

Slow Neutron Scattering by Molecular Hydrogen and Deuterium*

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The cross sections for neutrons scattering from H_2 and D_2 have been calculated taking into account the spin correlations, rotations, and vibrations of the molecules exactly, to the extent that the vibrations are harmonic and do not interact with the rotations. Free translations of the molecules are assumed, but this assumption is expected to be valid in the liquid, as well as the gas, for neutron energies above 0.007 eV, the Debye temperature for hydrogen. Numerical results are given for the total cross section for both ortho-hydrogen and parahydrogen gas at 20.4°K which agree reasonably well with the limited experimental results available. Also, curves of the double differential cross section are shown for selected incident neutron energies and scattering angles. These latter curves show very clearly the various rotational and vibrational transitions. The formulas given here are applicable at all temperatures below the thermal excitation of the first vibrational level.

I. INTRODUCTION

THE technique of slow neutron scattering has proved most valuable as a probe for studying molecular systems,¹ and much work has been devoted to the development of calculational methods and models which will adequately describe the scattering from such systems. The H_2 molecule is not only perhaps the simplest of all but, as a liquid, hydrogen is important both as a cold neutron source and, in special cases, as a shield. However, up to the present, the only calculations that have been performed^{2,3} are applicable to high temperatures ($\geq 400^\circ K$) with high incident neutron energies (≥ 0.06 eV), and to low temperatures ($\leq 30^\circ K$) with low neutron energies (≤ 0.09 eV).^{4,5} One other detailed calculation by Brimberg⁶ which has a wider range of validity is not applicable to differential cross sections, since a thermal average is taken only in the total cross section.

The purpose of this present paper is to derive the cross section for neutrons scattering from hydrogen gas in the energy range from 0 eV to roughly 3 eV, and for temperatures $\leq 3000^\circ K$ (the vibrational levels are assumed unpopulated). The results are applicable to liquid hydrogen, with the restriction that for incident neutron energies less than that of the Debye temperature (0.007 eV) the results are not expected to be accurate. Spin correlations, rotations, and vibrations are taken into account exactly, to the extent that vibration-rotation coupling can be neglected, and that the vibrations are harmonic. Some numerical results

are given, and these are compared with available experimental data. It is hoped that when more detailed experimental information is obtained, the calculation given here will prove useful in evaluating the assumptions that enter into the theory for more complicated molecules.

Since the only significant calculational difference between D_2 and H_2 is the effect of spin correlations, these have been evaluated, and thus we have derived the cross section for D_2 in addition to that for H_2 .

II. GENERAL FORMULATION

In units where $\hbar = 1$, the differential cross section for the scattering of neutrons from any molecular system with initial wave function ψ_i and final wave function ψ_f is¹

$$\frac{d^2\sigma}{d\Omega d\epsilon} = \frac{1}{2\pi} \frac{k}{k_0} \sum_f \int_{-\infty}^{\infty} dt e^{+i\epsilon t} [e^{-i(E_i - E_f)t} \times |\langle \psi_f | \sum_m \exp(i\mathbf{k} \cdot \mathbf{x}_m) a_m | \psi_i \rangle|^2]_T. \quad (1)$$

Here k_0 , k are initial and final neutron momenta, respectively, E_i , E_f are the initial and final neutron energies, ϵ the neutron energy transfer, \mathbf{k} the neutron momentum transfer, \mathbf{x}_m and a_m the position vector and scattering amplitude of the m th atom, respectively, and the subscript T implies that a thermal average is to be taken over the initial states.

The H_2 molecule is dumbbell-shaped with a separation a . The molecule can then vibrate along the line joining the atoms, and rotate about the center point of that line. We write then for the position vector of a hydrogen atom:

$$\mathbf{x}_m = \mathbf{r}_l + (-)^n \mathbf{R}_l / 2, \quad n = 1, 2,$$

where \mathbf{r}_l is the position coordinate of the molecular center-of-mass, and \mathbf{R}_l the relative coordinate of the two atoms of the l th molecule. Using this separation,

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¹ A. C. Zemach and R. J. Glauber, Phys. Rev. **101**, 118 (1956).

² A. M. L. Messiah, Phys. Rev. **84**, 204 (1951).

³ T. J. Krieger and M. Nelkin, Phys. Rev. **106**, 290 (1957).

⁴ G. Sarma, in *Proceedings of the International Atomic Energy Agency Symposium on Inelastic Scattering of Neutrons in Liquid and Solids, Vienna, 1960* (International Atomic Energy Agency, 1961).

⁵ M. Hammermesh and J. Schwinger, Phys. Rev. **71**, 678 (1947).

⁶ S. Brimberg, in *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, p. 79.

