Scattering of Slow Neutrons by H_2 and CH_4

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The mass tensor approximation, introduced by previous authors to calculate the scattering cross section of slow neutrons by molecules when the incident energy E_0 of the neutron is large compared with the average level spacing of the molecular rotations, is extended here to take into account the molecular vibrations. Calculations are made in the case of H_2 and CH_4 at room temperature for neutron energies below the threshold of excitation of higher vibrational states. The agreement with experiment is very satisfactory in the expected range of validity of this theory. The cross section for higher E_0 is also treated in the case of H_2, without taking into account, however, the thermal agitation of the scattering molecules: the asymptotic form of the total cross section for large values of E_0 is derived, and numerical calculations show that the asymptotic behavior is quickly reached.

THE ratio σ/σ_f of the neutron scattering cross section for nuclei in a molecule to that for free nuclei has been calculated by Sachs and Teller;[1] they treat the molecule as a rigid body and, using a classical approximation which introduces a mass tensor for the scattering nucleus, they obtain a simple expression for the scattering cross section of a molecule in a given orientation and with a given velocity of the scattering nucleus. The averaging of this expression over orientation and velocity is then carried out by an elaborate numerical procedure. They find that the numerical result for the energy dependence of σ/σ_f thus obtained can be well represented by the linear relationship:

$$\sigma/\sigma_f = a(kT/E_0)+b, \tag{1}$$

where T is the temperature of the scattering medium, k the Boltzmann constant, E_0 the energy of the incident neutron, a and b suitable numerical constants. It has been shown by Placzek[2] that this linear relationship may be obtained by performing this averaging analytically. He obtains the coefficients a and b as functions of the principal values of the mass tensor. His calculation leads to changes in the numerical values of a and b given by Sachs and Teller, especially in the a for the H_2 molecule.

This theory—referred to later as MTR theory—is based on the assumption that E_0 and kT are large compared with the mean level spacing of the initial rotational states of the molecule, and small compared with the vibrational quanta. Because of these two conditions, its region of applicability is rather limited. In the present paper, the mass tensor treatment, whose validity depends only on the first of the above restrictions, has been extended by taking into account the vibrations of the molecule (this will be referred to as MTV theory) and applied to the case of H_2 and CH_4 molecules.

The derivation of the basic formulas giving the scattering cross section by bound nuclei in the mass tensor approximation is given in Sec. I. In Sec. II is outlined the general analytical procedure which will be used to treat the particular cases in the next sections; the connection between MTV and MTR theory is shown. The scattering by H_2 molecules is treated in Sec. III. The elastic cross section is calculated, the doppler effect caused by thermal motion of the molecule being taken into account; the numerical results are given for a H_2 gas at room temperature. The general formulas for the inelastic cross sections are then given, neglecting the doppler effect. The asymptotic behavior of the total cross section at increasing E_0 is calculated, and a numerical study shows the rapidity of convergence toward this asymptotic form. In Sec. IV, the scattering cross section per proton in CH_4 molecules is treated. This study is restricted to the elastic cross section below the threshold of inelastic collision, and the same points are made as in the case of H_2. (The general formulas for the inelastic cross sections are given in Appendix.) Section V is devoted to the discussion of the validity of the theory and to the comparison with experimental results.

I

It is well known that, in calculating the transition probability for scattering of slow neutrons by bound nuclei, one can use a δ-type neutron-nucleus interaction and apply the Born approximation. The matrix element occurring in the perturbation calculation reduces then to $\langle F|\exp(i\mathbf{P}\cdot\mathbf{R}/\hbar)|I\rangle$, where $|I\rangle$ and $|F\rangle$ are the initial and final states of the molecule, $\mathbf{R}$ the position vector of the scattering nucleus,[3] $\mathbf{P}$ the momentum transferred to the molecule. The molecule is assumed to be initially in its ground state of vibration $|0\rangle$, its initial states of translation and rotation are denoted by $|T_I\rangle$ and $|R_I\rangle$; its final states of vibration, translation, and rotation by $|N\rangle$, $|T_F\rangle$, and $|R_F\rangle$. Call $\mathbf{R}_c$ the position vector of its center of mass; $\mathbf{r}$, the vector

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[1] R. G. Sachs and E. Teller, Phys. Rev. **60**, 18 (1941).
[2] G. Placzek (to be published).
[3] The limitations of the present theory for low values of the incident energy are such that the wavelengths of the neutrons considered here are always small compared with internuclear distances. Consequently, the interference terms between waves scattered by different nuclei are merely neglected.

joining this center of mass to the equilibrium position of the scattering nucleus; $\mathbf{w}$, the vibrational displacement of the latter:

$$\mathbf{R}=\mathbf{R}_c+\mathbf{r}+\mathbf{w}.$$

The matrix element considered above may be written:

$$\langle T_F|\exp(i\mathbf{P}\cdot\mathbf{R}_c/\hbar)|T_I\rangle\langle R_F|\exp(i\mathbf{P}\cdot\mathbf{r}/\hbar)W_N|R_I\rangle \quad (2)$$

with

$$W_N=\langle N|\exp(i\mathbf{P}\cdot\mathbf{w}/\hbar)|0\rangle.$$

W_N is a function of $\mathbf{P}$ and of the coordinates fixing the orientation of the molecule. If the temperature of the scattering medium is large compared with the rotational constant: $kT\gg B$, the initial rotational wave function can be replaced by a rotating wave packet with given orientation and angular velocity at the beginning of the collision. It is possible to give to W_N its value in this initial orientation and to take it out of the matrix element. If, in addition, the incident energy E_0 is large compared with the mean rotational level spacing, the wave function of the incident neutron can also be approximated by a wave packet of such a small extension that the time of collision is small compared with the period of rotation. Then, (1) reduces to the product of W_N and matrix elements involving the translational and rotational states whose unique role is to guarantee the conservation of linear and angular momentum respectively, i.e., to require that the energy transferred to the translational-rotational motion of the molecule has the same value E_{TR}^{cl} as in a classical collision. The latter quantity is defined as

$$E_{TR}^{cl}=\mathbf{V}_s\cdot\mathbf{P}+\tfrac{1}{2}\mathbf{P}M^{-1}\mathbf{P}, \quad (3)$$

where $\mathbf{V}_s$ is the initial velocity of the scattering nucleus and M the mass tensor introduced by Sachs and Teller.

Thus, apart from a constant factor, the transition probability for the neutron to be scattered into a given solid angle $(\Omega_1,\ \Omega_1+d\Omega_1)$ with the molecule suffering the vibrational transition $0\rightarrow N$, is equal to the product of $|W_N|^2$ and the density of final states, which can be written

$$d\Omega_1\int_0^\infty |W_N|^2\delta[E_1+E_N+E_{TR}^{cl}-E_0]P_1^2dP_1,$$

where E_1 and $\mathbf{P}_1$ are the energy and momentum, respectively, of the scattered neutron and E_N the energy transferred to the vibrations. Integrating over solid angle and dividing by the neutron initial velocity (P_0/m), one obtains the scattering cross section with transition $0\rightarrow N$:

$$\sigma_N=(c/P_0)\int |W_N|^2\delta[E_1+E_N+E_{TR}^{cl}-E_0]d_3\mathbf{P}_1. \quad (4)$$

The constant of proportionality c is determined by the condition that σ_N must reduce to the cross section σ_∞ for scattering by the rigidly fixed nucleus when $|W_N|^2=1$, $E_{TR}^{cl}=E_N=0$. This gives: $c=\sigma_\infty/4\pi m$. It is convenient to introduce

$$\mu=m^{-1}M,\quad \nu=(1+\mu^{-1})^{-1},$$

mass tensor and reduced mass tensor, respectively, expressed in neutron mass units. Then, substituting Eq. (3) in Eq. (4) and taking the transferred momentum $\mathbf{P}$ as new integration variable,

$$\sigma_N=(\sigma_\infty/2\pi P_0)\int B_N(\mathbf{P})\delta[\mathbf{P}\nu^{-1}\mathbf{P}+2m\mathbf{V}_s\cdot\mathbf{P}+2mE_N-2\mathbf{P}_0\cdot\mathbf{P}]d_3\mathbf{P} \quad (5)$$

with

$$B_N(\mathbf{P})\equiv|W_N|^2=|\langle N|\exp(i\mathbf{P}\cdot\mathbf{w}/\hbar)|0\rangle|^2. \quad (5')$$

Equations (5) and (5') constitute the basic formulas of the MTV theory. It reduces to the MTR theory if one writes $B_0=1$ and $B_N=0$ $(N\neq 0)$, which merely expresses the neglect of vibrational motion.

In case of degeneracy in the final state of vibration, it is convenient to deal with the sum of the scattering cross sections for transition to each one of the degenerate states. In such cases, σ_N will denote this sum and B_N the sum of the absolute squares of the matrix elements corresponding to a transition to each degenerate state. With these conditions, Eq. (5) will still hold.

