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

$$\begin{aligned}\sigma_l(\theta)/a^2=1-\tfrac{2}{3}(k_0b)^2(1-\cos\theta)&\\ +\tfrac{2}{3}l(2l+1)^{-1}(k_0b)^2[1+2l\tau]^{\frac{1}{2}}&\\ \times\{1+l\tau-\cos\theta[1+2l\tau]^{\frac{1}{2}}\}&\\ +\tfrac{2}{3}(l+1)(2l+1)^{-1}(k_0b)^2[1-2(l+1)\tau]^{\frac{1}{2}}&\\ \times\{1-(l+1)\tau-\cos\theta[1-2(l+1)\tau]^{\frac{1}{2}}\}&\end{aligned}$$

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

$$\sigma_l(\theta)=1-(4/3)(m/\mathfrak{M})(1-\cos\theta)+\tfrac{1}{3}(m/\mathfrak{M})^2(l^2+l+1)(k_0b)^{-2}+\cdots$$

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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* National Science Foundation Predoctoral Fellow.

[1] 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.

2. SPIN DEPENDENCE

In Sec. 2 of I we presented the differential cross section as an integral over time and energy transfer of the expectation values of certain functions of the molecular coordinates. If $\mathbf{r}_\nu$ denotes the position vector of the νth nucleus of a specified molecule, the expectation values take the form

$$\langle\chi_{\nu\nu'}\rangle=\langle\psi|\,a_\nu a_{\nu'}\exp[i\boldsymbol{\kappa}\cdot\mathbf{r}_\nu(t)]\times\exp[-i\boldsymbol{\kappa}\cdot\mathbf{r}_{\nu'}(0)]\,|\psi\rangle_T. \qquad (2.1)$$

These describe both direct scattering ($\nu=\nu'$) and interference effects ($\nu\neq\nu'$) that take place within a single molecule of the gas. Since the scattering lengths a_ν and $a_{\nu'}$ are in general spin-dependent, they have been included within the expectation value brackets.

To describe the spin-dependence more explicitly, we let $\mathbf{S}_\nu$ be the spin of nucleus ν, $\mathbf{s}$ the neutron spin, and $a_\nu^{(+)}$ and $a_\nu^{(-)}$ the scattering lengths for the system of neutron plus νth nucleus in states whose spins are $S_\nu+\frac{1}{2}$ and $S_\nu-\frac{1}{2}$ respectively. Then the scattering lengths appearing in (2.1) are spin-dependent operators which may be written

$$a_\nu=a_\nu^{(+)}\pi_\nu^{(+)}+a_\nu^{(-)}\pi_\nu^{(-)}, \qquad (2.2)$$

in terms of projection operators $\pi_\nu^{(+)}$, $\pi_\nu^{(-)}$ to the parallel and antiparallel spin states. The projection operators are easily expressed in terms of spin vectors:

$$\pi_\nu^{(+)}=(1+S_\nu+2\mathbf{s}\cdot\mathbf{S}_\nu)(1+2S_\nu)^{-1},$$
$$\pi_\nu^{(-)}=(S_\nu-2\mathbf{s}\cdot\mathbf{S}_\nu)(1+2S_\nu)^{-1}.$$

With the customary definitions of the coherent scattering amplitude A_ν,

$$A_\nu=\frac{S_\nu+1}{2S_\nu+1}a_\nu^{(+)}+\frac{S_\nu}{2S_\nu+1}a_\nu^{(-)},$$

and the incoherent scattering amplitude C_ν,

$$C_\nu=\frac{[S_\nu(S_\nu+1)]^{\frac{1}{2}}}{2S_\nu+1}(a_\nu^{(+)}-a_\nu^{(-)}),$$

the scattering length (2.2) becomes

$$a_\nu=A_\nu+2C_\nu(\mathbf{S}_\nu\cdot\mathbf{s})[S_\nu(S_\nu+1)]^{-\frac{1}{2}}. \qquad (2.3)$$

Employing the neutron spin averages appropriate to the diffraction of unpolarized neutron beams,

$$\tfrac{1}{2}\textstyle\sum_{s=\pm\frac{1}{2}}\langle\mathbf{s}\cdot\mathbf{S}\rangle_\nu=0,\quad \tfrac{1}{2}\textstyle\sum_{s=\pm\frac{1}{2}}\langle(\mathbf{s}\cdot\mathbf{S}_\nu)(\mathbf{s}\cdot\mathbf{S}_{\nu'})\rangle_5=\tfrac{1}{4}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'}),$$

we find, inserting (2.3) into (2.1),

$$\langle\chi_{\nu\nu'}\rangle=\langle\psi|\,f_{\nu\nu'}\{A_\nu A_{\nu'}+C_\nu C_{\nu'}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})\times[S_\nu(S_\nu+1)S_{\nu'}(S_{\nu'}+1)]^{-\frac{1}{2}}\}\,|\psi\rangle_T, \qquad (2.4)$$

where

$$f_{\nu\nu'}=\exp(i\mathbf{k}\cdot\mathbf{r}_\nu(t))\exp[-i\mathbf{k}\cdot\mathbf{r}_{\nu'}(0)].$$

The wave functions for a molecule containing identical nuclei are restricted by the requirements of nuclear statistics. Such restrictions introduce correlations of the otherwise independent nuclear spin orientations. In the absence of symmetry restrictions, each molecular wave function ψ may be chosen as a product

$$\psi=\phi\xi \qquad (2.5)$$

of a spatial function ϕ and a spin function ξ. Averages over molecular states then separate naturally into spatial averages and spin averages, performed independently of each other. In this case, we have

$$\langle\xi|\,(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})\,|\xi\rangle_{\mathrm{Av}}=S_\nu(S_\nu+1)\delta_{\nu\nu'}, \qquad (2.6)$$

and hence

$$\begin{aligned}\langle\chi_{\nu\nu'}\rangle&=A_\nu A_{\nu'}\langle\phi|\,f_{\nu\nu'}\,|\phi\rangle_T,\quad \nu\neq\nu',\\ \langle\chi_{\nu\nu}\rangle&=B_\nu^2\langle\phi|\,f_{\nu\nu}\,|\phi\rangle_T,\end{aligned} \qquad (2.7)$$

where

$$B_\nu^2=A_\nu^2+C_\nu^2.$$

When spin correlations are present, we find a further contribution to the expectation values $\langle\chi_{\nu\nu'}\rangle$ whose origin may be seen in (2.4). This takes the form of an additional interference effect between the amplitudes scattered by the identical nuclei. It contributes to $\langle\chi_{\nu\nu'}\rangle$ the term

$$C_\nu^2\langle\psi|\,f_{\nu\nu'}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})\,|\psi\rangle_T/S_\nu(S_\nu+1), \qquad (2.8)$$

which is seen to depend on the incoherent, rather than the coherent scattering amplitude. The correlation effect is most familiar in neutron scattering by ortho- and para-hydrogen. In diatomic molecules containing like nuclei, parity considerations imply a complete correlation of nuclear spins in an energy eigenstate.