Eq. (1) becomes

$$\begin{aligned} \frac{d^2\sigma}{d\Omega d\epsilon} &= \frac{1}{2\pi} \frac{k}{k_0} \sum_f \int_{-\infty}^{\infty} dt [e^{+i(\epsilon - E_i + E_f)t} \\ &\times \sum_{l \neq j, n, p=1}^2 \langle \psi_i | a_{jp} \exp[-i\mathbf{k} \cdot \mathbf{r}_j - i(-)^p \mathbf{R}_j \cdot \boldsymbol{\kappa}/2] | \psi_f \rangle \\ &\cdot \langle \psi_f | a_{ln} \exp[i\mathbf{k} \cdot \mathbf{r}_l + i(-)^n \mathbf{R}_l \cdot \boldsymbol{\kappa}/2] | \psi_i \rangle]_T \\ &+ \frac{1}{2\pi} \frac{k}{k_0} \sum_f \int_{-\infty}^{\infty} dt [e^{+i(\epsilon - E_i + E_f)t} \\ &\times \sum_{l, n, p=1}^2 \langle \psi_i | a_{lp} \exp[-i\mathbf{k} \cdot \mathbf{r} - i(-)^p \boldsymbol{\kappa} \cdot \mathbf{R}_l/2] | \psi_f \rangle \\ &\cdot \langle \psi_f | a_{ln} \exp[i\mathbf{k} \cdot \mathbf{r}_l + i(-)^n \boldsymbol{\kappa} \cdot \mathbf{R}_l/2] | \psi_i \rangle]_T. \quad (2) \end{aligned}$$

First we consider the second term which refers to a single H₂ molecule, and which we refer to as the "self" term. If we make the good assumption that the translational modes can be separated, then we can write for the "self" term

$$\begin{aligned} \frac{d^2\sigma_s}{d\Omega d\epsilon} &= \frac{1}{2\pi} \frac{k}{k_0} \sum_f \int_{-\infty}^{\infty} dt \sum_l \langle \psi_{il} | \exp[-i\mathbf{k} \cdot \mathbf{r}_l(0)] \\ &\times \exp[i\mathbf{k} \cdot \mathbf{r}_l(t)] | \psi_{il} \rangle_T \\ &\times [|\langle \psi_f | \sum_{n=1}^2 a_{ln} \exp[(-)^n i\mathbf{k} \cdot \mathbf{R}_l/2] | \psi_i \rangle|^2 \\ &\times \exp[+i(\epsilon - E_i' + E_f')t]]_T, \quad (3) \end{aligned}$$

where ψ_{il} is the initial wave function of the translational modes, and E_i' , E_f' are the energies of the rotational and vibrational states only. It is well to point out here that due to the vibrations R is not constant.

II.1 Spin Correlations

The nuclei of the H₂ molecule can form states of total spin $S=0$ or $S=1$, the former corresponding to parahydrogen and the latter to orthohydrogen. For $S=0$ only states of total angular momentum J having even values can occur, and for $S=1$ only odd values of J are allowed. In this section we compute the effects in Eq. (3) that are due to the nuclear spins. Call

$$g(t) = \left[\sum_f |\langle \psi_f | \sum_{n=1}^2 a_n \exp[(-)^n i\mathbf{k} \cdot \mathbf{R}/2] | \psi_i \rangle|^2 \times \exp[-i(E_i' - E_f')t] \right]_T. \quad (4)$$

We can write $g(t)$ explicitly in the form

$$\begin{aligned} g(t) &= \sum_{J,S} \frac{P_{JS}}{2J+1} \frac{1}{2S+1} \frac{1}{2} \sum_{\sigma_z, \sigma_z' = \pm \frac{1}{2}} \sum_{J'} e^{i(E_{J'} - E_J)t} \\ &\times \sum_{n=0} e^{in\omega t} \sum_{J_z J_z'} \sum_{S' S_z S_z'} |\langle J' J_z', S' S_z', \sigma_z', n \\ &\times | A | J J_z, S S_z, \sigma_z, n=0 \rangle|^2, \quad (5) \end{aligned}$$

which includes a thermal average over the initial states, where the parameters are defined as follows: J, J' are initial and final angular momenta of the molecule (J_z and J_z' are their z components); S, S' are initial and final total spin of the molecule (S_z and S_z' are their z components); σ_z, σ_z' are initial and final z components of the neutron spin; E_J is rotational energy of the state J ; P_{JS} is statistical weight of the state J with spin S ; ω is quantum of vibrational energy; n is vibrational quantum number;

$$A = a_1 \exp(i\mathbf{k} \cdot \mathbf{R}/2) + a_2 \exp(-i\mathbf{k} \cdot \mathbf{R}/2).$$

Thus, $g(t)$ refers to the rotational, vibrational, and spin-dependent parts of Eq. (3). The molecular Hamiltonian has been taken to be spin-independent and as stated previously the coupling between rotations and vibrations is neglected. Thus $|J, S, n\rangle = |J\rangle |S\rangle |n\rangle$.

Since $\hbar\omega = 0.546$ eV, for hydrogen, all the molecules are initially in their vibrational ground state, $n=0$, the higher states being frozen out except at very high temperature ($\gtrsim 3000^\circ\text{K}$). The rotational energy levels are given by

$$E_J = \frac{\hbar^2 J(J+1)}{4Ma^2} = 0.015 \frac{J(J+1)}{2} \text{ eV}, \quad (6)$$

where M is the proton mass, and a the equilibrium separation distance of the H-H bond. Now let us rewrite the operator A . We recall that the scattering length operator of the proton has the form

$$a_m = \frac{I_m + 1 + 2\mathbf{I}_m \cdot \boldsymbol{\sigma}}{2I_m + 1} a_+ + \frac{I_m - 2\mathbf{I}_m \cdot \boldsymbol{\sigma}}{2I_m + 1} a_-, \quad (7)$$

where I_m is the spin of the proton and a_+ and a_- are the triplet and singlet scattering lengths, respectively, of the neutron-proton interaction. Since $I = \frac{1}{2}$ and $\mathbf{I}_1 + \mathbf{I}_2 = \mathbf{S}$, we find

