\mathfrak{N}+3) \\ &- \mathfrak{N}^2\frac{(\mathfrak{N}+2-m)(\mathfrak{N}+1-m)}{(2\mathfrak{N}+3)(2\mathfrak{N}+1)} - \mathfrak{L}^2\frac{(\mathfrak{N}-1+m)(\mathfrak{N}+m)}{(2\mathfrak{N}-1)(2\mathfrak{N}+1)}. \end{split}$$

Further, if $f(\mathfrak{N})$ is any function of \mathfrak{N} ,

$$\Re f(\mathfrak{N}) = f(\mathfrak{N}-1)\Re, \quad \pounds f(\mathfrak{N}) = f(\mathfrak{N}+1)\pounds,$$

and

$$\mathfrak{LR} = 1, \quad \mathfrak{RL}(\mathfrak{N}^2 - m^2) = \mathfrak{N}^2 - m^2.$$
 (4.14)

We now expand $\exp[-it(H'-E_l)]$ in powers of t, retaining only those terms with diagonal matrix elements (terms which may be reduced to a form independent of \mathcal{R} and \mathcal{L}). Note that only even powers of β contribute to diagonal matrix elements. Moreover, as $H-E_l$ yields zero when standing adjacent to $P_l^m(x)$, the only terms involving $H - E_l$ which need be kept are those in which it has factors of β or γ on both sides of it. The use of the \Re , \mathfrak{L} , and \mathfrak{N} representation, with the simple prescriptions (4.14) for commutation relations provides a straightforward method for evaluation of the time expansion coefficients to any order. We exhibit here the calculation to terms in t^2 .

Keeping only terms with diagonal matrix elements, we have

$$\langle P_l^m | \exp[-it(H'-E_l)] | P_l^m \rangle = \langle P_l^m | 1 - \frac{1}{2}(it/\mathfrak{M})\kappa^2 \gamma + (it/\mathfrak{M})^2 (\frac{1}{8}\kappa^4 \gamma^2 - \frac{1}{2}\kappa^2 \beta^2 b^{-2}) + \cdots | P_l^m \rangle.$$

The part of β^2 which contributes is

$$-\mathfrak{R}\frac{(\mathfrak{N}+1-m)(\mathfrak{N}+1)}{2\mathfrak{N}+1}\mathfrak{L}\frac{(\mathfrak{N}+m)\mathfrak{N}}{2\mathfrak{N}+1} \\ -\mathfrak{L}\frac{(\mathfrak{N}+m)\mathfrak{N}}{2\mathfrak{N}+1}\mathfrak{R}\frac{(\mathfrak{N}+1-m)(\mathfrak{N}+1)}{2\mathfrak{N}+1}$$

Hence,

Since

 $N_{l,m} \langle P_{l}^{m} | \beta^{2} | P_{l}^{m} \rangle$

$$=\frac{-l^{2}(l^{2}-m^{2})}{(2l-1)(2l+1)}-\frac{\left[(l+1)^{2}-m^{2}\right](l+1)^{2}}{(2l+3)(2l+1)}$$
$$\frac{1}{2l+1}\sum_{m=-l}^{l}m^{2}=\frac{1}{3}l(l+1),$$

it follows that

$$(2l+1)^{-1}\sum_{m} N_{l,m} \langle P_l^m | \beta^2 | P_l^m \rangle = -\frac{1}{3}(l^2+l+1).$$

Spatial averages of powers of γ unmixed with powers of β or $H-E_l$ are easily obtained from the representation (4.13), so that

 $\langle \sin^2 \vartheta \rangle_{AV} = \frac{2}{3}, \quad \langle \sin^4 \vartheta \rangle_{AV} = 8/15.$

Thus.

$$\langle \chi \rangle_l = 1 - \frac{it}{\mathfrak{M}} \frac{\kappa^2}{3} + \left(\frac{it}{\mathfrak{M}}\right)^2 \left[\frac{\kappa^4}{15} + \frac{\kappa^2(l^2+l+1)}{6b^2}\right].$$

Therefore, by (4.7), we obtain

$$\sigma_{l}(\theta)/a^{2} = 1 - (m/\mathfrak{M}) [(4/3) - (4/3) \cos\theta] + (m/\mathfrak{M})^{2} \{ (1 - \cos\theta) [(4/3) - \frac{4}{5} \cos\theta] + \frac{1}{3} (l^{2} + l + 1) (k_{0}b)^{-2} \} - \cdots . \quad (4.15)$$

Each power of $(m/\mathfrak{M})^n$ in the series development multiplies a sequence of *n* terms of which the first is solely a function of angle. The further terms contain successively higher powers of $(k_0b)^{-2}$ as well as polynomials in l. For a gas in thermal equilibrium, thermal averages of these polynomials must be computed. The leading terms represent the formal expansion in powers of (m/\mathfrak{M}) of the classical cross section, $\sigma_{cl}(\theta)$. The calculation of the diffraction cross section for methane discussed in the paper to follow requires the consideration of terms of the latter category to order $(m/\mathfrak{M})^4$. The sum of the second terms constitutes the first correction $\sigma^{(1)}(\theta)$ to $\sigma_{cl}(\theta)$. For low energies, $k_0 b \ll 1$, the last terms of the sequences multiplying powers of m/\mathfrak{M} predominate. A low-energy approximation to the cross section given in appendix C is shown to constitute a selective summation of precisely these terms of the mass expansion. The closed expression representing this sum indicates that the mass expansion fails to converge for neutron energies smaller than the rotational level spacings.