The explicit evaluation of the spin correlation effect for molecules of arbitrary symmetry type is carried out in the Appendix. The result can be summarized as follows: Let η denote the number of occurrences of a given type of nucleus in a molecule and let $g_E(\Omega)$ be the fraction of molecular wave functions[2] of energy E for which the total spin of the like nuclei in question is Ω. Then, as a consequence of the nuclear symmetry, the value of $\langle\chi_{\nu\nu'}\rangle$ given by (2.7) must be replaced by the thermal average of

$$\left\{A_\nu^2+C_\nu^2\sum_\Omega g_E(\Omega)\frac{\Omega(\Omega+1)-\eta S_\nu(S_\nu+1)}{S_\nu(S_\nu+1)\eta(\eta-1)}\right\}\times\langle\phi|\,f_{\nu\nu'}\,|\phi\rangle, \qquad (2.9)$$

when ν and ν' denote identical nuclei. Evaluations of this expression in some particular cases are also given in the Appendix. It is noted that in all molecules save those of the smallest rotational inertia (e.g., H_2 and D_2) the close spacing of different rotational levels compensates the exclusion of certain values of Ω for particular levels. As a result, in all but the lightest molecules the correlation effect is confined to very low temperatures, and we may neglect it in further work.

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[2] Tables of $g_E(\Omega)$ for all molecules of common symmetry types are given by E. B. Wilson, J. Chem. Phys. 3, 276 (1935).

3. MOLECULAR SCATTERING

In order to treat the dynamical modes of the molecules simultaneously, it is convenient to separate the nuclear coordinates into a sum of terms corresponding to the different degrees of freedom. The position vector $\mathbf{r}_\nu$ of nucleus ν may be represented as

$$\mathbf{r}_\nu = \mathbf{R} + \mathbf{b}_\nu + \mathbf{u}_\nu,$$

where $\mathbf{R}$ is the position vector of the molecular center of mass, $\mathbf{b}_\nu$ is the displacement of the equilibrium position of the νth nucleus from the mass center, and $\mathbf{u}_\nu$ the instantaneous displacement of this nucleus from its equilibrium position due to vibration. The molecular Hamiltonian is taken as the sum of three parts

$$H = H_{\text{vib}} + H_{\text{rot}} + H_{\text{trans}},$$

which are separately the Hamiltonians for the translational, rotational, and vibrational motions of the molecule. If we neglect rotation-vibration interactions in constructing the molecular wave functions, these assume the form

$$\psi = \psi_{\text{spin}} \psi_{\text{vib}} \psi_{\text{rot}} \psi_{\text{trans}}, \qquad (3.1)$$

in which the effects of different dynamical modes are explicitly separated. In a treatment of the outer effect, (3.1) must, of course, be supplemented by a specification of intermolecular correlation.

We introduce normal coordinates, following the notation of I, but abbreviating the dynamical variables evaluated at $t=0$ by writing, e.g., $\mathbf{r}_\nu(0) = \mathbf{r}_\nu$, etc. Then

$$\begin{aligned} \mathbf{r}_\nu &= \mathbf{R} + \mathbf{b}_\nu + \textstyle\sum_\lambda \mathbf{c}_\nu^{(\lambda)} q_\lambda, \\ \mathbf{r}_\nu(t) &= \mathbf{R}(t) + \mathbf{b}_\nu(t) + \textstyle\sum_\lambda \mathbf{c}_\nu^{(\lambda)}(t) q_\lambda(t). \end{aligned} \qquad (3.2)$$

The coordinates of the type $\mathbf{R}$ and q commute with each other and with $\mathbf{b}$ and $\mathbf{c}$, regardless of the times at which they are evaluated. The coefficient vectors $\mathbf{c}$ used in defining the normal modes, like the vectors $\mathbf{b}$, are fixed in the molecule and rotate with it. The behavior the latter vectors have in common as rotator coordinates is reflected by their failure to commute save when evaluated at equal times. As a consequence, rotation and vibration effects cannot be completely separated from one another in the calculation of expectation values. This feature is due to the rotation of the axes of vibration in the course of a scattering event. It is to be distinguished from the effect of rotation-vibration interaction alluded to in the previous paragraph. The latter depends, characteristically, on the rotation which takes place during a vibrational period.

The substitution of (3.2) into the expression (2.1) for $\langle \chi_{\nu\nu'} \rangle$ provides a partial separation of the influence of the different degrees of freedom because of the separability of the wave functions (3.1). Thus, by (2.7) and (I, 5.1), we have

$$\begin{aligned} \langle \chi_{\nu\nu'} \rangle = (A_\nu A_{\nu'} + \delta_{\nu\nu'} C_\nu C_{\nu'}) & \\ \times \exp[-(it\kappa^2 + t^2\kappa^2 T)/2M] & \\ \times \langle \psi_{\text{rot}} | \exp(i\boldsymbol{\kappa}\cdot\mathbf{b}_\nu(t)) V_{\nu\nu'} & \\ \times \exp(-i\boldsymbol{\kappa}\cdot\mathbf{b}_{\nu'}) | \psi_{\text{rot}} \rangle_T, & \qquad (3.3) \end{aligned}$$

where

$$\begin{aligned} V_{\nu\nu'} = \langle \psi_{\text{vib}} | \exp[i\boldsymbol{\kappa}\cdot \textstyle\sum_\lambda \mathbf{c}_\nu^{(\lambda)}(t) q_\lambda(t)] & \\ \times \exp[-i\boldsymbol{\kappa}\cdot \textstyle\sum_\lambda \mathbf{c}_{\nu'}^{(\lambda)} q_\lambda] | \psi_{\text{vib}} \rangle_T. & \qquad (3.4) \end{aligned}$$