$$A/2 = a_{\text{coh}} \cos(\boldsymbol{\kappa} \cdot \mathbf{R}/2) + (2/\sqrt{3}) a_{\text{inc}} [\cos(\boldsymbol{\kappa} \cdot \mathbf{R}/2) \boldsymbol{\sigma} \cdot \mathbf{S} + i \sin(\boldsymbol{\kappa} \cdot \mathbf{R}/2) \boldsymbol{\sigma} \cdot (\mathbf{I}_1 - \mathbf{I}_2)], \quad (8)$$

with the usual definitions of coherent and incoherent scattering length:

$$a_{\text{coh}}^2 = \langle a \rangle^2 = [1/(2I+1)^2] [(I+1)a_+ + Ia_-]^2 = \frac{1}{16} (3a_+ + a_-)^2, \quad (9)$$

$$a_{\text{inc}}^2 = \langle a^2 \rangle - \langle a \rangle^2 = [I(I+1)/(2I+1)^2] (a_+ - a_-)^2 = \frac{3}{16} (a_+ - a_-)^2, \quad (10)$$

respectively. First we shall consider transitions between states of the same parity (and hence with the same molecular spin S). In this case only the symmetric part of A contributes to the matrix elements and calling α and β the initial and final states we have

$$|\langle \beta | A | \alpha \rangle_{S=S'}|^2 = |\langle J' J_z' n | \cos(\boldsymbol{\kappa} \cdot \mathbf{R}/2) | J J_z n=0 \rangle|^2 \times |\langle \sigma_z' S' S_z' | P | \sigma_z S S_z \rangle|^2, \quad (11)$$

where

$$P = 2a_{\text{coh}} + (4/\sqrt{3}) a_{\text{inc}} \boldsymbol{\sigma} \cdot \mathbf{S}.$$

Now we may sum over the final spin states with the result

$$\sum_{\sigma_z' S' S_z'} |\langle \beta | A | \alpha \rangle|^2 = |\langle J' J_z' n | \cos(\boldsymbol{\kappa} \cdot \mathbf{R}/2) | J J_z n = 0 \rangle|^2 \times \langle \sigma_z S S_z | P^2 | \sigma_z S S_z \rangle. \quad (12)$$

The square of the Hermitian operator P is

$$P^2 = P^+ P = 4a_{\text{coh}}^2 + (16/3)a_{\text{inc}}^2 (\boldsymbol{\sigma} \cdot \mathbf{S})^2 + (16/\sqrt{3})a_{\text{coh}} a_{\text{inc}} \boldsymbol{\sigma} \cdot \mathbf{S}. \quad (13)$$

Summing over the neutron spin states the last term averages to zero. Since

$$\sum_{\sigma_z} \langle \sigma_z | (\boldsymbol{\sigma} \cdot \mathbf{I}_\nu) (\boldsymbol{\sigma} \cdot \mathbf{I}_\mu) | \sigma_z \rangle = \frac{1}{2} \mathbf{I}_\nu \cdot \mathbf{I}_\mu \quad (14)$$

or

$$\sum_{\sigma_z} \langle \sigma_z | (\boldsymbol{\sigma} \cdot \mathbf{S})^2 | \sigma_z \rangle = \frac{1}{2} \mathbf{S}^2,$$

we get

$$\sum_{\sigma_z} \langle \sigma_z S S_z | P^2 | \sigma_z S S_z \rangle = 8[a_{\text{coh}}^2 + \frac{1}{3} a_{\text{inc}}^2 S(S+1)]. \quad (15)$$

Substituting now Eqs. (12) and (15) into (5) and summing over S we obtain for spin-conserving transitions

$$g_{S=S'}(t) = 4 \sum_J \frac{P_{JS}}{2J+1} \left[a_{\text{coh}}^2 + \frac{a_{\text{inc}}^2}{3} S(S+1) \right] \times \sum_{J'} \exp[i(E_{J'} - E_J)t] \sum_{n=0} e^{in\omega t} \times \sum_{J_z J_z'} |\langle J' J_z' n | \cos(\boldsymbol{\kappa} \cdot \mathbf{R}/2) | J J_z n = 0 \rangle|^2, \quad (16)$$

where the sum $\sum_{J'}$ is over states of same parity as J . For transitions between states of opposite parity (and hence of different spin S) only the antisymmetric part of the operator A gives a nonvanishing contribution to the matrix elements. In this case

$$|\langle \beta | A | \alpha \rangle|_{S \neq S'}^2 = |\langle J' J_z' n | \sin(\boldsymbol{\kappa} \cdot \mathbf{R}/2) | J J_z n = 0 \rangle|^2 \times |\langle \sigma_z' S' S_z' | Q | \sigma_z S S_z \rangle|^2, \quad (17)$$

with

$$Q = (4/\sqrt{3})a_{\text{inc}} \boldsymbol{\sigma} \cdot (\mathbf{I}_1 - \mathbf{I}_2). \quad (18)$$

Again, summing over final spin states and considering that

$$\sum_{\sigma_z} \langle \sigma_z | [\boldsymbol{\sigma} \cdot (\mathbf{I}_1 - \mathbf{I}_2)]^2 | \sigma_z \rangle = \frac{1}{2} (\mathbf{I}_1 - \mathbf{I}_2)^2 = \mathbf{I}_1^2 + \mathbf{I}_2^2 - \frac{1}{2} \mathbf{S}^2, \quad (19)$$

one finds

$$g_{S \neq S'}(t) = 4 \sum_J \frac{P_{JS}}{2J+1} a_{\text{inc}}^2 \left[1 - \frac{S(S+1)}{3} \right] \times \sum_{J'} e^{i(E_{J'} - E_J)t} \sum_{n=0} e^{in\omega t} \times \sum_{J_z J_z'} |\langle J' J_z' n | \sin(\boldsymbol{\kappa} \cdot \mathbf{R}/2) | J J_z n = 0 \rangle|^2, \quad (20)$$

where the sum $\sum_{J'}$ is over states of opposite parity to J . These same results can be obtained for D_2 and are given in Appendix A.