We consider next the interference terms which result from the presence of more than one scattering center in a rotating molecule. Since particle interference is inherently a quantum phenomenon, there is no classical approximation analogous to (4.5).

Let \mathbf{b}_1 , a_1 and \mathbf{b}_2 , a_2 be the position coordinates and scattering lengths of two nuclei rigidly bound to a molecule that rotates as a spherical top or linear configuration. Let *I* be the moment of inertia of the molecule. We employ a device earlier applied to the treatment of direct scattering by introducing a Hamiltonian $H_{(2)}'$,

$$H_{(2)}' = \exp(i\mathbf{\kappa} \cdot \mathbf{b}_2) H \exp(-i\mathbf{\kappa} \cdot \mathbf{b}_2) = [L^2 + 2\mathbf{\kappa} \cdot (\mathbf{b}_2 \times \mathbf{L} - i\mathbf{b}_2) + (\mathbf{\kappa} \times \mathbf{b}_2)^2](2I)^{-1}.$$

The expectation value for an interference term, averaged over all states ψ_l of angular momentum l,

$$\langle \chi_{\mathrm{int}} \rangle_l = \langle \psi_l | e^{iHt} \exp(i \mathbf{\kappa} \cdot \mathbf{b}_1) e^{-iHt} \exp(-i \mathbf{\kappa} \cdot \mathbf{b}_2) | \psi_l \rangle_{\mathrm{Av}},$$

can then we written as

 $\begin{aligned} \langle \chi_{\rm int} \rangle_l &= \langle \psi_l | \exp(i \mathbf{\kappa} \cdot \mathbf{b}_{12}) \exp[-it(H_{(2)}' - E_l)] | \psi_l \rangle_{\rm Av}(4.16) \\ \text{with } \mathbf{b}_{12} &= \mathbf{b}_1 - \mathbf{b}_2. \text{ To first order in } t, (4.16) \text{ becomes} \\ \langle \chi_{\rm int} \rangle_l &= \langle \psi_l | \exp(i \mathbf{\kappa} \cdot \mathbf{b}_{12}) | \psi_l \rangle_{\rm Av} - (it/2I) \\ &\times \langle \psi_l | \exp(i \mathbf{\kappa} \cdot \mathbf{b}_{12}) [- 2i \mathbf{\kappa} \cdot \mathbf{b}_2 + (\mathbf{\kappa} \times \mathbf{b}_2)^2] | \psi_l \rangle_{\rm Av} \\ &- (it/I) \langle \psi_l | \exp(i \mathbf{\kappa} \cdot \mathbf{b}_{12}) (\mathbf{\kappa} \times \mathbf{b}_2 \cdot \mathbf{L}) | \psi_l \rangle_{\rm Av}. \end{aligned}$

In the third term of (4.17) the operator L acts directly upon a wave function ψ_i . As is easily seen by choosing the axis of quantization along $(\mathbf{x} \times \mathbf{b}_2)$, the indicated average of a component of L over all orientations of the rotation axis vanishes. Averages of the other terms can be performed classically. The integrations which occur give rise to the spherical Bessel functions

$$j_0(x) = x^{-1} \sin x,$$

$$j_1(x) = -x^{-1} \cos x + x^{-2} \sin x.$$
(4.18)

With these definitions, we find

$$\begin{aligned} \langle \boldsymbol{\psi}_l | \exp(i \boldsymbol{\kappa} \cdot \boldsymbol{b}_{12}) | \boldsymbol{\psi}_l \rangle_{\text{Av}} &= (4\pi)^{-1} \boldsymbol{\int} \exp(i \boldsymbol{\kappa} \cdot \boldsymbol{b}_{12}) d\Omega \\ &= j_0(\kappa b_{12}), \end{aligned}$$