The cross section σ_N has to be averaged over all initial states of the scatterer, namely, over random orientation of the molecule, then over the initial values of $\mathbf{V}_s$, whose distribution depends on the temperature T of the scattering medium according to Boltzmann's statistics. Those two successive averages will be denoted by $\bar{\sigma}_N$ and $\langle\bar{\sigma}_N\rangle_T$. Then, the cross section σ for the whole scattering process is

$$\sigma=\textstyle\sum_N\langle\bar{\sigma}_N\rangle_T. \quad (6)$$

σ is the quantity which is experimentally measurable.

All σ_N, except the elastic one σ_0, vanish for sufficiently slow neutrons. It is clear from Eq. (5) that the threshold depends on the initial state of the molecule. One also gets a threshold for $\bar{\sigma}_N$ and for[4] $\langle\bar{\sigma}_N\rangle_T$, so that the sum Eq. (6) is in fact limited. Letting ν' be the largest component of ν, the threshold for $\langle\bar{\sigma}_N\rangle_T$ is $T_N=E_N/\nu'$.

II

The calculation of B_N implies the knowledge of the vibrational potential V in the molecule. Introducing a set of normal coordinates q_l and neglecting the effect of anharmonicity, one gets $V=\sum_l\omega_l^2q_l^2$, where the ω_l are the normal frequencies, and the vibrational wave functions are products of one-dimensional harmonic oscillators, $|N\rangle=\prod_l|n_l\rangle$. Expressing then $\mathbf{w}$ as a linear combination of the q_l, $\mathbf{w}=\sum_l q_l\mathbf{w}_l$, and setting $k_l=\mathbf{P}\cdot\mathbf{w}_l/\hbar$, B_N takes the form of a product—or a sum of products in the case of degeneracy—of absolute

[4] The threshold for $\langle\bar{\sigma}_N\rangle_T$ is defined here by neglecting $\mathbf{V}_s$. It is, in fact, smeared by the thermal effect; but, at the temperatures considered in the following, the values of $\langle\bar{\sigma}_N\rangle_T$ below its "threshold" are negligibly small.

squares of matrix elements of the type $\langle n_l|\exp(ik_lq_l)|0\rangle$. It can be expressed analytically with help of the formula[5]

$$|\langle n|\exp(ikq)|0\rangle|^2=(n!)^{-1}(\hbar k^2/2\omega)^n\exp(-\hbar k^2/2\omega); \quad (7)$$

in particular

$$|\langle 0|\exp(ikq)|0\rangle|^2=\exp(-\hbar k^2/2\omega). \quad (8)$$

The calculation of the multiple integrals involved in $\langle\bar\sigma_N\rangle_T$ and even $\bar\sigma_N$ is in general exceedingly complicated. However, in the cases of H_2 and CH_4, the expressions are greatly simplified, since the axis joining the scattering nucleus to the center of mass of the molecule is an axis of cylindrical symmetry for the scattering problem.

Call P, θ, φ the polar coordinates of $\mathbf{P}$ with the axis of symmetry as polar axis; M_1, μ_1, ν_1 the principal components of M, μ, ν along this axis; M_2, μ_2, ν_2 the other principal components ($M_1>M_2$, $\mu_1>\mu_2$, $\nu_1>\nu_2$); and introduce the quantities u, α, $v_{||}$, s such that

$$u=\cos\theta, \qquad \alpha=\cos(\mathbf{P}_0,\mathbf{P}),$$
$$v_{||}=\mathbf{V}_s\cdot\mathbf{P}/P, \quad 2/s=\nu_2^{-1}-(\nu_2^{-1}-\nu_1^{-1})u^2.$$

From Eq. (5)

$$\bar\sigma_N=(\sigma_\infty/2\pi P_0)\int B_N d_3\mathbf{P}[\tfrac{1}{2}\int_{-1}^{+1}\delta(\mathbf{P}\nu^{-1}\mathbf{P}+2m\mathbf{V}_s\cdot\mathbf{P}+2mE_N-2P_0P\alpha)d\alpha]. \quad (9)$$

Performing the α-integration and introducing polar coordinates, one gets

$$\bar\sigma_N=(\sigma_\infty/2\pi P_0^2)\int_{F_-<0<F_+} PB_N du d\varphi dP, \quad (10)$$

$$F_\pm=P^2+s(mv_{||}\pm P_0)P+smE_N. \quad (10')$$

(Note that the volume of integration is limited by an algebraic surface of order 4.) The thermal average leads to relatively simple analytic expressions for the elastic cross section only. This will be shown now and the final expression will exhibit the link with the MTR theory. B_0, as will be seen later, takes the form,

$$B_0=\exp[-t(2\hbar\omega)^{-1}(P^2/2m)], \quad (11)$$

where $t=A_1u^2+A_2$ (A_1, A_2 are nondimensional constants depending on the elastic forces in the molecule) and ω is chosen, for instance, as the lowest vibrational frequency in the molecule. It is convenient to introduce also $y=E_0/2\hbar\omega$. Then

$$\bar\sigma_0=(\sigma_\infty/8\pi P_0^2)\int_{-1}^{+1}du\int_0^{2\pi}d\varphi\int_{P'}^{P''}P\times\exp[-t(2\hbar\omega)^{-1}(P^2/2m)]dP. \quad (12)$$

[5] N. Arley, Kgl. Danske Videnskab. Selskab **16**, 1 (1938).

The range (P', P'') takes the following values:

(a) if $-P_0<mv_{||}<P_0$,
$$(P', P'')=(0, s(P_0-mv_{||})), \quad (13a)$$

(b) if $mv_{||}<-P_0$,
$$(P', P'')=(s(-P_0-mv_{||}), s(P_0-mv_{||})), \quad (13b)$$

(c) if $mv_{||}>P_0$, $(P', P'')=(0, 0)$. (13c)

As long as the initial velocity of the neutron is greater than the scatterer one V_s, case (a) necessarily holds and, from Eqs. (12) and (13a),

$$\bar\sigma_0=(\sigma_\infty/4y)\int_0^1 du\int_0^{2\pi}(d\varphi/2\pi)t^{-1}\times[1-\exp(-s^2t(P_0-mv_{||})^2/4m\hbar\omega)]. \quad (14)$$

If one performs the thermal average by assuming that case (a) holds for all values of $v_{||}$, one obtains a result $\langle\bar\sigma_0\rangle_T$ greater than the correct one by an amount which becomes vanishingly small with increasing E_0. This approximation will be discussed more precisely in Sec. V. Doing it, expanding the integrand in Eq. (14) in powers of $v_{||}$, and substituting in the successive powers of $v_{||}$ their thermal averages (which no longer depend on φ), one gets

$$\langle\bar\sigma_0\rangle_T=(\sigma_\infty/4y)\Big\{\int_0^1 t^{-1}[1-\exp(-s^2ty)]du + \int_0^1[\langle mv_{||}^2\rangle_T/2\hbar\omega][\tfrac{1}{2}-s^2ty]s^2\exp(-s^2ty)du + \int_0^1[\langle m^2v_{||}^4\rangle_T/(2\hbar\omega)^2][-\tfrac{1}{8}+\tfrac{1}{2}s^2ty-\tfrac{1}{6}s^4t^2y^2]s^4t\exp(-s^2ty)du+\cdots\Big\}. \quad (15)$$

If one assumes a maxwell distribution of velocities—which is consistent with our condition $kT\gg B$—the thermal averages $\langle mv_{||}^2\rangle_T$, $\langle m^2v_{||}^4\rangle_T$, $\cdots$, are obtained[1] by expressing $v_{||}$ in terms of the components of the vector $\varrho=M^{\frac{1}{2}}\mathbf{V}_s$, and then averaging in ϱ-space with the Boltzmann weight $\exp(-\rho^2/kT)$. This leads to

$$\langle mv_{||}^2\rangle_T=(2-s)s^{-1}kT, \qquad \langle m^2v_{||}^4\rangle_T=3(2-s)^2s^{-2}(kT)^2. \quad (16)$$

Introducing $\Theta=kT/2\hbar\omega$, and substituting Eq. (16) in Eq. (15),

$$\langle\bar\sigma_0\rangle_T=(\sigma_\infty/4y)\Big\{\int_0^1 t^{-1}[1-\exp(-s^2ty)]du + \Theta\int_0^1 s(2-s)(\tfrac{1}{2}-s^2ty)\exp(-s^2ty)du + \Theta^2\int_0^1 ts^2(2-s)^2(-\tfrac{3}{8}+\tfrac{3}{2}s^2ty-\tfrac{1}{2}s^4t^2y^2)\exp(-s^2ty)du+\cdots\Big\}. \quad (17)$$

For small energies, the most convenient way to calculate $\langle\bar{\sigma}_0\rangle_T$ is to express the exponentials in the integrands of Eq. (17) in power series; one obtains $\langle\bar{\sigma}_0\rangle_T$ as a double expansion in ascending powers of y and Θ whose coefficients are analytically integrable.

$$\langle\bar{\sigma}_0\rangle_T/(\sigma_\infty/4)=\left[\int_0^1 s^2du-y/2\int_0^1 s^4tdu+\cdots\right]$$
$$+(\Theta/y)\left[\frac{1}{2}\int_0^1 s(2-s)du-\frac{3}{2}y\int_0^1 s^3t(2-s)du\right.$$
$$\left.+\frac{5}{4}y^2\int_0^1 s^5t^2(2-s)du-\cdots\right]$$
$$+\frac{\Theta^2}{y}\left[-\frac{3}{8}\int_0^1 s^2t(2-s)^2du+\frac{15}{8}y\int_0^1 s^4t^2(2-s)^2du\right.$$
$$\left.-\frac{35}{16}y^2\int_0^1 s^6t^3(2-s)^2du+\cdots\right]+\cdots. \quad (18)$$