II.2 Vibrations

Since all the molecules are initially in their ground vibrational state, the vibrational transitions are important only if the incident neutron energy is great enough so that $\kappa^2/8M\omega \gtrsim 1$. The neutron then cannot gain energy from the vibrational modes.

The vibrational matrix elements can be evaluated exactly under the assumption that the restoring force of the H-H bond is harmonic in nature. We write $R = a + x$ where x is the amount the bond length is stretched. On expanding x ,

$$x = i(M\omega)^{-1/2} [b - b^\dagger],$$

where b^\dagger , b are the boson creation and annihilation operators, respectively; the matrix elements in Eqs. (16) and (20) are of the form

$$\langle n | e^{\pm i\kappa R \mu/2} | 0 \rangle = e^{\pm i\kappa a \mu/2} \times \langle n | \exp[\mp (\kappa \mu/2)(M\omega)^{1/2} (b - b^\dagger)] | 0 \rangle,$$

where $\mu = \cos\theta$, and θ is the angle between $\boldsymbol{\kappa}$ and \mathbf{R} . Using the relation

$$e^A e^B = e^{A+B+1/2[A, B]_-},$$

the above expression becomes

$$\langle n | e^{\pm i\kappa R \mu/2} | 0 \rangle = \exp\left(\pm i \frac{\kappa a \mu}{2}\right) \exp\left(-\frac{\kappa^2 \mu^2}{8M\omega}\right) \times \left\langle n \left| \exp\left(\pm \frac{\kappa \mu b^\dagger}{2(M\omega)^{1/2}}\right) \exp\left(\mp \frac{\kappa \mu b}{2(M\omega)^{1/2}}\right) \right| 0 \right\rangle = \left(\pm \frac{\kappa \mu}{2(M\omega)^{1/2}}\right)^n \frac{1}{(n!)^{1/2}} \exp\left(\pm \frac{i\kappa a \mu}{2}\right) \exp\left(-\frac{\kappa^2 \mu^2}{8M\omega}\right).$$

One readily observes then that in Eq. (16)

$$|\langle J' J_z' n | \cos(\boldsymbol{\kappa} \cdot \mathbf{R}/2) | J J_z n = 0 \rangle|^2 = \left(\frac{\kappa^2}{4M\omega}\right)^n \frac{1}{n!} \left| \left\langle J' J_z' \left| \mu^n \exp\left(-\frac{\kappa^2 \mu^2}{8M\omega}\right) \times \left(\frac{e^{i\kappa a \mu/2} + (-)^n e^{-i\kappa a \mu/2}}{2}\right) \right| J J_z \right\rangle \right|^2 = \left(\frac{\kappa^2}{4M\omega}\right)^n \frac{1}{n!} \left| \left\langle J' J_z' \left| \mu^n \exp\left(-\frac{\kappa^2 \mu^2}{8M\omega}\right) \times e^{i\kappa a \mu/2} \right| J J_z \right\rangle \right|^2, \quad (21)$$

since J' and J have the same parity. A similar expression holds if J' and J have opposite parity, so that

Eqs. (16) and (20) become

$$g(t)_{S=S'} = 4 \left(a_{\text{coh}}^2 + \frac{a_{\text{inc}}^2}{3} S(S+1) \right) \sum_J \frac{P_{JS}}{2J+1} \\ \times \sum_{J'}' e^{i(E_{J'} - E_J)t} \sum_n e^{in\omega t} \left(\frac{\kappa^2}{4M\omega} \right)^n \frac{1}{n!} \\ \times \sum_{J_z J_z'} \left| \left\langle J' J_z' \left| \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} \right. \right. \right. \right. \\ \left. \left. \left. + \frac{i\kappa a \mu}{2} \right) \right| J J_z \right\rangle \right|^2, \quad (22)$$

$$g(t)_{S \neq S'} = 4 a_{\text{inc}}^2 \left(1 - \frac{S(S+1)}{3} \right) \sum_J \frac{P_{JS}}{2J+1} \\ \times \sum_{J'}'' \exp[i(E_{J'} - E_J)t] \sum_{n=0} e^{in\omega t} \\ \times \left(\frac{\kappa^2}{4M\omega} \right)^n \frac{1}{n!} \sum_{J_z J_z'} \left| \left\langle J' J_z' \left| \mu^n \right. \right. \right. \\ \left. \left. \left. \times \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + \frac{i\kappa a \mu}{2} \right) \right| J J_z \right\rangle \right|^2, \quad (23)$$

Eqs. (22) and (23) are expected to be valid for n up to a value of 5, for above this value the H-H bond must begin to show its nonlinear character, and the harmonic approximation should fail.

II.3 Rotations

Since $E_J = (0.015/2)J(J+1)$, at liquid-hydrogen temperature, $kT \ll 0.015$ and all the molecules are in their lowest rotational state, which is $J=0$ for para-hydrogen and $J=1$ for orthohydrogen. At higher temperatures, however, we must know the transition probability between two arbitrary rotational states.