 $\langle \boldsymbol{\psi}_l | \exp(i \boldsymbol{\kappa} \cdot \mathbf{b}_{12}) i \boldsymbol{\kappa} \cdot \mathbf{b}_2 | \boldsymbol{\psi}_l \rangle_{\text{Av}} = \kappa b_{12}^{-1} (b_2^2 - \mathbf{b}_1 \cdot \mathbf{b}_2) j_1(\kappa b_{12}),$ and

$$\begin{aligned} \langle \boldsymbol{\psi}_l | \exp(i\boldsymbol{\kappa} \cdot \mathbf{b}_{12}) (\boldsymbol{\kappa} \times \mathbf{b}_2)^2 | \boldsymbol{\psi}_l \rangle_{\text{Av}} \\ &= \kappa^2 b_{12}^{-2} (\mathbf{b}_1 \times \mathbf{b}_2)^2 j_0 (\kappa b_{12}) \\ &+ \kappa b_{12}^{-1} [2b_2^2 - 3b_{12}^{-2} (\mathbf{b}_1 \times \mathbf{b}_2)^2] j_1 (\kappa b_{12}) \end{aligned}$$

The portion of the differential cross section arising from interference effects is derived from the expectation value (4.16) contributed by scatterers 1 and 2, which we have computed in first order, plus an equal term in which the roles of 1 and 2 are interchanged. To find the interference cross section contributed by this pair of scatterers, we substitute the averages computed above into an obvious adaptation of the time expansion formula (4.7), to interference terms, obtaining

$$\sigma_{\text{int}}^{(1,2)}(\theta)/a_{1}a_{2}=2j_{0}(\kappa b_{12})$$

$$-\frac{m}{I}\frac{1}{k_{0}^{2}}\frac{d}{dk}\left\{\frac{k\kappa^{2}(\mathbf{b}_{1}\times\mathbf{b}_{2})^{2}}{b_{12}^{2}}j_{0}(\kappa b_{12})\right.$$

$$\left.+\frac{k\kappa}{b_{12}}\left[2(\mathbf{b}_{1}\cdot\mathbf{b}_{2})-3\frac{(\mathbf{b}_{1}\times\mathbf{b}_{2})^{2}}{b_{12}^{2}}\right]j_{1}(\kappa b_{12})\right\}_{k=k_{0}}.$$
(4.19)

Further corrections would be rather lengthy expressions involving Bessel functions of higher order. However, inelastic effects are manifested characteristically as corrections to wide angle scattering while interference itself, for wavelengths smaller than molecular dimensions, is a forward angle effect. Thus, in (4.19), we find the combinations $\kappa^2 j_0(\kappa b_{12})$ and $\kappa j_1(\kappa b_{12})$ whose derivatives with respect to k are small for both large and small angles. The static approximation,

$$a_{\rm int}^{(1,2)}(\theta) = 2a_1a_2j_0(\kappa b_{12})$$

σ

is in fact adequate for the analysis of many diffraction experiments.

5. SCATTERING BY A MONATOMIC GAS

Neutrons scattered by a gas exchange energy with the molecules via translational recoil. To study the effect of such processes upon the cross sections, we consider the scattering by a gas of single particles which have no internal degrees of freedom. The techniques thereby developed, which apply directly to the case of a monatomic gas, will later be of use in treating the scattering by complex molecules.

To obtain the cross section for direct scattering, we utilize the form (2.18) of the expectation value. For gas densities which are not too great, we may neglect the possibility that a scattering atom collides with another during the time associated with a neutron collision. Accordingly, an atom participating in a direct scattering event may be treated as a free particle with a Hamiltonian which consists solely of its kinetic energy. (However, certain interference effects, to be discussed later, require a more careful examination of interparticle interactions.) If M is the atomic mass, we have

$$H = p^2/2M, \quad H' = (\mathbf{p} - \mathbf{\kappa})^2/2M.$$

Choosing wave functions ψ for the atom which are eigenfunctions of momentum as well as energy, we have

$$\langle \boldsymbol{\psi} | e^{iHt} e^{-iH't} | \boldsymbol{\psi} \rangle = \exp[it(2\mathbf{p}\cdot\mathbf{\kappa}-\kappa^2)/2M]$$

where **p** now specifies a momentum eigenvalue. The particle momenta are distributed with the Boltzmann weighting factor $f_T(\mathbf{p}) = (2\pi MT)^{-\frac{3}{2}} \exp(-\frac{p^2}{2MT})$ for a gas in thermal equilibrium. Hence the thermal average becomes

$$\langle e^{iHt}e^{-iH't}\rangle_{T} = \int \langle \psi | e^{iHt}e^{-iH't} | \psi \rangle f_{T}(\mathbf{p}) \ (d\mathbf{p})$$
$$= \exp\left[\frac{-it\kappa^{2}}{2M} - \frac{t^{2}T\kappa^{2}}{2M}\right], \quad (5.1)$$

so that we obtain

$$\langle \sigma(\theta, \epsilon) \rangle_T = \frac{a^2}{2\pi} \frac{k}{k_0} \int \exp\left[-it\left(\epsilon + \frac{\kappa^2}{2M}\right) - \frac{t^2 T \kappa^2}{2M}\right] dt$$
$$= a^2 \left(\frac{M}{2\pi T \kappa^2}\right)^{\frac{1}{2}} \frac{k}{k_0} \exp\left[\frac{-M}{2T \kappa^2}\left(\epsilon + \frac{\kappa^2}{2M}\right)^2\right]. \quad (5.2)$$

The operator formalism thus provides a compact evaluation of the energy spectrum of neutrons scattered at any angle.