The results of MTR theory are easily deduced from Eq. (18). The neglect of vibration consists mathematically in taking the limit of this expression for $\hbar\omega$ infinite, or expressed otherwise, in letting Θ and y be together infinitely small but keeping the ratio $\Theta/y=kT/E_0$ constant, i.e.,

$$\langle\bar{\sigma}_0\rangle_T/(\tfrac{1}{4}\sigma_\infty)=\int_0^1 s^2du+\tfrac{1}{2}(kT/E_0)\int_0^1 s(2-s)du, \quad (19)$$

which is exactly the expression (1) with coefficients in agreement with the analytic expressions given by Placzek. On the other hand, it is clear that the asymptotic form of $\langle\bar{\sigma}_0\rangle_T$ for low energies may also be written in the form,

$$\langle\bar{\sigma}_0\rangle_T/(\tfrac{1}{4}\sigma_\infty)\sim a'(kT/E_0)+b' \quad (20)$$

by dropping in Eq. (18) all the terms involving positive powers of y. a' and b' are expansions in power of Θ whose constant terms are the coefficients of MTR theory written explicitly in Eq. (19). The formulas (19) and (20), in the case of H_2 and CH_4 at room temperature are compared in Table I.

III

In the case of the scattering by H_2 molecules, the axis of the H_2 molecule constitutes the axis of cylindrical symmetry of the scattering problem. The mass tensors μ, ν have the following components: $\mu_1=2$, $\mu_2=1$, $\nu_1=\frac{2}{3}$, $\nu_2=\frac{1}{2}$, and $1/s=1-\frac{1}{4}u^2$. The rotational constant[6] $B=0.007356$ volt. The energy of vibrational quantum $\hbar\omega=0.54617$ volt. A vibrational state is designated by the number n of its vibrational quanta. The thresholds for the successive inelastic collisions, $T_n=\frac{3}{2}n\hbar\omega$. If q is the normal coordinate (corresponding to the hamiltonian for the vibrational motion $H_{\text{vib}}=\frac{1}{2}(p^2+\omega^2q^2)$), it is connected with the displacement w of the H nucleus from its equilibrium position by $w=(2m)^{-\frac{1}{2}}q$. Then, from the definition (5′) and with help of Eq. (8):

$$B_n=(n!)^{-1}(u^2P^2/4m\hbar\omega)^n\exp[-u^2P^2/4m\hbar\omega], \quad (21)$$
$$B_0=\exp[-u^2P^2/4m\hbar\omega]. \quad (22)$$

Here $t=u^2$. It is convenient to compare the calculated cross sections with the scattering cross section σ_f by free protons, rather than with σ_∞; $\sigma_f=\frac{1}{4}\sigma_\infty$.

A complete treatment of the elastic cross section is now given; the latter is equal to the total cross section as long as $E_0<0.819$ volt ($y<0.75$). The numerical calculations have been made at room temperature: $kT=0.0258$ volt, where the distribution of rotations is not yet maxwellian; the mean rotational energy $\langle E_R\rangle_T=0.0235$ volt. At such a temperature, about $\frac{2}{3}$ of the molecules have quantum number $J=1$. One may question the validity of considering those rotating wave packets in the initial state, as was done in Sec. I. On the other hand, one sees from formulas (15) or (17) that the theory gives $\langle\bar{\sigma}_0\rangle_T$ as a sum of terms: the first term is independent of the temperature (it may be obtained by writing $V_s=0$ in all the preceding steps); the following terms may be understood as corrections for the doppler effect due to thermal agitation; those last terms will turn out to be very small for the values of the incident energy y where the theory is valid—the third term in Eq. (15) is even completely negligible.[7] Consequently, $\langle\bar{\sigma}_0\rangle_T$ has been calculated from formula (15), retaining only the two first terms, and expressing the second term itself as a sum of two contributions: one given by assuming a maxwell distribution of rotation, the second being the correction due to deviation from this distribution, as deduced from

$$\langle mv_{||}^2\rangle_T=s^{-1}(2-s)kT-\tfrac{1}{2}(1-u^2)(kT-\langle E_R\rangle_T). \quad (23)$$

Writing merely σ_0 for $\langle\bar{\sigma}_0\rangle_T$, this gives

$$\sigma_0/\sigma_f=Q+R(kT/E_0)-R'[(kT-\langle E_R\rangle_T)/E_0]+\cdots \quad (24)$$

TABLE I. Comparison between the expressions of σ/σ_f given by MTR theory and MTV theory (at low incident energy) for scattering per proton in H_2 and CH_4 at room temperature ($kT=0.0258$ volts). A maxwellian distribution of molecular rotations is assumed.

	MTR expression	Asymptotic MTV expression
H_2	$0.491(kT/E_0)+1.216$	$0.488(kT/E_0)+1.200$
CH_4	$0.3468(kT/E_0)+2.3778$	$0.3314(kT/E_0)+2.2058$

[6] G. Herzberg, *Molecular Spectra and Molecular Structures* (Prentice-Hall, Inc., New York, 1939), Vol. I.

[7] It may be shown to be less than $0.003(kT/E_0)\sigma_f$. It has been taken into account, however, in the comparison between MTV and MTR theory.

TABLE II. Numerical values of the coefficients of the first powers of y, in the Taylor expansion of Q, R, R' [see Eqs. (24a), (24b), and (24c)].

	Q	R	R'
y^0	1.2159728	0.490626	0.186633
y^1	−0.3379821	-0.671468_6	−0.164495
y^2	0.1174249	0.535065	0.094721
y^3	−0.03685118	−0.299159	−0.041201
y^4	0.0101218	0.12829_5	0.014424
y^5	-0.0024428_4	-0.04453_1	-0.00422_5
y^6	0.0005227_1	0.01299_6	
y^7	-0.0001005_3	-0.00325_4	
y^8	0.0000175		
y^9	-0.0000027_8		
y^{10}	0.0000004_1		

TABLE III. Energy dependence of the scattering cross section per protons in H_2 in the region of pure elastic scattering when $T=300°K$. Comparison with experiment.

E_0	y	Q	$R(kT/E_0)$	$R'[kT-\langle E_R\rangle_T/E_0]$	Theoretical value σ/σ_f	Experimental value (σ/σ_f)exp
0.0256	0.0234	1.20812	0.47888	0.01656	1.67	1.725±0.01
0.0347	0.0317	1.20536	0.34935	0.01213	1.54	1.600±0.01
0.0530	0.0485	1.19964	0.22360	0.00783	1.42	1.435±0.005
0.0830	0.0760	1.19096	0.13759	0.00488	1.32	1.319±0.005
0.146	0.1337	1.17280	0.07238	0.00264	1.24	1.232±0.005
0.187	0.1710	1.16142	0.05380	0.00200	1.21	1.194±0.005
0.238	0.2180	1.14749	0.03972	0.00151	1.18	1.175±0.005
0.325	0.2975	1.12487	0.02621	0.00104	1.150	1.141±0.005
0.387	0.3543	1.10947	0.02044	0.00083	1.129	1.109±0.005
0.470	0.4303	1.08966	0.01523	0.00064	1.104	1.093±0.005
0.582	0.5328	1.06438	0.01073	0.00048	1.074	1.071±0.005
0.737	0.6747	1.03188	0.00695	0.00034	1.038	1.034±0.005
0.819	0.7500	1.01569	0.00611	0.00020	1.022	

with

$$Q=y^{-1}\int_0^1[1-\exp(-u^2s^2y)](du/u^2), \tag{24a}$$

$$R=\int_0^1 s(2-s)(\tfrac{1}{2}-u^2s^2y)\exp(-u^2s^2y)du, \tag{24b}$$

$$R'=\int_0^1 (s^2/2)(1-u^2)(\tfrac{1}{2}-u^2s^2y)\exp(-u^2s^2y)du. \tag{24c}$$

Then it has been merely considered that the correction term for deviation from Maxwell distribution $R'[(kT-\langle E_R\rangle_T)/E_0]$ gives the limit of accuracy of the theory. It is given as such in Table III, and the numerical values of σ_0/σ_f are indicated accordingly. Q, R, R' have been calculated by expanding the integrand in power of y and by analytic integrations of the coefficients; the numerical values of the coefficients of the lowest powers are given in Table II. Table III gives,

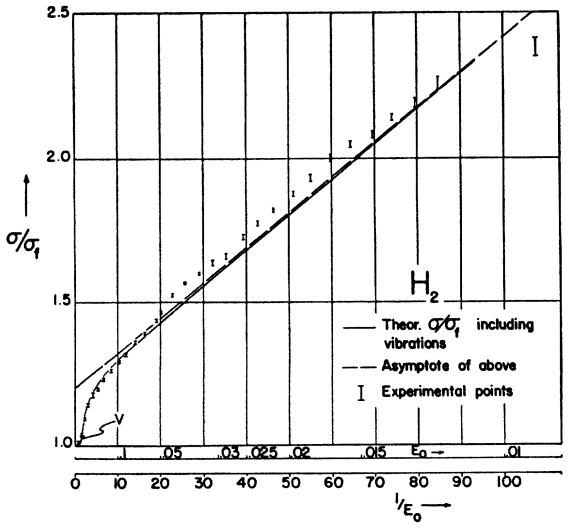

FIG. 1. Energy dependence of the slow neutron scattering cross section per proton by H_2 molecules at room temperature ($T=300°K$): theoretical curve given by MTV theory and experimental points. Arrow V indicates the energy threshold for inelastic collision.

at some specific value of E_0, the values of σ_0/σ_f at room temperature and the contributions of each term in the sum (24). In Fig. 1 is shown the curve σ_0/σ_f at room temperature in a $1/E_0$ diagram with its asymptote for low energies.