The wave functions for the rotational states of the linear H_2 molecule (if we assume no rotational-vibration coupling, and that there is no hindrance to the rotation) are just the spherical harmonics

$$|JJ_z\rangle = Y_{JJ_z}(\theta, \phi).$$

We choose a coordinate system with κ along the z axis. The rotational matrix elements then are of the form

$$\left\langle J' J_z' \left| \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i \frac{\kappa a \mu}{2} \right) \right| J J_z \right\rangle \\ = \int d\Omega \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i \frac{\kappa a \mu}{2} \right) Y_{J' J_z'}^*(\theta, \phi) \\ \times Y_{J J_z}(\theta, \phi). \quad (24)$$

If now we make use of the coupling theorem⁷ for the

⁷ M. E. Rose, *Elementary Theory of Angular Momentum* (John Wiley & Sons, Inc., New York, 1957).

spherical harmonics, namely,

$$Y_{l_1 m_1}(\theta, \phi) Y_{l_2 m_2}(\theta, \phi) = \sum_{l=|l_1-l_2|}^{l_1+l_2} \left[\frac{(2l_1+1)(2l_2+1)}{4\pi(2l+1)} \right]^{1/2} \\ \times C(l_1 l_2 l; m_1 m_2) C(l_1 l_2 l; 00) Y_{l, m_1+m_2}(\theta, \phi),$$

where the $C(l_1 l_2 l; m_1 m_2)$ are the Clebsch-Gordan coefficients which vanish (for $m_1 = m_2 = 0$) unless $l_1 + l_2 + l$ is even, and notice that J_z' must equal J_z , Eq. (24) can be written as

$$\left\langle J' J_z' \left| \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i \frac{\kappa a \mu}{2} \right) \right| J J_z \right\rangle \\ = \delta_{J_z' J_z} (-)^{J_z'} \sum_{l=|J'-J|}^{J'+J} \left[\frac{(2J'+1)(2J+1)}{4\pi(2l+1)} \right]^{1/2} \\ \times C(J J' l; J_z - J_z') C(J J' l; 00) \\ \cdot \int d\Omega \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i \frac{\kappa a \mu}{2} \right) Y_{l, J_z - J_z'}(\theta, \phi).$$

Thus,

$$\sum_{J_z' J_z} \left| \left\langle J' J_z' \left| \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i \frac{\kappa a \mu}{2} \right) \right| J J_z \right\rangle \right|^2 \\ = \frac{(2J'+1)(2J+1)}{4} \sum_{l=|J'-J|}^{J'+J} |A_{nl}|^2 C^2(J J' l; 00), \quad (25)$$

where we have used the closure relation for the Clebsch-Gordan coefficients:

$$\sum_{J_z} C(J J' l'; J_z - J_z) C(J J' l; J_z - J_z) = \delta_{l'l'}.$$

The A_{nl} are defined by

$$A_{nl} = \int_{-1}^1 d\mu \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + \frac{i\kappa a \mu}{2} \right) P_l(\mu),$$

and $P_l(\mu)$ is the Legendre polynomial of order l .

Using Eq. (25), Eqs. (22) and (23) become

$$g(t)_{S=S'} = [a_{\text{coh}}^2 + (a_{\text{inc}}^2/3)S(S+1)] \sum_J P_{JS} \\ \times \sum_{J'}' \exp[i(E_{J'} - E_J)t] (2J'+1) \\ \cdot \sum_{n=0} e^{in\omega t} \left(\frac{\kappa^2}{4M\omega} \right)^n \frac{1}{n!} \sum_{l=|J'-J|}^{J'+J} |A_{nl}|^2 \\ \times C^2(J J' l; 00), \quad (26)$$

$$g(t)_{S \neq S'} = a_{\text{inc}}^2 \left(1 - \frac{S(S+1)}{3} \right) \sum_J P_{JS} \\ \times \sum_{J'}'' \exp[i(E_{J'} - E_J)t] (2J'+1) \\ \cdot \sum_{n=0} e^{in\omega t} \left(\frac{\kappa^2}{4M\omega} \right)^n \frac{1}{n!} \sum_{l=|J'-J|}^{J'+J} |A_{nl}|^2 \\ \times C^2(J J' l; 00). \quad (27)$$

The coefficients $C^2(JJ'l; 00)$ are given in Appendix B for $J=0, 1, 2, 3, 4$.

II.4 Translations

For hydrogen we assume that the molecular translations are free. Using familiar results, the translational part of Eq. (3) becomes

$$\langle \psi_{it} | \exp[-i\mathbf{k} \cdot \mathbf{r}_i(0)] \exp[i\mathbf{k} \cdot \mathbf{r}_i(t)] | \psi_{it} \rangle_T = \exp[-(t\kappa^2/4M)(-i+tT)], \quad (28)$$

where the temperature T is measured in electron volts.

This approximation is likely to be an excellent one for gaseous hydrogen.