In order to carry out the integration over final energies, we introduce a dimensionless variable $z = k/k_0$ proportional to the scattered neutron momentum and a parameter α defined by

$$\alpha^2 = (mT)/(M\mathcal{E}_0).$$

An equivalent relation is

$$\alpha = (\frac{2}{3})^{\frac{1}{2}} (v_m / v_0), \tag{5.3}$$

where v_m is the mean thermal gas velocity and v_0 the incident neutron velocity. We also write λ for the mass ratio m/M. We find then, by (2.12) and (5.2),

$$\begin{split} \langle \sigma(\theta) \rangle_T &= \int_{-\infty}^{\infty} \langle \sigma(\theta, \epsilon) \rangle_T d\epsilon \\ &= \int_{0}^{\infty} \langle \sigma(\theta, \epsilon) \rangle_T (k_0^2/m) z dz \\ &= a^2 \pi^{-\frac{1}{2}} \alpha^{-1} \int_{0}^{\infty} \phi(\theta, z) \exp\{-[h(\theta, z)/\alpha]^2\} dz, (5.4) \end{split}$$

 $\phi(\theta,z) = z^2(1-2z\cos\theta+z^2)^{-\frac{1}{2}},$

where

$$h(\theta, z) = \frac{z^2 (1+\lambda) - 2\lambda z \cos\theta - (1-\lambda)}{2(z^2 - 2z \cos\theta + 1)^{\frac{1}{2}}}.$$
 (5.5)

Thus, the details of the differential cross section are determined by two physical parameters, the ratio λ of the masses of projectile and target, and the mean velocity ratio α .

In many experiments on neutron diffraction, the relevant velocity ratio is rather small. An expansion of (5.4) about the value assumed at $\alpha=0$ is therefore of practical value. The cross section for scattering by a monatomic gas may, in any event, be computed numerically from (5.4). However, the approximation for small α is applicable, as we shall see in the second paper, to the treatment of more complicated scattering systems where analogous but more complex integrals appear.

Because the integrand in question is not analytic at $\alpha = 0$, the expansion of $\langle \sigma(\theta) \rangle_T$ about this value need not be a simple Taylor series in powers of α . As α approaches zero, the function $\alpha^{-1} \exp[h(\theta, z)/\alpha]^2$ vanishes strongly for all values of z save those which satisfy

$$h(\theta, z) = 0. \tag{5.6}$$

Thus, the dominant contributions to the integral come from the neighborhoods of the roots of this equation. The expansion of (5.4) is accomplished by a device which approximates the integrand well in the vicinity of these roots, though crudely elsewhere.

The extrema of $h(\theta,z)$ as a function of z are determined from $(d/dz)h(\theta,z)=0$ which is a cubic equation. Its sole real root, which we denote by z_m , corresponds to a minimum value $h_m(\theta)$ of $h(\theta,z)$. If we define a new variable of integration s by

$$s = h(\theta, z) / \alpha,$$
 (5.7)

the implicitly defined function $z=z(\alpha s)$ has two branches. These are specified by the notation

$$z=z^{(+)}(\alpha s) \text{ for } z>z_m,$$

$$z=z^{(-)}(\alpha s) \text{ for } z$$

An inspection of (5.5) shows that z_m is positive and thus within the region of integration of (5.4) only when $\theta < 90^{\circ}$.

Introducing the change of variable into (5.4), we obtain

$$\frac{\langle \sigma(\theta) \rangle_T}{a^2 \pi^{-\frac{1}{2}}} = \int_{h_m(\theta)/\alpha}^{\infty} \exp(-s^2) \Phi^{(+)}(\alpha s) ds$$
$$+ \int_{h_m(\theta)/\alpha}^{h(\theta,0)/\alpha} \exp(-s^2) \Phi^{(-)}(\alpha s) ds \quad \text{for} \quad \theta < 90^\circ, \quad (5.8)$$

and

$$\frac{\langle \sigma(\theta) \rangle_T}{a^2 \pi^{-\frac{1}{2}}} = \int_{h(\theta,0)/\alpha}^{\infty} \exp(-s^2) \Phi^{(+)}(\alpha s) ds \text{ for } \theta \ge 90^\circ, (5.9)$$

where Let

$$\Phi^{(\pm)}(\alpha s) = \phi(z^{(\pm)}(\alpha s))dz^{\pm}/d(\alpha s).$$

$$\Phi^{(\pm)}(\alpha s) = \sum_{n=0}^{\infty} c_n^{(\pm)} \alpha^n s^n$$
 (5.10)

be the formal Taylor series for $\Phi^{(+)}$ and $\Phi^{(-)}$ about s=0. Upon substitution of these series into (5.8) and (5.9), we encounter integrals over terms of the general type $\exp(-s^2)\alpha^n s^n$. It is now assumed, as part of the approximation, that α is small enough to permit the replacement of the (negative) limits of integration $h(\theta,0)/\alpha$ and $h_m(\theta)/\alpha$ by $-\infty$. This corresponds to the neglect of contributions to the cross section of order $\alpha \exp[-h(\theta,0)/\alpha]^2$ and $\alpha \exp[-h_m(\theta)/\alpha]^2$.