In a calculation of the vibrational expectation values $V_{\nu\nu'}$, the noncommutativity of $\mathbf{c}_\nu^{(\lambda)}(t)$ and $\mathbf{c}_{\nu'}^{(\lambda)}$ must be respected. The time-dependent coefficient vectors all lie to the left of their time-independent counterparts and an expression for $V_{\nu\nu'}$ inserted into (3.3) must retain the ordering of the coefficient vectors. With this understanding we infer, by comparison of (3.4) with Eqs. (I, 3.6) and (I, 3.19) that

$$\begin{aligned} V_{\nu\nu'} = \prod_\lambda \sum_{n_\lambda} \exp[n_\lambda \omega_\lambda (it - \tfrac{1}{2}T^{-1})] & \\ \times \exp\{-\tfrac{1}{4}\omega_\lambda^{-1} [\boldsymbol{\kappa}\cdot\mathbf{c}_\nu^{(\lambda)}(t)]^2 \coth(\omega_\lambda/2T)\} & \\ \times I_{n_\lambda}\left(\frac{(\boldsymbol{\kappa}\cdot\mathbf{c}_\nu^{(\lambda)}(t))(\boldsymbol{\kappa}\cdot\mathbf{c}_{\nu'}^{(\lambda)})}{2\omega_\lambda \sinh(\omega_\lambda/2T)} \right) & \\ \times \exp\{-\tfrac{1}{4}\omega_\lambda^{-1} [\boldsymbol{\kappa}\cdot\mathbf{c}_{\nu'}^{(\lambda)}]^2 \coth(\omega_\lambda/2T)\}. & \qquad (3.5) \end{aligned}$$

The explicit time-ordering of the argument of the modified Bessel function may be accomplished by introducing the power series representation of the latter. In practice, the arguments of the I_n are often so small that only the leading term of each series is required.

The cross section for scattering by an ideal gas in thermal equilibrium is then given by

$$\langle \sigma(\theta) \rangle_T = \textstyle\sum_{\nu,\nu'} (2\pi)^{-1} \int\int (k/k_0) e^{-i\epsilon t} \langle \chi_{\nu\nu'} \rangle dt d\epsilon, \qquad (3.6)$$

with $\langle \chi_{\nu\nu'} \rangle$ determined from (3.3). The outer effect, which is due to the mutual interactions of gas molecules, is considered at the close of this section.

We have seen in the previous paper how expressions similar to (3.3) can be evaluated with the aid of various approximate techniques. The characteristics of the scattering of neutrons by the rotational and translational degrees of molecular freedom were investigated separately in the limits of both high and low incident neutron energy. The method of most general utility is based on an expansion of the expectation values $\langle \chi_{\nu\nu'} \rangle$ in powers of t and has already been applied in the discussion of rotator scattering. With the aid of this device, the cross section is represented as a series whose initial term is the familiar static approximation. The succeeding terms describe the consequences of energy transfer between the neutron and the scatterer. Since the various inelastic processes contribute additively to the cross section, the time expansion method is well suited to the simultaneous treatment of neutron interactions with different degrees of freedom.

As a preliminary step in the application of this procedure, it is convenient to effect a transformation of variables in (3.6). A consequence of this transformation, as will appear later, is the more compact expression of certain quantities which the time expansion method would otherwise yield as infinite series. We begin by introducing a center-of-mass coordinate frame. We take this to be the frame of reference which moves with the mass center of the system formed by an incident neutron plus a molecule at rest in the laboratory. The change of variable contemplated is from $\mathbf{k}$, $\mathbf{k}_0$, ϵ to similarly defined quantities which, however, are referred to the center-of-mass frame. The velocity of the center-of-mass frame relative to the laboratory is (μ/M) times the incident neutron velocity, where μ is the reduced mass of the neutron-molecule system. Then the transformation of momenta becomes

$$\begin{aligned}\mathbf{k}'&=\mathbf{k}-(\mu/M)\mathbf{k}_0,\\ \mathbf{k}_0'&=\mathbf{k}_0-(\mu/M)\mathbf{k}_0=(\mu/m)\mathbf{k}_0,\end{aligned}\tag{3.7}$$

while the momentum transfer $\boldsymbol{\kappa}$ is expressible as

$$\boldsymbol{\kappa}=\mathbf{k}'-\mathbf{k}_0'.$$

The energy transfer ϵ', given by

$$\epsilon'=(2\mu)^{-1}(k'^2-k_0'^2),$$

differs from ϵ by a term equal to the recoil energy of the molecule,

$$\epsilon'=\epsilon+\kappa^2/2M.\tag{3.8}$$

The angle of scattering θ' in the center-of-mass coordinate system is determined from the well-known relation

$$\tan\theta=\frac{\sin\theta'}{\cos\theta'+(mk_0')/(Mk')},$$

which implies $\theta'=\theta+\sin^{-1}[(mk_0'/Mk')\sin\theta]$. By formal algebraic manipulation, we find that

$$\frac{k}{k_0}d\epsilon=\left(\frac{\mu}{m}\right)^2\frac{k'}{k_0'}\left(\frac{k^2}{k'^2}\frac{dk}{dk'}\right)d\epsilon'.\tag{3.9}$$

The factor

$$Z(\theta,k'/k_0')=(k^2/k'^2)(dk/dk')$$

may be determined from (3.7). It is a double-valued function,

$$Z(\theta,k'/k_0')=\left\{\frac{mk_0'}{Mk'}\cos\theta\pm\left[1-\left(\frac{mk_0'}{Mk'}\right)^2\sin^2\theta\right]^{\frac{1}{2}}\right\}^2\times\left[1-\left(\frac{mk_0'}{Mk'}\right)^2\sin^2\theta\right]^{-\frac{1}{2}},\tag{3.10}$$

in which the sign preceding the square root is negative when $\theta<90°$ and $k/k_0<(\mu/M)\cos\theta$, and positive otherwise.