One now turns to the inelastic cross sections. The expressions are much more complicated than in the elastic case and it is no longer possible to take so simply the effect of thermal agitation into account. The inelastic cross sections are treated here, neglecting the thermal agitation. It follows from Eqs. (10) and (10′) where one makes $v_{||}=0$, $E_n=n\hbar\omega$ and substitutes for B_n expression (21). Again, the φ and P integration may be carried out analytically and one is left with a single integral in u.

If one sets

$$f_i(v)=\int_0^v (x^ie^{-x}/i!)dx=1-e^{-v}[1+(v/1!)+\cdots+(v^i/i!)] \qquad (i=0,\,1,\,2,\,\cdots),$$

$$v_\pm=\tfrac{1}{4}u^2s[(sy)^{\frac{1}{2}}\pm(sy-n)^{\frac{1}{2}}]^2,$$

$$u_0=\begin{cases} 0 & \text{if } y>n \\ 2[1-(y/n)]^{\frac{1}{2}} & \text{if } n>y>\tfrac{3}{4}n, \\ 1 & \text{if } \tfrac{3}{4}>y \end{cases}$$

the final result is

$$\bar\sigma_n/\sigma_f=(1/y)\int_{u_0}^1[f_n(v_+)-f_n(v_-)](du/u^2). \tag{25}$$

The right-hand side can be expressed in terms of elementary functions and the error integral in the special case: $y=n$ (see Appendix). For all other values of the energy, one has to perform the integration numerically. Then the total cross section is given by

$$\sigma=\textstyle\sum_n \bar\sigma_n. \tag{26}$$

It is interesting to study the asymptotic behavior of the total cross section σ, and to see if it agrees, in this special case, with the general expression of the scattering cross section by a bound nucleus, which is obtained by assuming that the nucleus scatters as free and by

taking the doppler effect into account,[2] i.e.,

$$\sigma/\sigma_f=1+[\langle K\rangle/3\mu_s E_0], \tag{27}$$

where $\langle K\rangle$ is the average kinetic energy of the nucleus and μ_s = mass of scattering nucleus/mass of the neutron. In the case of H_2, neglecting thermal agitation, the latter formula reduces to

$$\sigma/\sigma_f=1+(1/48y). \tag{28}$$

To solve this asymptotic problem, it is convenient to put Eqs. (25) and (26) in a slightly different form. One introduces $\sigma_n(u)$ such that

$$\frac{\sigma_n(u)}{\sigma_f}=\begin{cases}(1/u^2y)[f_n(v_+)-f_n(v_-)] & \text{if } sy>n\\ 0 & \text{if } sy<n\end{cases}$$

$(\sigma_n(u)du$ is the scattering cross section with energy transfer of n quanta and momentum transfer directed in the cone between u and $u+du$), and also $\sigma(u)$:

$$\frac{\sigma(u)}{\sigma_f}=\sum_{n=0}^{[sy]}\left(\frac{\sigma_n(u)}{\sigma_f}\right). \tag{29}$$

The symbol $[sy]$ means: the greatest integer contained in sy. Then Eqs. (25) and (26) may be written

$$\bar\sigma_n/\sigma_f=\int_0^1(\sigma_n(u)/\sigma_f)du,\quad \sigma/\sigma_f=\int_0^1(\sigma(u)/\sigma_f)du. \tag{30}$$

Writing the right side of Eq. (29) explicitly, one gets

$$\frac{\sigma(u)}{\sigma_f}=\sum_{n=0}^{[sy]}\left(\frac{1}{u^2y}\right)\int_{v_-}^{v_+}\left(\frac{v^ne^{-v}}{n!}\right)dv$$

with

$$v_\pm=\tfrac{1}{4}u^2s[(sy)^{\frac{1}{2}}\pm(sy-n)^{\frac{1}{2}}]^2.$$

Setting

$$\mu'=4/u^2s=(4/u^2)-1,$$
$$\epsilon'\equiv(E'/\hbar\omega')=sy,$$
$$x_\pm=\mu'^{-1}[\epsilon'^{\frac{1}{2}}\pm(\epsilon'-n)^{\frac{1}{2}}]^2,$$

the last expression may be written

$$\sigma(u)/\sigma_f=[(\mu'+1)^2/4\mu']\epsilon'^{-1}\sum_{n=0}^{[\epsilon']}\int_{x_-}^{x^+}(x^ne^{-x}/n!)dx.$$

In this form, $\sigma(u)$ is easily seen to be equal to the scattering cross section σ' of a neutron of energy E' by a nucleus of mass $M'=m\mu'$ oscillating harmonically and isotropically in its ground state around an infinitely heavy center with frequency ω'. The asymptotic behavior of σ' has been derived explicitly. If $\mu'=1$, Eq. (27) gives[2] only the average of the asymptotic cross section over an energy region of width $\hbar\omega'$ around which the cross section fluctuates with period $\hbar\omega'$. If $\mu'\neq1$, however, Eq. (27) is valid without averaging.[8]

$$\sigma'/\sigma_f\sim1+(1/4\mu'\epsilon').$$

[8] A. Messiah, J. phys. et radium **12**, 670 (1951).

In the present problem, μ' ranges from 3 to ∞ and one deals only with the latter case. Accordingly

$$\sigma(u)/\sigma_f\sim1+(u^2/16y).$$

Integrating over u

$$\sigma/\sigma_f\sim1+(1/48y),$$

which agrees with Eq. (28).

A numerical study of the total cross section above the threshold for inelastic collision has been made, up to the energy: $y=3$. The result is shown in Fig. 2, where σ/σ_f and also $\bar\sigma_0/\sigma_f$, $\bar\sigma_1/\sigma_f$, $\cdots$, are plotted against $(1/y)$. In order to see the rapidity of convergence

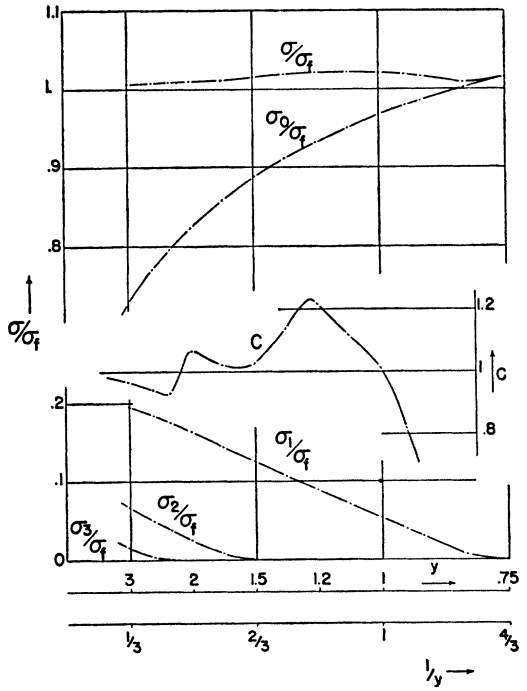

FIG. 2. Energy dependence of the slow neutron scattering cross section per proton by H_2 molecules above the threshold for inelastic collisions without thermal effect: elastic σ_0/σ_f, inelastic σ_1/σ_f, σ_2/σ_f, $\cdots$, and total σ/σ_f cross sections. The plot: $c=48y(\sigma/\sigma_f-1)$ *vs* $1/y$, emphasizes the quick fluctuating trend of σ/σ_f toward its asymptotic form $\sigma/\sigma_f\sim1+(1/48y)$.

of σ towards its asymptotic form (28), the plot of c against $(1/y)$, where c is defined by

$$\sigma/\sigma_f=1+(c/48y),$$

has been drawn on the same diagram. One sees that σ/σ_f exhibits a fluctuating trend towards its asymptotic expression; such fluctuations vanish very quickly as y increases and the asymptotic formula is practically reached when $y=3$ $(c=0.97)$.