Then the differential cross section takes the form of a power series in α^2 ,

$$\langle \sigma(\theta) \rangle_T = a^2 \pi^{-\frac{1}{2}} \int_{-\infty}^{\infty} \exp(-s^2) \Phi^{(+)}(\alpha s) ds$$

= $a^2 \pi^{-\frac{1}{2}} \sum_{n=0}^{\infty} \alpha^{2n} c_{2n}^{(+)} \Gamma(n+\frac{1}{2}),$ (5.11)

the odd powers of (αs) canceling in the integration. The computation of the initial terms of the series is straightforward, if tedious, and yields

$$\langle \sigma(\theta) \rangle_T = \langle \sigma^{(0)}(\theta) \rangle_T + \langle \sigma^{(1)}(\theta) \rangle_T + \cdots,$$
 (5.12)

$$\langle \sigma^{(0)}(\theta) \rangle_T = a^2 \frac{\left[\lambda \cos\theta + (1 - \lambda^2 \sin^2\theta)^{\frac{1}{2}}\right]^2}{(1 + \lambda)^2 (1 - \lambda^2 \sin^2\theta)^{\frac{1}{2}}},$$

$$\langle \sigma^{(1)}(\theta) \rangle_T = \frac{1}{2} a^2 \alpha^2 \left[1 + (\lambda + \frac{1}{2}\lambda^2)(3\cos^2\theta - 1) + \frac{3}{2}\lambda^3 \sin^2\theta(5\cos^2\theta - 1) + O(\lambda^4)\right].$$

Because of the complexity of the exact expression for $\langle \sigma^{(1)}(\theta) \rangle_T$ it is written here as a power series in the mass ratio. The cross section for $\alpha = 0$, or equivalently, zero temperature, is given by the leading term of (5.12). It is just the result ordinarily derived by kinematical transformation to the rest system of the scatterer from the center-of-mass system, in which the cross section is isotropic.

We have noted earlier the nonanalytic dependence on α of the function whose integral determines the cross section. In consequence, the series (5.11) likewise fails to behave analytically at $\alpha = 0$. Convergence of (5.11) for any range of values of α implies that $c_{2n}^{(+)}$ tends to zero with increasing n with sufficient rapidity to assure that $\sum c_{2n}^{(+)} s^{2n}$ is an entire function. Since the even part of $\Phi^{(+)}(\alpha s)$ is manifestly not an entire function, we recognize that (5.11) and (5.12) represent an asymptotic series.¹²

The utility of the expansion of the cross section is limited by its asymptotic character; its region of applicability is dependent on the accuracy desired. For scattering by particles whose mass is at least twice the neutron mass, the use of solely the zeroth-order term of (5.12) may be shown to cause an error of less than 1% when $\alpha < 1/10$. The cross section for somewhat heavier scatterers, e.g., $\lambda \leq 1/5$, is given to within 1%by retaining only the first two terms of (5.12), provided that $\alpha < 1/5$.

It may be remarked that the series (5.12) is precisely the result which would have emerged had the time expansion method of Sec. 4 been applied to the crosssection formula. The present method of deriving the expansion indicates the extent to which the former procedure represents the cross section accurately, and permits a study of the analytical properties of the series. These considerations will be seen in the next paper to apply equally well to the scattering by complex nuclei. The application of the time expansion method in the more general context yields additive corrections to the static cross section which are associated with the different molecular degrees of freedom with which the neutron may exchange energy. Among these corrections will appear the terms of (5.12).

The form of the approximate cross section in the limit of large α , i.e., high temperature or low incident neutron energy, is easily derived. If the region of integration in (5.4) is separated, for example, into $(0,\alpha^4)$ and (α^4,∞) , then for $\alpha\gg1$, the integral over the first region is less than

$$a^2 \pi^{-\frac{1}{2}} \alpha^{-\frac{1}{2}} \int_0^{\alpha^{\frac{1}{2}}} (z^2 - 2z \cos\theta + 1)^{-\frac{1}{2}} dz$$

which goes to zero, whereas the integral over the second region becomes

$$a^2\pi^{-\frac{1}{2}}\alpha^{-1}\int_{\alpha^{\frac{1}{4}}}^{\infty}z\,\exp[-z^2(1+\lambda)^2/4\alpha^2]dz.$$

Setting $y=z^2(1+\lambda)^2/4\alpha^2$, the latter integral becomes simply that of an exponential, and as α becomes large the lower limit goes to zero. Hence we find asymptotically for large α ,

$$\langle \sigma(\theta) \rangle_T \sim a^2 (1+\lambda)^{-2} (2\alpha \pi^{-\frac{1}{2}}).$$

For intermediate values of α a weak logarithmic singularity in the forward direction is contributed to the differential cross section by scatterers moving with nearly the same velocity and direction as the incoming neutron. The scattering by these particles is clearly elastic and confined to small angles.