We have then, by (3.6), (3.8), and (3.9),

$$\langle\sigma(\theta)\rangle_T=\textstyle\sum_{\nu,\nu'}(2\pi)^{-1}(\mu/m)^2\times\int\int(k'/k_0')Ze^{-i\epsilon' t}\langle\chi_{\nu\nu'}'\rangle dtd\epsilon',\tag{3.11}$$

where $\langle\chi_{\nu\nu'}'\rangle$ is obtained from $\langle\chi_{\nu\nu'}\rangle$ by dropping the factor $\exp(-it\kappa^2/2M)$. The arguments of the Z function have been omitted for brevity.

If we now introduce the time expansion

$$\langle\chi_{\nu\nu'}'\rangle=\sum_{n=0}^{\infty}c_n(it)^n,$$

we obtain, in analogy to (I, 4.7),

$$\begin{aligned}\langle\sigma(\theta)\rangle_T&=\textstyle\sum_{\nu,\nu'}(\mu/m)^2\int(k'/k_0')Z\sum_n(-1)^nc_n\delta^{(n)}(\epsilon')d\epsilon'\\ &=\sum_{\nu,\nu',n}\left(\frac{\mu}{m}\right)^2\left(\frac{\mu}{k'}\frac{d}{dk'}\right)^n[(k'/k_0')Zc_n]_{k'=k_0'}.\end{aligned}\tag{3.12}$$

The $n=0$ term of (3.12) represents the static approximation to the cross section. It follows directly from (3.11) by ignoring the time dependence in $\langle\chi_{\nu\nu'}'\rangle$. A delta function of energy transfer thereby emerges from the integration over t. The succeeding terms of (3.12) are inelastic corrections which take into account the role of scattering processes in which an appreciable fraction of the incident neutron energy is exchanged between the neutron and the molecule.

The apparent inelasticity of scattering processes depends on the coordinate system in which they are viewed. A static approximation applied directly to (3.6) would signify elasticity in the laboratory and the inelastic corrections would include infinitely many terms arising from the time expansion of $\exp(-it\kappa^2/2M)$. These terms constitute the development of $Z(\theta,1)$ in powers of (m/M).

The terms of (3.12) which are due to the thermal motion of the gas molecules form a power series in α^2. They are, in fact, the terms appearing in (I, 5.12). The inelastic effects of rotator scattering are given by the correction terms of (I, 4.15), (I, 4.19) provided that the neutron mass in the latter formulas is replaced by the reduced mass, and additional factors of $(\mu/m)^2Z(\theta,1)$ are appended. Finally, there are additional corrections in which the effects of the translational, vibrational, and rotational degrees of freedom are mixed.

When vibration effects and time dependence are both ignored in the rotational expectation values, the latter reduce to spherical Bessel functions:

$$\langle\psi_{\mathrm{rot}}|\exp[i\boldsymbol{\kappa}\cdot(\mathbf{b}_\nu-\mathbf{b}_{\nu'})]|\psi_{\mathrm{rot}}\rangle_T=j_0(\kappa b_{\nu\nu'}),\quad \mathbf{b}_{\nu\nu'}=\mathbf{b}_\nu-\mathbf{b}_{\nu'}.$$

If, in addition, we were to neglect the translational motion of the molecules (i.e., $\alpha\rightarrow0$), then (3.11) would become

$$\langle\sigma(\theta)\rangle_T=(\mu/m)^2Z(\theta,1)\textstyle\sum_{\nu,\nu'}(A_\nu A_{\nu'}+C_\nu C_{\nu'}\delta_{\nu\nu'})\times j_0(\kappa b_{\nu\nu'})|_{k'=k_0'}.\tag{3.13}$$

Equation (3.13) is the "semiclassical approximation" of Spiers,[3] and Alcock and Hurst[4] for scattering by rigid

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[3] J. A. Spiers, Phys. Rev. **75**, 1765 (1949); National Research Council (Canada) Report CRT-417, April, 1949; N.R.C. No. 1940 (unpublished).

[4] N. Z. Alcock and D. G. Hurst, Phys. Rev. **75**, 1609 (1949) and Phys. Rev. **83**, 1100 (1951).

molecules. It is applicable when the energy (measured in the center-of-mass frame) gained or lost by the rotational modes and by the translational modes of the molecule is a small fraction of the incident neutron energy. The considerations of I have shown that the first condition rests upon the validity of the inequalities

$$(\mu/\mathfrak{M}_\nu)\ll 1, \quad (\langle \Delta E_l\rangle_T/\mathcal{E}_0)\ll 1,$$

where the $\mathfrak{M}_\nu$ are appropriately defined rotational masses of the nuclei. The second condition depends on

$$(mT/M\mathcal{E}_0)\ll 1.$$

When these conditions do not obtain, corrections to the semiclassical approximation may be derived in the manner prescribed by (3.12).

In treating the outer effect, we shall be content with the Debye approximation (I, 5.13). The independence of the internal coordinates of different molecules permits the separation of internal expectation values. The expectation value $\langle\zeta_{\nu\nu'}\rangle$ for interference between nuclei ν, ν' of different molecules becomes

$$\begin{aligned}\langle\zeta_{\nu\nu'}\rangle = -A_\nu A_{\nu'}[(4/3)\pi\rho d^3](\tfrac{1}{3}\kappa d)^{-1}j_1(\kappa d)&\\ \times\langle\psi_{\text{rot}}|V_\nu{}^* \exp(i\boldsymbol{\kappa}\cdot\mathbf{b}_\nu)|\psi_{\text{rot}}\rangle_T&\\ \times\langle\psi_{\text{rot}}|V_{\nu'}\exp(-i\boldsymbol{\kappa}\cdot\mathbf{b}_{\nu'})|\psi_{\text{rot}}\rangle_T&,\end{aligned}$$

where

$$V_\nu=\langle\psi_{\text{vib}}|\exp(-i\boldsymbol{\kappa}\cdot\textstyle\sum_\lambda \mathbf{c}_\nu{}^{(\lambda)}q_\lambda|\psi_{\text{vib}}\rangle_T.$$