IV

In a CH_4 molecule, the carbon nucleus is not involved in the molecular rotation; therefore no mass tensor treatment is needed to derive the scattering cross section

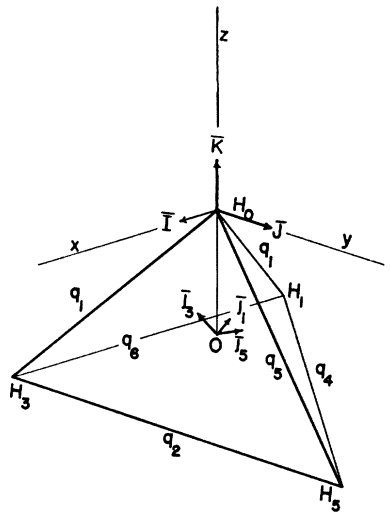

FIG. 3. Coordinate system for the methane molecule.

by the C nucleus. This section is concerned only with the scattering cross section per proton in CH_4. The total scattering cross section per proton will be called σ and the notations of Sec. III will be used all along.

The axis joining the center of mass of the molecule to the scattering proton is then the axis of cylindrical symmetry of the scattering problem.

The mass tensors μ, ν have the following components: $\mu_1=16$, $\mu_2=16/7$, $\nu_1=16/17$, $\nu_2=16/23$, and $1/s=(23-6u^2)/32$. The rotational constant:[9] $B=0.0006$ volt. The distribution of rotations is a maxwell one at room temperature.

There are 4 normal vibrations:[9] a nondegenerate one involving the H's only (along the C−H bond): $\hbar\omega_1=0.3756_3$ volt; a twofold degenerate one involving the H's only ($\perp$ to the C−H bond): $\hbar\omega_2=0.1723_6$ volt; and two threefold degenerate: $\hbar\omega_3=0.1683_2$ volt, $\hbar\omega_4=0.39139$ volt. Those last two belong to the same 3-dimensional irreducible representation of the group of symmetries of the CH_4 molecule. As ω_3 is the lowest frequency, according to our conventions,

$$y=E_0/2\hbar\omega_3, \quad \Theta=kT/2\hbar\omega_3.$$

A vibrational level will be indicated by its 4 quantum numbers listed in the order given above (the ground level will be more simply referred to by the index 0). The thresholds for inelastic collisions are

$$T_{n_1n_2n_3n_4}=(1/\nu_1)(n_1\hbar\omega_1+n_2\hbar\omega_2+n_3\hbar\omega_3+n_4\hbar\omega_4).$$

The thresholds (in volts) for the first inelastic cross sections are indicated below:

T_{0010}	T_{0100}	T_{0020}	T_{0110}	T_{0200}	T_{1000}	T_{0001}
0.1788	0.1831	0.3577	0.3620	0.3662	0.3991	0.4158

One expresses now the displacement $\mathbf{w}$ of the scattering proton as a linear combination of the normal coordinates, in view of the calculation of $B_{n_1n_2n_3n_4}$. In the derivation of $\mathbf{w}$, some symbols which were introduced before will be used with a different meaning; the new meaning will be given in time and will be used for this specific derivation only, so that any confusion may be easily avoided.

Call H_0 the proton nucleus whose displacement will be considered, H_1, H_3, and H_5, the three others, 0 the center of mass of the molecules; $\mathbf{I}$, $\mathbf{J}$, $\mathbf{K}$ a right-handed triorthogonal basis of fixed unit vectors, such that $\mathbf{K}$ be along the rest position of OH_0 and $\mathbf{I}$ in the rest position of the plane H_1OH_0—the sense of those vectors is indicated on Fig. 3. Introduce also (according to Dennison's notation) q_1 and q_2, q_3 and q_4, q_5 and q_6, the variations in magnitude of the three pairs of opposite edges; ξ, η, ζ, the coordinates of the displacement of the carbon nucleus in the reference frame ($\mathbf{j}_1$, $\mathbf{j}_3$, $\mathbf{j}_5$) of unit vectors respectively orthogonal to each couple of opposite edge.

T and V, the kinetic and potential energy of vibration of the CH_4 molecule, are given by

$$2T=3m(\dot\xi^2+\dot\eta^2+\dot\zeta^2)+\tfrac{5}{8}m\Sigma_i\,\dot q_i^2-\tfrac{1}{4}m\Sigma_{\text{n.op.}}\,\dot q_i\dot q_j+\tfrac{1}{4}m\Sigma_{\text{op.}}\,\dot q_i\dot q_j, \quad (31a)$$

$$2V=a(\xi^2+\eta^2+\zeta^2)+b\Sigma_i\,q_i^2+2c\Sigma_{\text{n.op.}}\,q_iq_j+2d\Sigma_{\text{op.}}\,q_iq_j +2e[\xi(q_1-q_2)+\eta(q_3-q_4)+\zeta(q_5-q_6)], \quad (31b)$$

where $\Sigma_{\text{n.op.}}$ means the sum of all cross products of non-opposite q_i, q_j (12 terms), and $\Sigma_{\text{op.}}$ means the sum of all cross products of opposite q_i, q_j (3 terms). $a=8.5881\times10^5$ dyne/cm, $b=1.6468\times10^5$ d/cm, $c=0.3584\times10^5$ d/cm, $d=-0.4278\times10^5$ d/cm, $e=-2.0104\times10^5$ d/cm. Due to the degeneracies, there exists a certain freedom in the choice of the normal coordinates. The most suitable choice will appear obvious in the course of the derivation.

As a first step, one introduces the following "geometrical symmetry"[10] coordinates:

$$\begin{array}{llll} s=q_1+q_2+q_3+q_4+q_5+q_6 & d_1=q_1-q_2 & \xi & \\ p=(q_3+q_4)-(q_5+q_6) & d_3=q_3-q_4 & \eta & (32)\\ r=2(q_1+q_2)-(q_3+q_4)-(q_5+q_6) & d_5=q_5-q_6 & \zeta & \end{array}$$

One obtains $\mathbf{w}$ as a linear combination of those 9 coordinates by inspection of 9 particular displacements (without translation or rotation):

$$\mathbf{w}=[s(6^{\frac12}/24)+(d_1+d_3+d_5)(6^{\frac12}/12)-(\xi+\eta+\zeta)\sqrt3/4]\mathbf{K} +[r(\sqrt3/12)+(2d_1-d_3-d_5)(\sqrt3/24)+(2\xi-\eta-\zeta)(6^{\frac12}/8)]\mathbf{I} +[\tfrac14 p+(d_3-d_5)\tfrac18+(\eta-\zeta)3\sqrt2/8]\mathbf{J}. \quad (33)$$

A simpler expression is obtained if one introduces the new coordinates,[11]

$$\begin{array}{ll} X_1=(\sqrt3/3)(\xi+\eta+\zeta), & D_1=(\sqrt3/3)(d_1+d_3+d_5),\\ X_2=(6^{\frac12}/6)(2\xi-\eta-\zeta), & D_2=(6^{\frac12}/6)(2d_1-d_3-d_5),\\ X_3=(\sqrt2/2)(\eta-\zeta), & D_3=(\sqrt2/2)(d_3-d_5). \end{array} \quad (34)$$

Then

$$\mathbf{w}=\left[\frac{6^{\frac12}}{24}s+\frac14(2)^{\frac12}D_1-3X_1)\right]\mathbf{K}+\left[\frac{\sqrt3}{12}r+\frac14\left(\frac{\sqrt2}{2}D_2+3X_2\right)\right]\mathbf{I} +\left[\frac14 p+\frac14\left(\frac{\sqrt2}{2}D_3+3X_3\right)\right]\mathbf{J}. \quad (35)$$

From Eqs. (31), (32), and (34)

$$2T=3m\Sigma_i\,\dot X_i^2+\tfrac14m\Sigma_i\,\dot D_i^2+24ms^2+\tfrac{1}{12}m(\dot r^2+3\dot p^2), \quad (36a)$$

$$2V=a\Sigma_i\,X_i^2+2e\Sigma_i\,X_iD_i+\tfrac12(b-d)\Sigma_i\,D_i^2 +\tfrac16(4c+b+d)s^2+\tfrac{1}{12}(b+d-2c)(r^2+3p^2). \quad (36b)$$

This suggests the following choice of normal coordinates: $\sigma=m^{\frac12}s/24$ related to frequency ω_1; $\tilde\omega=(\frac14m)^{\frac12}p$ and $\chi=(\frac{1}{12}m)^{\frac12}r$ to ω_2; U_1, U_2, and U_3 to ω_3; V_1, V_2, and V_3 to ω_4. The U_i and V_i ($i=1, 2, 3$) are derived from $(3m)^{\frac12}X_i$ and $(\frac14m)^{\frac12}D_i$ by the linear orthogonal transformation

$$\begin{array}{l} (3m)^{\frac12}X_i=U_i\cos\beta+V_i\sin\beta\\ (\frac14m)^{\frac12}D_i=-U_i\sin\beta+V_i\cos\beta, \end{array} \quad (37)$$

such that the cross terms vanish in the expression of V as a function of the normal coordinates.

[9] D. M. Dennison, Revs. Modern Phys. **12**, 175 (1940); see also J. E. Rosenthal, Phys. Rev. **45**, 538 (1936), and Phys. Rev. **46**, 730 (1934).

[10] J. E. Rosenthal and G. M. Murphy, Revs. Modern Phys. **8**, 317 (1938).