Interference between the amplitudes scattered by independent atoms of the gas (the "outer effect") produces a change in the intensity well known in the case of x-ray diffraction. To calculate the intensity exactly, however, requires detailed information on the interparticle interactions and the position correlations they lead to. In particular, to treat the inelastic part exactly a knowledge of the time dependence of the position correlations is required as well. Since the interference effect varies as the square of the density of the scattering system it is not often of great importance in gases. Furthermore, at the neutron energies commonly employed the effect is confined to very small scattering angles. Since high accuracy is not required for its estimation we may employ the simple model introduced by Debye¹³ for the x-ray case. We assume the interparticle correlations vary relatively slowly with time and replace $\mathbf{r}_{\nu}(t)$ by $\mathbf{r}_{\nu}(0)$ in (2.15). If now the particles are treated as hard spheres of diameter d, whose overlap is forbidden, we have for the interference cross section per atom:

$$\sigma_{\rm int}(\theta) = -a^2 \rho \left(4\pi d^3/3\right) \left(\frac{1}{3}\kappa d\right)^{-1} j_1(\kappa d) \left| k = k_0. \quad (5.13)$$

Here ρ is the number of particles per unit volume, and j_1 is the first-order spherical Bessel function (4.18).

APPENDIX A

In Sec. 3, use was made of the thermally averaged expectation value of an exponential function of a harmonic oscillator coordinate. The theorem employed is

$$\langle \exp Q \rangle_T = \exp\{\frac{1}{2} \langle Q^2 \rangle_T\},$$
 (A.1)

where Q is any multiple of a harmonic oscillator coordinate. This relation, a corollary of Bloch's theorem, may be demonstrated quite directly by employing the operator representation for Q. We express Q in the form

$$Q = \lambda a + \lambda^* a^\dagger$$

¹³ P. Debye, Physik. Z. 28, 135 (1927).

¹² See, for example, E. T. Whittaker and G. N. Watson, *A Course in Modern Analysis* (The Macmillan Company, New York, 1945), pp. 150–159.

where a and a^{\dagger} are the familiar quantization operators, obeying the commutation relations (3.7), and the coefficients λ and λ^* may contain an oscillating time dependence. To find a matrix element of a function of Q it is convenient to carry out first a rearrangement of the operators a and a^{\dagger} which collects the powers in which they occur and fixes their order. For this purpose we employ the identity (3.9), by means of which the function expQ may be expressed as

$$\exp Q = \exp(\lambda^* a^{\dagger}) \exp(\lambda a) \exp(\frac{1}{2} |\lambda|^2),$$

a form which is chosen so that the annihilation operators precede the creation operators. The diagonal element of $\exp Q$ in the *n*th excitation state is then found by introducing the series expansions of the functions $\exp(\lambda^*a^{\dagger})$ and $\exp(\lambda a)$, which may be terminated at the *n*th order since no more than *n* quanta may be annihilated.

$$\langle \exp Q \rangle_n = \sum_{p=0}^n \frac{|\lambda|^{2p}}{(p!)^2} \langle (a^{\dagger})^p (a)^p \rangle_n \exp(\frac{1}{2}|\lambda|^2). \quad (A.2)$$

Using the elementary rules for the application of the quantization operators to the state vectors Ψ_n ,

$$a\Psi_n = n^{\frac{1}{2}}\Psi_{n-1}, \quad a^{\dagger}\Psi_n = (n+1)^{\frac{1}{2}}\Psi_{n+1},$$

we have

$$\langle (a^{\dagger})^{p}a^{p}\rangle_{n} = n!/(n-p)!$$

which, substituted in (A.2), yields

$$\langle \exp Q \rangle_n = \sum_{p=0}^n \frac{n!}{(p!)^2(n-p)!} |\lambda|^{2p} \exp(\frac{1}{2}|\lambda|^2).$$
 (A.3)

The thermal average of the operator $\exp Q$ is given by

$$\langle \exp Q \rangle_T = \operatorname{Trace}[\exp(-H/T) \exp Q] / \operatorname{Trace}[\exp(-H/T)]$$
$$= [1 - \exp(-\omega/T)] \sum_n \exp(-n\omega/T)$$
$$\times \langle \exp Q \rangle_n. \quad (A.4)$$