The quantity V_ν is a particular form of the general expression (3.4). It is obtained from the latter by placing each $\mathbf{c}_\nu{}^{(\lambda)}(t)$ therein equal to zero. Since $I_0(0)=1$, $I_n(0)=0$ for $n\neq 0$,

$$V_\nu=\textstyle\prod_\lambda \exp[-\tfrac{1}{4}\omega_\lambda{}^{-1}(\boldsymbol{\kappa}\cdot\mathbf{c}_\nu{}^{(\lambda)})^2\coth(\omega_\lambda/2T)]|_{k=k_0}.$$

Then the outer scattering term which adds to (3.6) to complete the expression for the differential cross section is

$$\begin{aligned}-\textstyle\sum_{\nu,\nu'} A_\nu A_{\nu'}[(4/3)\pi\rho d^3](\tfrac{1}{3}\kappa d)^{-1}j_1(\kappa d)&\\ \times j_0(\kappa b_\nu)j_0(\kappa b_{\nu'})|V_\nu|^2|_{k=k_0}&.\end{aligned}$$

4. DIFFERENTIAL CROSS SECTION OF METHANE

The slow-neutron cross sections of methane and of a number of other gases have been measured by Alcock and Hurst.[4] Their approximate formula, Eq. (3.13), sufficed in most cases to reproduce the observed angular distributions. Pope[5] has applied this formula, suitably generalized to include the effects of molecular vibration, to the analysis of the scattering data on methane and carbon tetraflouride. He obtained good agreement with the experimental cross section for carbon tetrafluoride where the masses of the constituent atoms are large compared with the neutron mass, and interference scattering is dominant. However, the approximation did not adequately account for the observed decrease of scattering intensity at large angles in methane.

The moment of inertia of a methane molecule, expressed in terms of the carbon-hydrogen bond length b, is given by

$$I=(8/3)mb^2.$$

Therefore, the hydrogen atoms in methane each possess a rotational mass $\mathfrak{M}$, in the sense defined in Sec. 4 of I equal to $8m/3$. We have seen that inelastic corrections to the static or semiclassical approximation which are due to the excitation of rotational energy levels are of order of magnitude $(\mu/\mathfrak{M})$. Consequently, the effect of inelastic processes in the scattering of slow neutrons by methane may be considerable. The evaluation of this effect affords an illustration of the methods developed in the present paper, and the preceding one.

The experiment on methane was performed by Alcock and Hurst at room temperature with 0.0732-ev neutrons. Since the neutron energy was too low to excite vibrational transitions, only the elastic term (no change in vibrational quantum numbers) of the summation (3.5) is required. Pope has already determined the normal coordinates for methane and CF_4, and calculated the time-independent analog of (3.5). His result for the elastic term of the summation is

$$\exp\{-\kappa^2(\gamma_\nu+\gamma_{\nu'})-3\delta_\nu(\boldsymbol{\kappa}\cdot\mathfrak{b}_\nu)^2-3\delta_{\nu'}(\boldsymbol{\kappa}\cdot\mathfrak{b}_{\nu'})^2\}, \tag{4.1}$$

where $\mathfrak{b}_\nu$, $\mathfrak{b}_{\nu'}$ are unit vectors oriented along $\mathbf{b}_\nu$, $\mathbf{b}_{\nu'}$ respectively and the γ_ν, δ_ν are constants related to the force constants of the molecule. For methane, Pope found

$$4(k_0')^2\gamma_{\text{H}}=0.2863, \quad 4(k_0')^2\delta_{\text{H}}=0.1017,$$
$$4(k_0')^2\gamma_{\text{C}}=0.01323, \quad \delta_{\text{C}}=0.$$

The subscripts refer to carbon and hydrogen nuclei, and $k_0'=5.563\times 10^8$ cm^{-1} is the incident neutron wave number in the center-of-mass coordinate system. A value of 1.093×10^{-8} cm was used for the carbon-hydrogen bond length.

Bearing in mind the ordering of time-dependent and time-independent operators which must be preserved in the vibration expectation values, we infer from (4.1),

$$\begin{aligned}V_{\nu\nu'}&=\exp[-\kappa^2(\gamma_\nu+\gamma_{\nu'})]\exp[-3\delta_\nu(\boldsymbol{\kappa}\cdot\mathfrak{b}_\nu(t))^2]\\&\qquad\times\exp[-3\delta_{\nu'}(\boldsymbol{\kappa}\cdot\mathfrak{b}_{\nu'})^2]\\&=\exp[-\kappa^2(\gamma_\nu+\gamma_{\nu'})]\exp(iH_{\text{rot}}t)\exp[-3\delta_\nu(\boldsymbol{\kappa}\cdot\mathfrak{b}_\nu)^2]\\&\qquad\times\exp(-iH_{\text{rot}}t)\exp[-3\delta_{\nu'}(\boldsymbol{\kappa}\cdot\mathfrak{b}_{\nu'})^2].\end{aligned}$$

The coherent scattering amplitudes of carbon and hydrogen are[6]

$$A_{\text{H}}=-0.40\times 10^{-12}\text{ cm}, \quad A_{\text{C}}=0.64\times 10^{-12}\text{ cm}.$$

The bound-scattering cross sections for these nuclei are given by[7]

$$4\pi B_{\text{H}}{}^2=80.4\times 10^{-24}\text{ cm}, \quad 4\pi B_{\text{C}}{}^2=4\pi A_{\text{C}}{}^2.$$

Carbon, which is spinless, has no incoherent cross section. The outer scattering by methane is negligible.

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[5] N. K. Pope, Can. J. Phys. **30**, 597 (1952).

[6] C. G. Shull and E. O. Wollan, Phys. Rev. **81**, 527 (1951).

[7] R. K. Adair, Revs. Modern Phys. **22**, 249 (1950).