III. SCATTERING CROSS SECTIONS

By using Eqs. (26), (27), and (28) in Eq. (3), and performing the indicated integration over t , the "self" scattering cross section per molecule from parahydrogen becomes

$$\begin{aligned} \left. \frac{d^2\sigma_s}{d\Omega d\epsilon} \right|_{\text{para}} &= \frac{k}{k_0} \left(\frac{M}{\pi\kappa^2 T} \right)^{1/2} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega} \right)^n \sum_{J=0,2,4,\dots} P_J \\ &\times [a_c^2 \sum_{J'=0,2,4,\dots} + a_i^2 \sum_{J'=1,3,5,\dots} (2J'+1)] \\ &\times \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M} \right) / \left(\frac{\kappa^2 T}{M} \right) \right\} \\ &\times \sum_{l=|J'-J|}^{J'+J} C^2(JJ'l; 00) |A_{nl}|^2, \quad (29) \end{aligned}$$

where $\Delta E = E_{J'} - E_J + n\omega$, and for orthohydrogen

$$\begin{aligned} \left. \frac{d^2\sigma_s}{d\Omega d\epsilon} \right|_{\text{ortho}} &= \frac{k}{3k_0} \left(\frac{M}{\pi\kappa^2 T} \right)^{1/2} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega} \right)^n \sum_{J=1,3,5,\dots} P_J \\ &\times [a_i^2 \sum_{J'=0,2,4,\dots} + (3a_c^2 + 2a_i^2) \sum_{J'=1,3,5,\dots}] \\ &\times (2J'+1) \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M} \right) / \left(\frac{\kappa^2 T}{M} \right) \right\} \\ &\times \sum_{l=|J'-J|}^{J'+J} C^2(JJ'l; 00) |A_{nl}|^2, \quad (30) \end{aligned}$$

where for compactness in writing we treat the summation sign as an operator. Eqs. (29) and (30) have special cases of interest. If the initial neutron energy is sufficiently less than 0.546 eV so that we satisfy the condition $\kappa^2/8M\omega \ll 1$ for all scattering angles, then the vibrations need not be considered, and

$$A_{0l} = \int_{-1}^1 d\mu e^{i\kappa a \mu/2} P_l(\mu) = 2i^l j_l \left(\frac{\kappa a}{2} \right),$$

where j_l is the spherical Bessel function of order l .

Thus, in this limit, Eqs. (29) and (30) become

$$\begin{aligned} \left. \frac{d^2\sigma_s}{d\Omega d\epsilon} \right|_{\text{para}} &= 4 \frac{k}{k_0} \left(\frac{M}{\pi\kappa^2 T} \right)^{1/2} \sum_{J=0,2,4,\dots} P_J \\ &\times [a_c^2 \sum_{J'=0,2,4,\dots} + a_i^2 \sum_{J'=1,3,5,\dots} (2J'+1)] \\ &\times \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M} \right) / \left(\frac{\kappa^2 T}{M} \right) \right\} \\ &\times \sum_{l=|J'-J|}^{J'+J} j_l^2 \left(\frac{\kappa a}{2} \right) C^2(JJ'l; 00), \quad (31) \end{aligned}$$

$$\begin{aligned} \left. \frac{d^2\sigma_s}{d\Omega d\epsilon} \right|_{\text{ortho}} &= \frac{4}{3} \frac{k}{k_0} \left(\frac{M}{\pi\kappa^2 T} \right)^{1/2} \sum_{J=1,3,5,\dots} P_J \\ &\times [a_i^2 \sum_{J'=0,2,4,\dots} + (3a_c^2 + 2a_i^2) \sum_{J'=1,3,5,\dots}] \\ &\times (2J'+1) \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M} \right) / \left(\frac{\kappa^2 T}{M} \right) \right\} \\ &\times \sum_{l=|J'-J|}^{J+J'} j_l^2 \left(\frac{\kappa a}{2} \right) C^2(JJ'l; 00). \quad (32) \end{aligned}$$

Another special case is that of low temperature, where the molecules are in their ground rotational as well as vibrational states. Here we find

$$\begin{aligned} \left. \left(\frac{d^2\sigma_s}{d\Omega d\epsilon} \right) \right|_{\text{para}} &= \frac{k}{k_0} \left(\frac{M}{\pi\kappa^2 T} \right)^{1/2} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega} \right)^n \\ &\cdot [a_c^2 \sum_{J'=0,2,4,\dots} + a_i^2 \sum_{J'=1,3,5,\dots} (2J'+1)] \\ &\times \exp \left\{ - \left(\epsilon + E_{J'} + n\omega - \frac{\kappa^2}{4M} \right) / \left(\frac{\kappa^2 T}{M} \right) \right\} \\ &\times |A_{n,J'}|^2, \quad (33) \end{aligned}$$

and for orthohydrogen,

$$\begin{aligned} \left. \left(\frac{d^2\sigma_s}{d\Omega d\epsilon} \right) \right|_{\text{ortho}} &= \frac{k}{3k_0} \left(\frac{M}{\pi\kappa^2 T} \right)^{1/2} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega} \right)^n \\ &\times [a_i^2 \sum_{J'=0,2,4,\dots} + (3a_c^2 + 2a_i^2) \sum_{J'=1,3,5,\dots}] \\ &\times \exp \left\{ - \left(\epsilon + E_{J'} - E_1 + n\omega - \frac{\kappa^2}{4M} \right) / \left(\frac{\kappa^2 T}{M} \right) \right\} \\ &\times ((J'+1) |A_{n,J'+1}|^2 + J' |A_{n,J'-1}|^2). \quad (34) \end{aligned}$$

At room temperature one might ask if the inclusion of spin correlations is important. The answer seems to be affirmative, since, as an investigation of P_J readily shows, at this temperature only the rotational states $J=0, 1, 2, 3$ are present in any appreciable amount.