Letting $z = \exp(-\omega/T)$ and combining (A.4), (A.3), we have

$$\langle \exp Q \rangle_T = (1-z) \sum_{n,p} \frac{n! |\lambda|^{2p} z^n}{(p!)^2 (n-p)!} \exp(\frac{1}{2} |\lambda|^2)$$

= $(1-z) \sum_{n,p=0}^{\infty} \frac{|\lambda|^{2p} z^p}{(p!)^2} \frac{d^p}{dz^p} z^n \exp(\frac{1}{2} |\lambda|^2).$

In the latter form, the summation over n is seen to be that of a geometric series, which yields

$$\begin{split} \langle \exp Q \rangle_T &= (1-z) \sum_{p=0}^{\infty} \frac{|\lambda|^{2p} z^p}{(p!)^2} \frac{d^p}{dz^p} (1-z)^{-1} \exp(\frac{1}{2} |\lambda|^2) \\ &= \sum (p!)^{-1} |\lambda|^{2p} z^p (1-z)^{-p} \exp(\frac{1}{2} |\lambda|^2) \\ &= \exp\{|\lambda|^2 [\frac{1}{2} + z(1-z)^{-1}]\}. \end{split}$$

The argument of the latter exponential may immediately be identified as $\frac{1}{2}\langle Q^2 \rangle_T$ for

$$\langle Q^2 \rangle_T = |\lambda|^2 \langle 2n+1 \rangle_T = 2 |\lambda|^2 [\frac{1}{2} + z(1-z)^{-1}]$$

which establishes directly the result (A.1).

APPENDIX B

A classical approximation to the cross section $\sigma(\theta, \epsilon)$ for scattering of neutrons at a given angle and at a given energy, valid for large $k_0 b$, has already been derived in the text. To compute $\sigma^{(1)}(\theta, \epsilon)$, which includes the first quantum-mechanical correction, we begin by examining the operator function $f(\lambda)$,

$$f(\lambda) = e^{-\lambda C} e^{\lambda (A+B+C)}$$

where, in terms of previous notation, $A = H - E_l$, $B = (-i\kappa/b\mathfrak{M})\beta$, $C = (\kappa^2/2\mathfrak{M})\gamma$. The classical approximation consisted of treating the term C exactly and neglecting A and B. To find corrections of relative order $(\kappa b)^{-2}$, we note that C contains implicitly a factor of k_0^2 . Hence we expand $f(\lambda)$ to first order in A and second order in B. Since the commutators [[A,C],C] and [B,C] commute with C, we find, after a short calculation,

$$f(\lambda) = \sum_{n=0}^{\infty} \lambda^n f^{(n)}(0) / n!$$

= 1+\lambda (A+B)+\frac{1}{2}\lambda^2([A+B,C]+B^2)
+\frac{1}{6}\lambda^3([[A,C],C]+2[B,C]B+B[B,C])
+\frac{1}{8}\lambda^4([B,C])^2.

Noting that the expectation values of terms containing either a factor of A adjacent to a wave function or odd powers of B vanish, we have

$$\begin{aligned} \langle \chi \rangle_{l} &= \langle e^{-it(A+B+C)} \rangle_{l} = \frac{1}{2} \int_{-1}^{1} \exp[-it(1-x^{2})\kappa^{2}/2\mathfrak{M}] \\ &\times [1+(-it)^{2}X_{2}(x)+(-it)^{3}X_{3}(x)+(-it)^{4}X_{4}(x)] dx \end{aligned}$$

with

$$\begin{split} X_2(x) &= \frac{1}{4} \kappa^2 (\mathfrak{M}b)^{-2} [3 - 7x^2 + (1 - x^2)l(l+1)], \\ X_3(x) &= \frac{1}{6} \kappa^4 \mathfrak{M}^{-3} b^{-2} (7x^4 - 8x^2 + 1), \\ X_4(x) &= -\frac{1}{8} \kappa^6 \mathfrak{M}^{-4} b^{-2} x^2 (1 - x^2)^2. \end{split}$$

Employing the formal relation

$$-ite^{-i\epsilon t} = (\partial/\partial \epsilon)e^{-i\epsilon t},$$

we infer

$$\sigma^{(1)}(\theta, \epsilon) = \frac{1}{2}a^2(k/k_0) \int_{-1}^{1} \left[1 + (\partial/\partial \epsilon)^2 X_2(x) + (\partial/\partial \epsilon)^3 X_3(x) + (\partial/\partial \epsilon)^4 X_4(x) \right]$$

 $\times \delta(\epsilon + \kappa^2 (1-x^2)/2\mathfrak{M}) dx$

where
$$\begin{split} &= \! \left[1 \!+\! \frac{\partial^2}{\partial \epsilon^2} X_2(\bar{x}) \!+\! \frac{\partial^3}{\partial \epsilon^3} X_3(\bar{x}) \!+\! \frac{\partial^4}{\partial \epsilon^4} X_4(\bar{x}) \right] \! \sigma_{cl}(\theta, \epsilon), \\ & \bar{x}^2 \!=\! \kappa^{-2} (\kappa^2 \!+\! 2\mathfrak{M}\epsilon). \end{split}$$

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Applying the indicated differential operations to all explicit occurrences of ϵ lying to the right of the operators, we obtain the expression for $\sigma^{(1)}(\theta, \epsilon)$ given in the text.