The differential cross section of the methane molecule becomes

$$\langle\sigma(\theta)\rangle_T=4B_{\mathrm{H}}^2F_1+12A_{\mathrm{H}}^2F_2+8A_{\mathrm{H}}A_{\mathrm{C}}F_3+A_{\mathrm{C}}^2F_4 \quad (4.2)$$

where

$$F_i=(2\pi)^{-1}(\mu/m)^2\int\int(k/k_0)Ze^{-it\epsilon'}\times\exp(-t^2T\kappa^2/2M)\langle\chi_i\rangle d\epsilon dt,$$

$$\langle\chi_1\rangle=\exp(-2\kappa^2\gamma_{\mathrm{H}})\langle\psi_{\mathrm{rot}}|\exp(iH_{\mathrm{rot}}t)\times\exp[i\boldsymbol{\kappa}\cdot\mathbf{b}-3\delta_{\mathrm{H}}(\boldsymbol{\kappa}\cdot\mathfrak{b})^2]\times\exp(-iH_{\mathrm{rot}}t)\exp[-i\boldsymbol{\kappa}\cdot\mathbf{b}-3\delta_{\mathrm{H}}(\boldsymbol{\kappa}\cdot\mathfrak{b})^2]|\psi_{\mathrm{rot}}\rangle_T,$$

and the expressions for $\langle\chi_i\rangle$, $i=2, 3, 4$, are similar. The terms $4B_{\mathrm{H}}^2F_1$ and $A_{\mathrm{C}}^2F_4$ of (4.2) describe the direct scattering by the four hydrogen nuclei and the single carbon nucleus, respectively, while the hydrogen-hydrogen and carbon-hydrogen interference scattering are accounted for by the remaining two terms.

The calculation of $\langle\sigma(\theta)\rangle_T$ based on the time expansion method will be briefly described. To compare theory and experiment, errors of less than $\frac{1}{2}\%$ in the numerical computations may be disregarded. The static approximation to F_1 takes the form

$$(F_1)_{\mathrm{stat}}=[(\mu/m)^2Z(\theta,1)\exp(-2\kappa^2\gamma_{\mathrm{H}})\times(4\pi)^{-1}\int\exp[-3\delta_{\mathrm{H}}(\boldsymbol{\kappa}\cdot\mathfrak{b})^2]d\Omega]_{k'=k_0'}.$$

To this, we must add the inelastic corrections due to terms proportional to powers of t in the expansion of $\exp(-t^2T\kappa^2/2M)\langle\chi_1\rangle$. The velocity ratio, as defined in section 5 of the preceding paper is given by $\alpha^2=0.022$. It follows that only the lowest order corrections for the thermal motion of the gas molecules, namely those proportional to α^2 need be retained. Accordingly, the factor $\exp(-t^2T\kappa^2/2M)$ whose expansion generates the α-dependent terms may be approximated by $(1-t^2T\kappa^2/2M)$. The magnitude of the methane cross section is altered less than 3% by the inclusion of the effect of random thermal motion.

To obtain the rotational corrections, we first expand in powers of the small quantity δ_{H}. Then the techniques employed in Sec. 4 of I in the evaluation of the direct scattering and interference expectation values to orders t^2 and t, respectively, are applicable. In the present case, some terms of order t^3 and t^4, resulting in rotational corrections to the cross section proportional to $(\mu/\mathfrak{M})^3$ and $(\mu/\mathfrak{M})^4$ must be included in the calculation, as well as terms to order t^2 which describe rotation-vibration interactions. The sole isotropic contribution to inelastic rotator scattering of appreciable size is given by the term of (I, 4.15) proportional to (l^2+l+1). The thermal average of the latter polynomial, evaluated at room temperature, is 61.6 so that the magnitude of this term is about 8% of the cross section in the forward direction. All other corrections contain one or more factors of $(1-\cos\theta)$ and contribute predominantly to the large angle scattering.

The terms F_2, F_3, and F_4 may be calculated with less accuracy than F_1 because the coherent scattering factors $12A_{\mathrm{H}}^2$, $8A_{\mathrm{H}}A_{\mathrm{C}}$, and A_{C}^2 are much smaller than $4B_{\mathrm{H}}^2$. Thus, the effect of thermal motion of the molecules may be disregarded in these particular terms. The inelastic rotational corrections to the interference scattering by two hydrogen nuclei, determined by (I, 4.19) are likewise negligible. No inelastic rotational corrections contribute to the direct scattering by the carbon nucleus, since this nucleus does not rotate. Such corrections are also absent from the carbon-hydrogen interference scattering, for the relevant expectation value $\langle\chi_3\rangle$ is composed of terms of the two forms

$$\exp[-\kappa^2(\gamma_{\mathrm{C}}+\gamma_{\mathrm{H}})]\times\langle\psi_{\mathrm{rot}}|\exp(iH_{\mathrm{rot}}t)\exp(i\boldsymbol{\kappa}\cdot\mathbf{b}-3\delta_{\mathrm{H}}(\boldsymbol{\kappa}\cdot\mathfrak{b})^2)\times\exp(-iH_{\mathrm{rot}}t)|\psi_{\mathrm{rot}}\rangle_T$$

and

$$\exp[-\kappa^2(\gamma_{\mathrm{C}}+\gamma_{\mathrm{H}})]\times\langle\psi_{\mathrm{rot}}|\exp(iH_{\mathrm{rot}}t)\exp(-iH_{\mathrm{rot}}t)\times\exp(-i\boldsymbol{\kappa}\cdot\mathbf{b}-3\delta_{\mathrm{H}}(\boldsymbol{\kappa}\cdot\mathfrak{b})^2)|\psi_{\mathrm{rot}}\rangle_T$$

in both of which the time dependence cancels. The physical basis of this simplification is easily understood as follows: a transfer of angular momentum to the molecule cannot occur if the neutron collides with the centrally located carbon nucleus. Hence any rotational inelasticity implies that the neutron has collided with a hydrogen nucleus. The rotationally inelastic scattering therefore contains no carbon-hydrogen interferences.