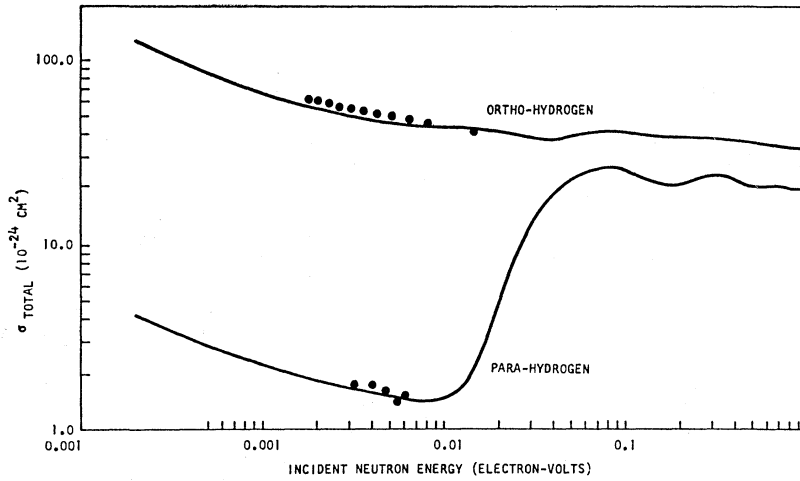


FIG. 1. Calculated total cross sections for parahydrogen and orthohydrogen gas at $T = 20.4^\circ \text{K}$ compared to the experimental results of Squires and Stewart (Ref. 10).

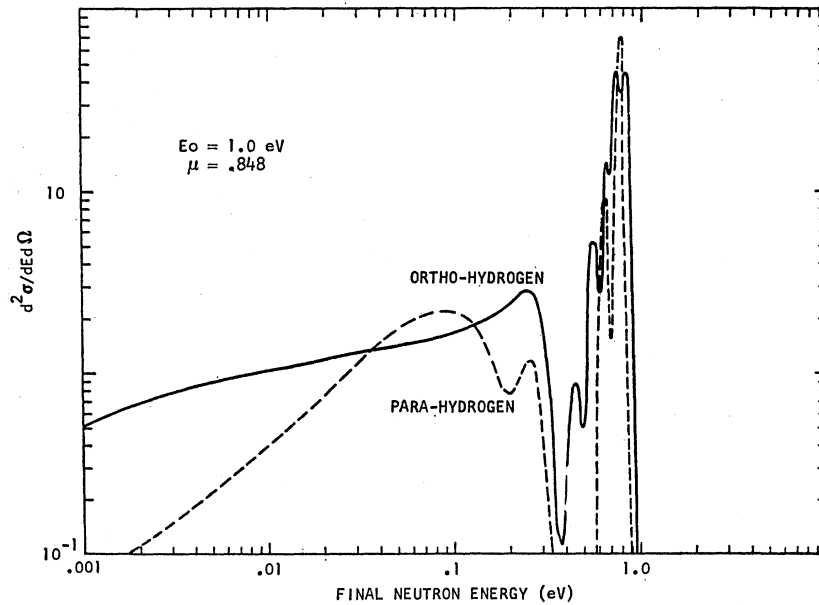


FIG. 2. The double differential cross section $d^2\sigma/dE d\Omega$, for an initial neutron energy of 1 eV and a scattering angle of 32° for both parahydrogen and orthohydrogen at $T = 20.4^\circ \text{K}$.

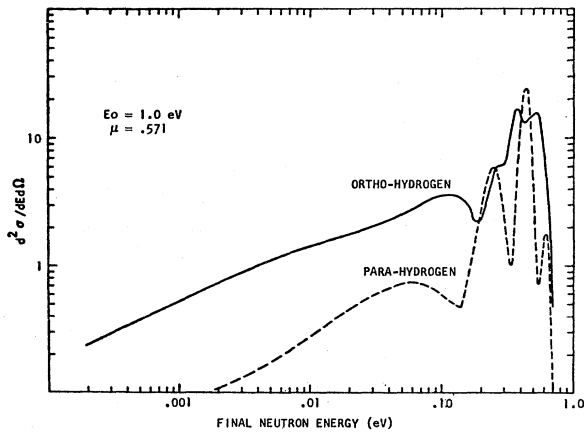


FIG. 3. The double differential cross section $d^2\sigma/dE d\Omega$, for an initial neutron energy of 1 eV and a scattering angle of 55° for both parahydrogen and orthohydrogen at $T = 20.4^\circ \text{K}$.

The cross section for scattering from an ortho-para mixture is given by

$$\frac{d^2\sigma_s}{d\Omega d\epsilon} = \frac{N_{\text{para}}}{N_{\text{para}} + N_{\text{ortho}}} \left. \frac{d^2\sigma_s}{d\Omega d\epsilon} \right|_{\text{para}} + \frac{N_{\text{ortho}}}{N_{\text{para}} + N_{\text{ortho}}} \left. \frac{d^2\sigma_s}{d\Omega d\epsilon} \right|_{\text{ortho}},$$

where N_{para} and N_{ortho} are the number densities of para and ortho molecules, respectively. For $kT \gg 0.015$, $N_{\text{para}}/N_{\text{ortho}} = \frac{1}{3}$, and at room temperature (0.0258 eV), $N_{\text{para}}/N_{\text{ortho}} = 1/2.91$.