APPENDIX C

The approximations thus far used in treating the rotator have been aimed at summing the effects of the large number of rotational transitions which occur for all but very small neutron energies. When the neutron energies are low the convergence of these methods may be slow or may fail entirely. To supplement them in this region it is convenient to develop the cross section as an expansion in positive powers of $(k_0b)^2$. This may be accomplished either by summing explicitly the partial cross sections due to the individual rotational transitions, which are few in number in the lowest approximations, or by using operator techniques similar to those described earlier. For example in the approximation to order $(k_0b)^2$ the operator $\exp[\lambda(A+B+C)]$ (using notation defined in Appendix B) is expanded to second order in B and first order in C before carrying out the required integrations. In this approximation the scattering cross section of a rotator of angular momentum l is

$$\begin{aligned} (\theta)/a^{2} &= 1 - \frac{2}{3} (k_{0}b)^{2} (1 - \cos\theta) \\ &+ \frac{2}{3} l(2l+1)^{-1} (k_{0}b)^{2} [1 + 2l\tau]^{\frac{1}{2}} \\ &\times \{1 + l\tau - \cos\theta [1 + 2l\tau]^{\frac{1}{2}}\} \\ &+ \frac{2}{3} (l+1) (2l+1)^{-1} (k_{0}b)^{2} [1 - 2(l+1)\tau]^{\frac{1}{2}} \\ &\times \{1 - (l+1)\tau - \cos\theta [1 - 2(l+1)\tau]^{\frac{1}{2}}\} \end{aligned}$$

where $\tau = m/(\mathfrak{M}k_0^2b^2)$. The contributions on the first line represent the partial cross section for the transition $l \rightarrow l$, while the remaining terms correspond to $l \rightarrow l-1$ and $l \rightarrow l+1$ respectively. For $(m/\mathfrak{M}) \ll 1$, it is of interest to expand this result in the mass ratio. We obtain

$$\sigma_{l}(\theta) = 1 - (4/3) (m/\mathfrak{M}) (1 - \cos\theta) + \frac{1}{3} (m/\mathfrak{M})^{2} (l^{2} + l + 1) (k_{0}b)^{-2} + \cdots$$

which is seen to contain the terms which dominate at low energies in the more general expansion (4.15). Since the present series fails to converge for values of τ larger than $(2l+2)^{-1}$, the expansion (4.15) may be expected to break down for $(k_0b)^2 < (m/\mathfrak{M})(2l+2)$ or for energies \mathcal{E}_0 less than the rotational level spacing $\Delta E_l = E_{l+1} - E_l$. The expansion in positive powers of k_0b , together with the earlier methods, covers the entire range of neutron energies of interest.

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Neutron Diffraction by Gases

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The methods developed in the previous paper for the treatment of neutron scattering by simplified molecular models are extended to form a unified description of scattering by real molecules. The generalization takes into account the influence on scattering of the interactions that appear among the different molecular degrees of freedom. The effect of spin correlations within molecules due to symmetries occurring in the presence of like nuclei is explicitly determined. The slow-neutron cross section of methane is calculated and compared with experiment.

1. INTRODUCTION

MOLECULAR scattering of neutrons is of principal interest at neutron energies low enough to produce significant interference effects. These energies are in practice comparable with those of thermally excited molecular degrees of freedom. Therefore, single collisions often suffice to alter the neutron energy by amounts comparable to or larger than its initial value.

In the preceding paper,¹ the scattering of slow neutrons by various simplified molecular systems was studied by a method which facilitates the treatment of inelastic processes. The method utilizes operator representations for the dynamical variables of the scattering system and yields simple formal expressions for the cross sections. The systems treated include coupled harmonic oscillators, rotators, and a gas of particles in thermal equilibrium.

By combining these results we construct, in the following sections, a unified description of neutron diffraction by gases. Certain new features are encountered, principally the interactions of the various dynamical modes, e.g., the rotation of the axes of vibration during a collision. At the same time, considerations of the spin dependence of neutron scattering and of spin correlations within the molecules are introduced.

The diffraction of neutrons by methane is studied in detail in the last section. The inelasticity of the scattering by this gas is an important feature in virtue of the small rotational inertia of the methane molecules.

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A. C. Zemach and R. J. Glauber, preceding paper [Phys. Rev.
 100, 118 (1955)], hereafter called I. In references to equations of this paper, the numeral I will precede the equation numbers.