In Fig. 1, the calculated differential cross section[8] is compared with the data of Alcock and Hurst. The dashed line is the semiclassical approximation computed by Pope. The theoretical curves have been normalized to unity at $\theta=0°$. The normalization of the experimental points is that which gives closest fit at large angles, since in the forward direction the experimental errors are rather large. Our curve reproduces the data quite well at large angles. Although the agreement is less

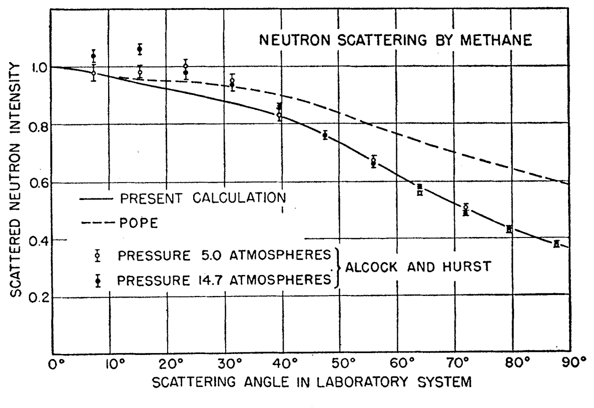

FIG. 1. Scattering of 0.0723-ev neutrons by methane at room temperature.

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[8] The total cross section for methane as a function of energy has been calculated by A. M. L. Messiah, Phys. Rev. 84, 204 (1951). We note that the oscillatory deviations of the experimental points from Messiah's curve are accounted for to within experimental error by the interference scattering which he neglected.

satisfactory at small angles, the experimental points for high and low gas pressures do not themselves agree here, suggesting the possibility of a slight experimental inaccuracy. Because of the relatively small values of the coherent scattering amplitudes of carbon and hydrogen, diffraction maxima and minima are not a prominent feature of the methane cross section. The most pronounced characteristic is the steady decrease of scattering intensity as a function of angle, due primarily to the effects of molecular vibration (included in Pope's curve), and to the large probability for inelastic scattering through interaction with the rotational degrees of freedom.

APPENDIX

It was shown in Sec. 2 that spin correlations which result from the requirements of nuclear statistics in the presence of like nuclei lead to additional interference terms in the scattering. The term which adds to (2.7), summed over distinct pairs ($\nu\neq\nu'$) of like nuclei has the form

$$\langle\chi\rangle_{\text{corr}}=\sum\nolimits'_{\nu,\nu'}C_\nu^2\langle\psi|f_{\nu\nu'}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})|\psi\rangle_T/S_\nu(S_\nu+1). \quad \text{(A.1)}$$

Let Ω denote the total spin of the like nuclei of a given type. Since the total spin is a symmetric function of the individual nuclear spins, it is a good quantum number for the energy eigenfunctions of the molecule. Let P_α, $\alpha=1, 2, \cdots, n$ be the elements in the group P of permutations of the nuclei.

We wish to characterize wave functions which transform either symmetrically or antisymmetrically under the permutations P_α. Let ξ_i, $i=1, 2, \cdots, m$, be molecular spin wave functions with quantum number Ω which transform like basis elements belonging to an irreducible representation $\Gamma(P_\alpha)$ of P. A total wave function of the desired symmetry may be formed by writing

$$\psi=\sum_{i=1}^{m}\phi_i{}^*\xi_i. \quad \text{(A.2)}$$

Here, the ϕ_i, $i=1, 2, \cdots, m$, are spatial wave functions of energy E belonging to the irreducible representation $\epsilon(P_\alpha)\Gamma(P_\alpha)$ where $\epsilon(P_\alpha)$ equals unity for Bose statistics or $(-1)^{P_\alpha}$, the parity of the permutation, for Fermi statistics. The proof that (A.2) has the correct symmetry follows from the unitary property of the representations. It is easy to show, using the theory of characters, that this mode of construction is unique.

The spin operators $\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'}$ appearing in (A.1) may be represented as matrices with elements $(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})_{ij}$ connecting the spin states ξ_i and ξ_j. In virtue of the scalar product form (A.2), matrices of this type may be replaced by their adjoints, operating on the $\{\phi_i\}$.

Let the permutation P_α be represented by a matrix $\bar{P}_\alpha$ in its action upon the spin states $\{\xi_i\}$. The same permutation applied to the spatial wave functions is represented by $\bar{P}_\alpha{}^\dagger$. Since $\sum'_{\nu,\nu'}f_{\nu\nu'}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})$ is completely symmetric as a *spatial* operator, spatial permutations represented by the matrices $\bar{P}_\alpha{}^\dagger$ may be freely applied to the wave functions in the expectation value. Averaging over the group, we find

$$\langle\chi\rangle_{\text{corr}}=\frac{1}{n}\sum_{\alpha=1}^{n}\left\langle\bar{P}_\alpha{}^\dagger\psi\left|\sum_{\nu,\nu'}{}'\frac{C_\nu^2}{S_\nu(S_\nu+1)}f_{\nu\nu'}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})\right|\bar{P}_\alpha{}^\dagger\psi\right\rangle_T$$

$$=\langle\psi|\sum\nolimits_{\nu,\nu'}{}'C_\nu^2/[S_\nu(S_\nu+1)]f_{\nu\nu'}K_{\nu\nu'}{}^\dagger|\psi\rangle_T,$$

where the spatial operator

$$K_{\nu\nu'}{}^\dagger=n^{-1}\sum\nolimits_\alpha(\bar{P}_\alpha{}^\dagger)^{-1}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})\bar{P}_\alpha{}^\dagger$$

can be immediately reinterpreted as a spin operator:

$$K_{\nu\nu'}=n^{-1}\sum\nolimits_\alpha\bar{P}_\alpha(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})\bar{P}_\alpha{}^{-1}.$$

But $K_{\nu\nu'}$, by its form, commutes with all permutation matrices $\bar{P}_\alpha$ of the irreducible representation Γ and is therefore a constant matrix. The dimension of Γ is m. Therefore, with $K_{\nu\nu'}=c\cdot 1$, we have

$$c=(mn)^{-1}\,\text{Trace}[\sum\nolimits_\alpha\bar{P}_\alpha(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})\bar{P}_\alpha{}^{-1}]$$
$$=m^{-1}\,\text{Trace}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'}).$$