[11] The X_i and D_i are related to the ξ, η, ζ, α, β, γ introduced by Rosenthal (reference 10) in the following way: $X_1=\zeta$, $X_2=\eta$, $X_3=-\xi$, $D_1\sqrt2=-\gamma$, $D_2\sqrt2=-\beta$, $D_3\sqrt2=\alpha$.

Substituting Eq. (37) in Eq. (36) leads to the condition

$$\cot\beta=(\sqrt{3}/2)e^{-1}[b-d-\tfrac{1}{6}a]=-0.27709.$$

There is an irrelevant arbitrariness in the choice of β itself; in order to have the U_i and the V_i correspond to ω_3 and ω_4, respectively, one takes

$$\cos\beta=0.79595, \quad \sin\beta=-0.60540. \tag{38}$$

Substitution of Eq. (37) in Eq. (35) gives the desired expression for $\mathbf{w}$:

$$m^{\frac{1}{2}}\mathbf{w}=(\tfrac{1}{2}\sigma+a_1U_1+b_1V_1)\mathbf{K}+(\tfrac{1}{2}\chi+a_2U_2+b_2V_2)\mathbf{I}+(\tfrac{1}{2}\bar{\omega}+a_2U_3+b_2V_3)\mathbf{J}, \tag{39}$$

with

$$a_1=-\tfrac{1}{4}\sqrt{3}\cos\beta-\tfrac{1}{2}\sqrt{2}\sin\beta, \quad a_2=\tfrac{1}{4}\sqrt{3}\cos\beta-\tfrac{1}{4}\sqrt{2}\sin\beta,$$
$$b_1=\tfrac{1}{2}\sqrt{2}\cos\beta-\tfrac{1}{4}\sqrt{3}\sin\beta, \quad b_2=\tfrac{1}{4}\sqrt{2}\cos\beta+\tfrac{1}{4}\sqrt{3}\sin\beta. \tag{40}$$

The calculation of $B_{n_1n_2n_3n_4}$ follows from Eq. (39). One gives here the derivation of B_0 only. The B's relative to inelastic scattering, except for more complication in writing, are derived in the same way and their expression is given in the Appendix:

$$B_0=|\langle 0|\exp(i\mathbf{P}\cdot\mathbf{w}/\hbar)|0\rangle|^2. \tag{41}$$

Calling P_x, P_y, P_z the components of $\mathbf{P}$ in the referential ($\mathbf{I}$, $\mathbf{J}$, $\mathbf{K}$), one gets from Eq. (39)

$$\mathbf{P}\cdot\mathbf{w}/\hbar=(1/\hbar m^{\frac{1}{2}})\{(\tfrac{1}{2}\sigma+a_1U_1+b_1V_1)P_z+(\tfrac{1}{2}\chi+a_2U_2+b_2V_2)P_x+(\tfrac{1}{2}\bar{\omega}+a_2U_3+b_2V_3)P_y\}. \tag{42}$$

Substituting Eq. (42) in Eq. (41), one obtains B_0 as a product of 9 absolute squared matrix elements of one-dimensional oscillators of the type given in Eq. (8).

$$B_0=\exp[-(P_z^2/2m)((1/4\hbar\omega_1)+(a_1^2/\hbar\omega_3)+(b_1^2/\hbar\omega_4))-((P_x^2+P_y^2)/2m)((1/4\hbar\omega_2)+(a_2^2/\hbar\omega_3)+(b_2^2/\hbar\omega_4))].$$

Introducing spherical coordinates for $\mathbf{P}$ with $0H_0$ as polar axis, it can be put in the form of Eq. (11):

$$B_0=\exp[-t(2\hbar\omega_3)^{-1}(P^2/2m)], \quad t=A_1u^2+A_2, \tag{43}$$

$$A_1=(\omega_3/2\omega_1)-(\omega_3/2\omega_2)+2(a_1^2-a_2^2)+(b_1^2-b_2^2)(2\omega_3/\omega_4) \tag{44a}$$
$$=-0.2895,$$

$$A_2=(\omega_3/2\omega_1)+2a_2^2+b_2^2(2\omega_3/\omega_4) \tag{44b}$$
$$=1.1128.$$

With those numerical values, the treatment of the elastic cross section follows at once from Eq. (17), namely,

$$\sigma_0/\sigma_f=Q+(R/y)\Theta+(S/y)\Theta^2+\cdots \tag{45}$$

with

$$Q=y^{-1}\int_0^1 t^{-1}[1-\exp(-s^2ty)]du, \tag{45a}$$

$$R=\int_0^1 s(2-s)(\tfrac{1}{2}-s^2ty)\exp(-s^2ty)du, \tag{45b}$$

$$S=\int_0^1 ts^2(2-s)^2(-\tfrac{3}{8}+\tfrac{3}{2}s^2ty-\tfrac{1}{2}s^4t^2y^2)\exp(-s^2ty)du. \tag{45c}$$

The numerical calculations have been carried through at room T ($kT=0.0258$ volt) for E_0 below the threshold of the first inelastic collision. The lower range of energy has been treated by expanding Q, R, S in power of y and integrating analytically the coefficients of this expansion, exactly in the same way as in the H_2 case; for greater values of E_0, Q, R, S merely have been integrated numerically. Contrary to the H_2 case, the term in Θ^2 in the expansion could not be neglected; in fact, for the same value of T, Θ is not so small here and the convergence of the series (45) in powers of Θ is slower. For the same reason the asymptotic form for low E_0 differs much more from the MTR result than in the H_2 case, as may be seen in Table I.

TABLE IV. Energy dependence of the scattering cross section per proton in CH_4 at room T in the region of pure elastic scattering when $T=300°K$. Comparison with experiment.

E_0	y	Q	$(R/y)\Theta$	$(S/y)\Theta^2$	Theoretical value σ/σ_f	Experimental value (σ/σ_f)exp
0.0056	0.0166	2.337	1.416	−0.058	3.695	3.551±0.045
0.0068	0.0202	2.333	1.137	−0.046	3.424	3.431±0.055
0.0083	0.0246	2.307	0.901	−0.035	3.174	3.174±0.065
0.0104	0.0309	2.291	0.677	−0.026	2.943	2.894±0.04
0.0126	0.0374	2.273	0.538	−0.019	2.792	2.758±0.02
0.0144	0.0428	2.258	0.446	−0.015	2.689	2.740±0.025
0.0168	0.0499	2.240	0.364	−0.012	2.592	2.648±0.02
0.0196	0.0582	2.218	0.291	−0.009	2.500	2.532±0.02
0.0233	0.0692	2.192	0.222	−0.006	2.408	2.411±0.01
0.0256	0.0760	2.172	0.190	−0.005	2.357	2.387±0.01
0.0439	0.1304	2.040	0.059	−0.002	2.097	2.115±0.01
0.0979	0.2908	1.708	−0.016		1.692	1.677±0.01
0.146	0.4337	1.474	−0.023		1.451	1.460±0.01
0.1788	0.53125	1.341	−0.021		1.320	

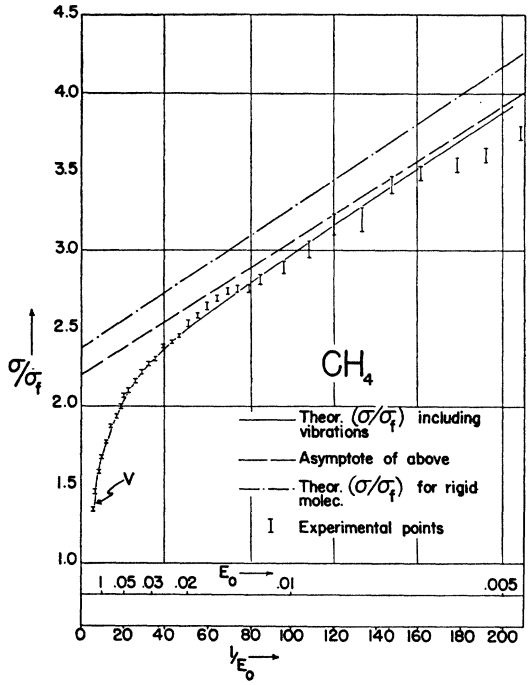

FIG. 4. Energy dependence of the slow neutron scattering cross section per proton by CH_4 molecules at room temperature ($T=300°K$): theoretical curve given by MTV theory and experimental points. The line given by MTR theory is also reported. Arrow V indicates the energy threshold for inelastic collisions.

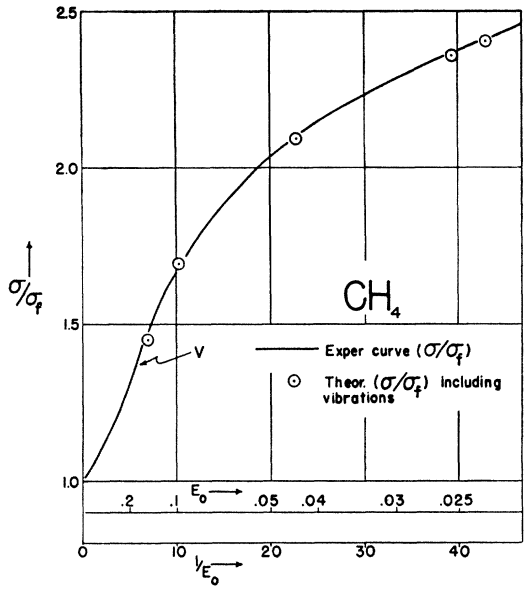

FIG. 5. Energy dependence of the slow neutron scattering cross section per proton by CH_4 molecules at room temperature ($T=300°K$): experimental curves and theoretical points given by MTV theory. Arrow V indicates the energy threshold for inelastic collisions.