We observe that c is independent of the indices ν, ν' for, introducing the permutation $\bar{P}_1$ which interchanges ν' and ν'' (where $\nu''\neq\nu$), we obtain

$$\text{Trace}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu''})=\text{Trace}[\bar{P}_1{}^{-1}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})\bar{P}_1]$$
$$=\text{Trace}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'}).$$

Therefore, if η is the number of identical nuclei, c is given by

$$c=[m\eta(\eta-1)]^{-1}\,\text{Trace}[\sum\nolimits_{\nu,\nu'}(\mathbf{S}_\nu\cdot\mathbf{S}_{\nu'})-\sum\nolimits_\nu S_\nu^2]$$
$$=\frac{\Omega(\Omega+1)-\eta S_\nu(S_\nu+1)}{\eta(\eta-1)}.$$

It follows that

$$\langle\chi\rangle_{\text{corr}}=\sum_{\nu,\nu'}{}'C_\nu^2\left\langle\psi\left|f_{\nu\nu'}\frac{\Omega(\Omega+1)-\eta S_\nu(S_\nu+1)}{S_\nu(S_\nu+1)\eta(\eta-1)}\right|\psi\right\rangle_T.$$

The quantity $\langle\chi\rangle_{\text{corr}}$ retains its form as a summation over distinct pairs (ν,ν') of like nuclei. Therefore, if $g_E(\Omega)$ denotes the statistical weight of the wave functions of energy E for which the total spin of the like nuclei in question is Ω the effect of spin coupling is described by adding

$$C_\nu^2\left\{\left[\sum_\Omega g_E(\Omega)\frac{\Omega(\Omega+1)-\eta S_\nu(S_\nu+1)}{S_\nu(S_\nu+1)\eta(\eta-1)}\right]\langle\phi|f_{\nu\nu'}|\phi\rangle\right\}_T \quad \text{(A.3)}$$

to the value of $\langle\chi_{\nu\nu'}\rangle$ as given in (2.7).

The deviations, at a given energy level, of the values of $g_E(\Omega)$ from the statistical weights appropriate to a molecule without symmetry restrictions are compensated by opposite deviations in adjacent energy levels.

Thus, symmetry requirements affect the scattering by a gas only at such low temperatures that very few energy levels are excited. Although it has been indicated heretofore that the correlation effect is small except at low temperatures, a general evaluation of it has not been previously given. The tables in reference 2 may be applied to the computation of (A.3) for various molecules. Typical results are that the cross section in the forward direction (where interference is greatest) is reduced 1% in gaseous methane at 45°K and 6% in gaseous ammonia at 200°K by the coupling of spins. Among the polyatomic molecules, ammonia probably exhibits the largest effect. These figures for the corrections decrease rapidly with increasing temperature.

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Properties of Em^{221}†

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A new isotope of the element emanation has been identified among the spallation products of thorium bombarded with 110-Mev protons. This isotope, Em^{221}, decays with a half-life of 25 minutes. The decay goes 80 percent by beta emission to Fr^{221} identified by its characteristic alpha particle groups and by the observance of the alpha particles of the decay products, At^{217} and Po^{213}. In 20 percent of its disintegrations Em^{221} emits an alpha particle of 6.0-Mev energy unresolved from the 6.0-Mev alpha particle of Fr^{221}, and gives rise to Po^{217}, a previously unreported isotope of polonium. Po^{217} has a half-life of less than 10 seconds and emits alpha particles of 6.54 ± 0.02 Mev.

An attempt was made to isolate Em^{221} from a sample of Th^{229} where it should be found as a result of slight alpha branching of Ra^{225}. The experiment was negative and a lower limit of 500 years was placed on the partial alpha half-life of Ra^{225}.

I. INTRODUCTION

During the course of a study of emanation isotopes of low mass number prepared by bombardment of thorium metal targets with 340-Mev protons, a technique was discovered enabling deposition of tracer rare gas activities on metal foils or wires. The radioactivity so deposited could be counted by any of the methods normally applicable to weight-free non-gaseous samples. This technique and its application to the study of emanation isotopes of mass numbers 212, 211, 210, 209, and 208 is discussed in detail in two recent papers.[1,2]

Briefly, the glow-discharge deposition method consists of introducing emanation gaseous activity, previously separated from a thorium target, into a glass tube equipped with two metal electrodes maintained at several hundred volts dc potential difference. Carrier gas is added to a pressure great enough to maintain a glow discharge. The emanation atoms are ionized and the positive ions are accelerated and collected on the cathode which may be a thin foil or a wire. The activity adheres strongly to the metal surface unless it is heated above room temperature.

As an extension of the previous study it was decided to search for the hitherto unobserved isotope, Em^{221}, among the spallation products of thorium and to attempt to determine its decay properties. This was successfully done and the information obtained on its decay scheme is summarized in Fig. 1. The details of the experiments will now be described.

II. EXPERIMENTAL DETAILS AND DISCUSSION

To facilitate the search for Em^{221}, the proton energy was dropped from 340 to 110 Mev so that the amount of emanation of mass number 212 and lower would be drastically reduced. At the higher energy the yield of emanation isotopes in the mass number region 212 and below is several hundred times greater than the yield of isotopes in the region of 221. Originally it was hoped that a good yield of Em^{221} could be obtained at a proton energy too low for the production of any Em^{212}. This proved to be impossible as will be discussed below, but at 110 Mev the yields were of comparable magnitude.

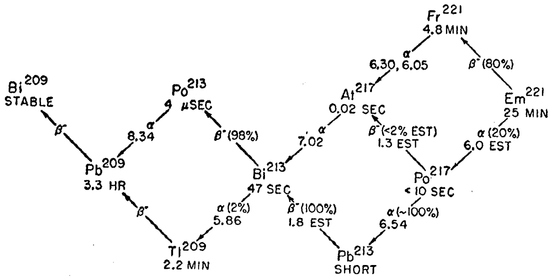

Fig. 1. Decay scheme of Em^{221}.

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† This research was performed under the auspices of the U. S. Atomic Energy Commission.

[1] F. F. Momyer, Jr., and E. K. Hyde, J. Inorg. Nuc. Chem. **1**, 274 (1955).

[2] Momyer, Asaro, and Hyde, J. Inorg. Nuc. Chem. **1**, 267 (1955).