σ/σ_f is plot against $1/E_0$ on Fig. 4; the asymptote of the curve and the MTR line are also drawn on Fig. 4. Table IV gives, at some specific values of E_0 and at room temperature, the values of σ/σ_f and the contribution to them of the first terms in the expansion in power of Θ. One sees that the thermal effect is much less important than in the H_2 case; this must be expected because, on the average, the velocity $\mathbf{V}_s$ of the scattering proton is much smaller in CH_4 than in H_2 at the same temperature.

When E_0 exceeds $T_{0010}=0.1788$ volt, inelastic collisions contribute to the total scattering process. Even if one neglects the rather small thermal effect, the numerical calculation of σ/σ_f soon becomes very cumbersome. The general analytical formula for the inelastic cross sections neglecting thermal effect is given in the Appendix.

V

This section is devoted to a discussion of the previous results and to a comparison with experimental data.

The limitation of the theory to scattering media of sufficiently large temperature has been already studied in the preceding sections. As far as the incident energy of the neutron is concerned, the calculation of the elastic cross section is based, in both cases, on two approximations: (a) the mass tensor approximation; (b) the thermal approximation, i.e., the neglect of higher values of $v_{||}$ which allowed one to make expansion (18). As E_0 decreases, both approximations become poorer and the whole theory progressively loses its validity. To put the thing in a more quantitative fashion, one may define critical values E_a, E_b, of E_0, under which, crudely speaking, each of the above approximations breaks down, namely: (a) A "mass tensor" critical value E_a. When $E_0=E_a$, the neutron is able to transfer to half of the molecules the energy necessary for a transition to the next higher rotational level; i.e., the reduced energy of the neutron is equal to the average level spacing of the rotational states of the molecule. (b) A "thermal critical" value E_b. When $E_0=E_b$, the number of states such that $\frac{1}{2}mv_{||}^2>E_0$, must be small compared with the number of other states. We shall take for E_b the angular average (integration over u) of $\langle\frac{1}{2}mv_{||}^2\rangle_T$.

The following numerical values (in volts) are found, for the E_a, E_b, relative to the scattering cross section per proton in H_2 and CH_4, at room temperature:

	E_a	E_b	
H_2	0.06	0.01	(46)
CH_4	0.01	0.004	

Below the threshold of inelastic collisions our numerical results can be compared with the recent measurements performed with the Columbia velocity selector on H_2 and CH_4 samples at room temperature.[12] For this purpose, one defines $(\sigma/\sigma_f)_{\text{exp}}=\sigma_{\text{exp}}/\sigma_{f\text{exp}}$. σ_{exp} is the experimental scattering cross section per proton. In the H_2 case, it is deduced at once from the experimental data by subtracting the capture cross section. In the CH_4 case, one has in addition to subtract the scattering cross section $\sigma^{(c)}$ by the C nucleus. The value $\sigma^{(c)}=\sigma_f{}^{(c)}=4.70$ barns has been taken ($\sigma_f{}^{(c)}$ cross section by free carbon nuclei). Strictly speaking,[13] $\sigma^{(c)}$ depends on E_0 and the exact formula for the ratio $\sigma^{(c)}/\sigma_f{}^{(c)}$ is given in the Appendix. But, in the range of energy that is considered here, the binding effect is very small and may be neglected in the derivation of σ_{exp}. $\sigma_{f\text{exp}}$ is an experimental value of the scattering cross section by free protons. The value derived by Melkonian from his scattering experiments on H_2 at higher energy has been adopted, namely, 20.36 barns.[14]

The values $(\sigma/\sigma_f)_{\text{exp}}$ so obtained have been reported in Figs. 1 (H_2), 4 (CH_4), and 5 (CH_4), and also in Tables III (H_2) and IV (CH_4). The experimental errors indicated in the tables and graphs correspond to statistical errors in σ_{exp}; all experimental points are subject to a common supplementary error due to a possibly inaccurate value of $\sigma_{f\text{exp}}$. The arrow, V, in each of Figs. 1, 4, and 5 indicates the threshold of the first inelastic collision.

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[12] E. Melkonian, Phys. Rev. **76**, 1750 (1949).

[13] $\sigma^{(c)}$ is in fact slightly greater than $\sigma_f{}^{(c)}$. This may be seen as the consequence of two effects: (1) The binding effect proper. It amounts to give to the neutron a reduced mass somewhere in between the one for the system (neutron+C) and the one for (neutron+CH_4). (2) The thermal (or doppler) effect, which gives a net increase of $(kT/32E_0)\sigma^{(c)}$ in the cross section. It follows that

$$1<\sigma^{(c)}/\sigma_f{}^{(c)}<1.04+(kT/32E_0).$$

[14] E. Melkonian, Phys. Rev. **76**, 1744 (1949).

Looking at H_2 first, one notices a quite satisfactory agreement where it is expected. In Fig. 1 the diagram σ/σ_f *vs* $1/E_0$ has been extended far below its expected range of validity. It may be crudely divided in three regions: when $E_0 > E_a = 0.06$ volt, theory and experiment agree; when E_0 goes from E_a to $E_b = 0.01$ volt, i.e., in a region where most of the H_2 molecules can suffer only elastic or hyperelastic collisions as far as rotations are concerned, discrepancies occur, but they are not very large and the experimental points follow a somewhat fluctuating line approximately 5 percent above the theoretical curve; when E_0 goes below E_b, the calculated thermal effect is notably too big and the experimental points drop below the theoretical curve.

The same features are noted with CH_4. The agreement is very satisfactory at higher energy, $E_0 > E_a = 0.01$ volt. This is particularly well emphasized on Fig. 5, where the experimental curve is drawn and 5 theoretical points are reported. Figure 4 shows again a diagram σ/σ_f *vs* $1/E_0$ extended far beyond the expected range of validity. Following the experimental points on the side of low E_0, one sees them fluctuate more and more around the theoretical curve as E_0, goes beyond E_a, and drop below it as E_0 approaches $E_b = 0.004$ volt.

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APPENDIX

A. H_2 Case: Exact Integration of $\bar{\sigma}$ When $y = n$

At such value of y: $v_\pm = \frac{1}{4}[u/(1 \mp \frac{1}{2}u)]^2 n$, and $\bar{\sigma}_n/\sigma_f = (1/n) \times \int_0^1 [f_n(v_+) - f_n(v_-)](du/u^2)$. Integration by parts gives

$$n\bar{\sigma}_n/\sigma_f = f_n(n/9) - f_n(n) + \int_0^1 [(v_+{}^n e^{-v_+}/n!)(dv_+/du) - (v_-{}^n e^{-v_-}/n!)(dv_-/du)][(du/u).$$

Taking v_+, v_- as new variables leads to

$$n\bar{\sigma}_n/\sigma_f = f_n(n/9) - f_n(n) + \int_0^n [(n^{\frac{1}{2}} + v^{\frac{1}{2}})/2v^{\frac{1}{2}}][v^n e^{-v}/n!]dv - \int_0^{n/9} [(n^{\frac{1}{2}} - v^{\frac{1}{2}})/2v^{\frac{1}{2}}][v^n e^{-v}/n!]dv$$
$$= \tfrac{3}{2} f_n(n/9) - \tfrac{1}{2} f_n(n) + \tfrac{1}{2} n^{\frac{1}{2}} \int_{n/9}^n (v^{n-\frac{1}{2}} e^{-v}/n!)dv.$$

The integral in the lower right-hand side is easily expressible as a linear combination of elementary functions and error integrals.

B. General Formulas for Diatomic Molecules

The formulas given here are relative to the cross section for scattering by a nucleus in any diatomic molecule. Let S be the scattering nucleus, T the other one; call their masses, respectively,

$$m_s = \mu_s m, \quad m_t = \mu_t m.$$

The components of the mass tensors are

$$\mu_1 = \mu_s + \mu_t, \ \mu_2 = \mu_s, \ \nu_1 = (\mu_s + \mu_t)/(1 + \mu_s + \mu_t), \ \nu_2 = \mu_s/(1 + \mu_s),$$

and

$$2/s = [(1 + \mu_s)/\mu_s] - [\mu_t/\mu_s(\mu_s + \mu_t)]u^2.$$

The displacement is $w = (m\mu_n)^{\frac{1}{2}} q$, with $\mu_n = \mu_s(\mu_s + \mu_t)/\mu_t$. The relation between σ_∞ and σ_f is $\sigma_f = [\mu_s/(1 + \mu_s)]^2 \sigma_\infty$.

The axis of the molecule is again an axis of symmetry for the scattering problem. The treatment of Sec. II applies, taking $t = 2u^2/\mu_n$. The expression corresponding to Eq. (24) is then easily deduced.

As far as the inelastic cross sections are concerned, Eq. (25) has to be replaced by

$$\bar{\sigma}_n/\sigma_f = (\mu_s/(1 + \mu_s))^2 (\mu_n/2y) \int_{u_0}^1 [f_n(v_+) - f_n(v_-)](du/u^2)$$

with the same definition of f_n and

$$v_\pm = (u^2 s/2\mu_n)[(sy)^{\frac{1}{2}} \pm (sy - n)^{\frac{1}{2}}]^2,$$
$$u_0 = \begin{cases} 0 & \text{if } y > n/2\nu_2 \\ (\mu_n/\nu_2)^{\frac{1}{2}}(1 - 2\nu_2 y/n)^{\frac{1}{2}} & n/2\nu_2 > y > n/2\nu_1 \\ 1 & n/2\nu_1 > y. \end{cases}$$

The same asymptotic study may be carried through with help of the model of the isotropic oscillator, taking $\mu' = 2\mu_n/u^2 s$ and $\epsilon' = sy$. This leads to

$$\sigma/\sigma_f \sim 1 + (1/24\mu_n y)$$

in agreement with expression (27).

C. Inelastic Scattering Cross Section per Proton in CH_4, Neglecting Thermal Effect

$B_{n_1 n_2 n_3 n_4}$ is calculated from Eqs. (39), (40) and (7). Setting

$$N = n_1 + n_2 + n_3 + n_4,$$
$$\tau_1 = (\omega_3/2\omega_1)u^2, \quad \tau_2 = (\omega_3/2\omega_2)(1 - u^2),$$
$$\tau_3 = 2[(a_1^2 - a_2^2)u^2 + a_2^2], \quad \tau_4 = (2\omega_3/\omega_4)[(b_1^2 - b_2^2)u^2 + b_2^2],$$

one gets

$$B_N = \left[\prod_{i=1}^4 (\tau_i^{n_i}/n_i!)\right][(2\hbar\omega_3)^{-1}(P^2/2m)]^N \exp[-t(2\hbar\omega_3)^{-1}(P^2/2m)].$$

(The subscript N is a short writing for $n_1 n_2 n_3 n_4$.) Also

$$E_N \equiv 2\hbar\omega_3 y_N = \sum_{i=1}^4 n_i \hbar\omega_i.$$

Then, applying Eqs. (10) and (10′) with $v_{||} = 0$,

$$\bar{\sigma}_N/\sigma_f = (1/y) \int_{u_0}^1 [f_N(v_+) - f_N(v_-)](N!/t^N) \left[\prod_{i=1}^4 (\tau_i^{n_i}/n_i!)\right](du/t)$$

with

$$v_\pm = \tfrac{1}{4} ts[(sy)^{\frac{1}{2}} \pm (sy - 2y_N)^{\frac{1}{2}}]^2,$$
$$u_0 = \begin{cases} 0 & \text{if } y > y_N/\nu_2 \\ [\nu_1/(\nu_1 - \nu_2)]^{\frac{1}{2}}[1 - (\nu_2 y/y_N)]^{\frac{1}{2}} & \text{if } y_N/\nu_2 > y > y_N/\nu_1 \\ 1 & \text{if } y_N/\nu_1 > y. \end{cases}$$

D. Scattering Cross Section by the Carbon Nucleus in CH_4

The carbon nucleus is involved only in the 3-fold degenerate vibrations. Its displacement $\mathbf{w}_c$ is given by

$$(48m)^{\frac{1}{2}} \mathbf{w}_c = (U_1 \cos\beta + V_1 \sin\beta)\mathbf{K} - (U_2 \cos\beta + V_2 \sin\beta)\mathbf{I} - (U_3 \cos\beta + V_3 \sin\beta)\mathbf{J}.$$

The sum of squared matrix elements $B_{n_3n_4}{}^{(c)}$ is

$$B_{n_3n_4}{}^{(c)}=(\gamma_3{}^{n_3}/n_3!)(\gamma_4{}^{n_4}/n_4!)[(2\hbar\omega_3)^{-1}(P^2/2m)]^{n_3+n_4}\times\exp[c(2\hbar\omega_3)^{-1}(P^2/2m)],$$

where

$$\gamma_3=(1/24)\cos^2\beta,\quad \gamma_4=(1/24)(\omega_3/\omega_4)\sin^2\beta,\quad c=\gamma_3+\gamma_4.$$

The reduced mass of the system (neutron+CH_4) is $\nu=16/17$. Setting $s=2\nu$, one obtains formulas quite analogous to those of the cross section per proton. It must be emphasized, however, that these formulas do not involve any mass tensor approximation. Below the threshold of the first inelastic collision, 0.1788 volt, one gets

$$\sigma^{(c)}/\sigma_f{}^{(c)}=\tfrac{1}{4}(13/12)^2\{[1-\exp(-s^2cy)]/cy+(\Theta/y)s(2-s)(\tfrac{1}{3}-s^2cy)\exp(-s^2cy)+(\Theta^2/y)cs^2(2-s)^2(-\tfrac{3}{8}+\tfrac{3}{2}s^2cy-\tfrac{1}{2}s^4c^2y^2)\exp(-s^2cy)+\cdots\}.$$

The general formula for inelastic scattering is

$$\bar{\sigma}_{n_3n_4}{}^{(c)}/\sigma_f{}^{(c)}=\tfrac{1}{4}(13/12)^2(1/y)(N!/c^{N+1})\times(\gamma_3{}^{n_3}/n_3!)(\gamma_4{}^{n_4}/n_4!)[f_N(v_+)-f_N(v_-)]$$

with

$$N=n_3+n_4,\quad v_\pm=\tfrac{1}{4}cs[(sy)^{\frac{1}{2}}\pm(sy-2y_N)^{\frac{1}{2}}]^2,$$
$$y_N\equiv(2\hbar\omega_3)^{-1}E_N=\tfrac{1}{2}[n_3+(\omega_4/\omega_3)n_4].$$

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The Scattering of Fast Neutrons by Bismuth and Lead

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Using good geometry, homogeneous fast neutrons of energy 4.3 Mev, supplied by the D−D reaction in the Bartol Van de Graaff statitron, have been scattered in bismuth and lead. Using binocular microscopes and Eastman NTA plates, fifty thousand fields of view were examined to obtain 5000 acceptable recoil proton tracks. The energy spectrum of the scattered neutrons was studied to establish the presence or absence of any inelastic groups. From a consideration of gross effects, it is concluded that no inelastic group exists in the bismuth data with an intensity greater than five percent of the elastically scattered neutrons, or that no level in Bi^{209} lying between the ground state and 3.4 Mev is excited by a neutron group of more than five percent of the intensity of the elastic group. Appreciable inelastic scattering was noted in naturally occurring lead, the inelastic scattering cross section being less than half the elastic cross section. The energy distribution of the inelastically scattered neutrons is interpreted to indicate an energy level in lead in the vicinity of 3.3 Mev. This level is consistent with radioactivity measurements which show that most of the beta-disintegrations of $ThC''\xrightarrow{\beta}{}^*Pb^{208}$ lead to energy levels in the vicinity of 3.2 Mev, and with Q-values of the reaction $Pb^{207}(d,p)Pb^{208}$ which disclose an energy level at 3.45 Mev in the nucleus Pb^{208}.

INTRODUCTION

WHEN neutrons are scattered by nuclei, both elastic and inelastic scattering can occur. The elastic process takes either of two forms, potential scattering or resonance scattering. The former is sometimes referred to as "hard sphere" or "diffraction" scattering, and the latter may be termed "capture elastic scattering." The capture elastic scattering and inelastic scattering are considered to be two phases of the same nuclear process, because an intermediate compound nucleus is actually formed, the product nucleus being left in the ground state after the elastic collision and in an excited state after the inelastic collision. In the case of potential or diffraction scattering, the compound nucleus is not formed. The foregoing terminology has been previously outlined by Feld.[1]

Neutrons from the deuteron-deuterium reaction have been employed in two previous scattering experiments dealing with the elastic and inelastic scattering of fast neutrons by lead.[2,3] These measurements have been interpreted by Feld[1] to give energy levels in naturally occurring lead at ~0.8 Mev,[2] and in the vicinity of 0.92, 1.87, and 2.67 Mev.[3]

It was decided to irradiate both bismuth and lead with fast neutrons to observe the inelastically scattered neutrons, because in both cases magic numbers are involved.[4] All of the naturally occurring isotopes of lead are "magic" for protons, since each of them contains 82 protons; Pb^{208}, in particular, may be regarded as a double closed shell nucleus, because it contains 82 protons and 126 neutrons, both of which are magic numbers. The monoisotopic Bi^{209} contains 83 protons and 126 neutrons and therefore is magic for neutrons.[4] Magic number nuclei should have widely spaced energy levels and should not, therefore, follow the statistical theory of Weisskopf.[5]

The previously mentioned data[2,3] were obtained with the use of "poor geometry," whereas the measurements of the present paper employed "good geometry" for better resolution.

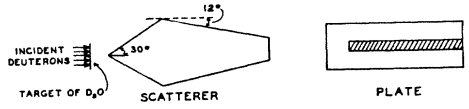

FIG. 1. Geometric arrangement for fast neutron scattering. The average length of the scatterer is 6.1 cm.

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