While all the above results have been essentially exact for gaseous hydrogen, it would be desirable to apply them to the case of liquid hydrogen. The Debye temperature of liquid hydrogen has been calculated⁸ to be 0.007 eV, and thus for neutron energies higher

⁸ J. Schwinger and E. Teller, Phys. Rev. **51**, 775 (1937); **52**, 286 (1937).

than this the results of this paper should be applicable. An additional supporting fact is that even the lowest rotational level has an energy twice that of the Debye temperature.

IV. INTERFERENCE EFFECTS

So far our calculations have been for the "self" scattering, i.e., referring to a single H_2 molecule; and for gaseous hydrogen the influence of the interference of neutron waves scattered from different molecules will be very small. For liquid hydrogen, however, interference effects might be of importance for very low neutron energies where para \rightarrow para transitions in Eq. (33) are the only competition, i.e., for neutron energies below 0.015 eV. Even for these low energies, the self-scattering para \rightarrow para cross section will dominate. Interference effects for liquid hydrogen in terms of the translational correlation function for neutron energies below the first vibrational level were calculated by Sarma,⁴ but for completeness we quote the formula here with the inclusion of the vibrations

$$\frac{d^2\sigma_{\text{int}}}{d\Omega d\epsilon} = \frac{a_c^2 k}{2\pi k_0} \int_{-\infty}^{\infty} dt e^{i\epsilon t} \times \sum_{l \neq j} \langle \psi_{il} | \exp[-i\mathbf{k} \cdot \mathbf{r}_j(0)] \exp[i\mathbf{k} \cdot \mathbf{r}_l(t)] | \psi_{il} \rangle_T \cdot \left[\int_{-1}^1 d\mu \cos\left(\frac{\kappa a \mu}{2}\right) e^{-\kappa^2 \mu^2 / 8M\omega} \right]^2. \quad (35)$$

For a free gas, the translational correlation function

$$\sum_{l \neq j} \langle \psi_{il} | \exp[-i\mathbf{k} \cdot \mathbf{r}_j(0)] \exp[+i\mathbf{k} \cdot \mathbf{r}_l(t)] | \psi_{il} \rangle_T$$

vanishes.

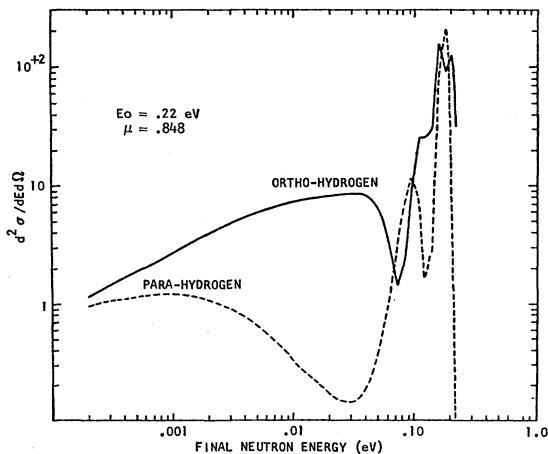


FIG. 4. The double differential cross section $d^2\sigma/dE d\Omega$, for an initial neutron energy of 0.22 eV and a scattering angle of 32° for both parahydrogen and orthohydrogen at $T=20.4^\circ\text{K}$.

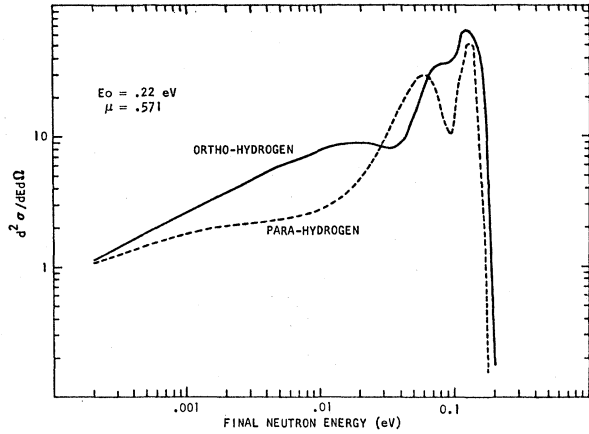


FIG. 5. The double differential cross section $d^2\sigma/dE d\Omega$, for an initial neutron energy of 0.22 eV and a scattering angle of 55° for both parahydrogen and orthohydrogen at $T=20.4^\circ\text{K}$.

If, for neutron energies $\lesssim 0.015$ eV so that the first rotational transition cannot be made, the results of the previous section are added to Eq. (35), we obtain for parahydrogen

$$\frac{d^2\sigma}{d\Omega d\epsilon} = \frac{2a_c^2 k}{\pi k_0} j_0^2\left(\frac{\kappa a}{2}\right) \int_{-\infty}^{\infty} dt e^{i\epsilon t} \times \sum_{l,j} \langle \psi_{il} | \exp[-i\mathbf{k} \cdot \mathbf{r}_j(0)] \exp[i\mathbf{k} \cdot \mathbf{r}_l(t)] | \psi_{il} \rangle_T, \quad (36)$$

where $\sigma = \sigma_s + \sigma_{\text{int}}$. Equation (36) has precisely the form for a purely coherent scatterer,⁹ and thus measurements of the double differential scattering cross section will give directly the double Fourier transform of the total translational correlation function and hence information concerning the intermolecular correlations in liquid parahydrogen. It is also interesting to note that for neutron energies $\gtrsim 0.015$ eV, parahydrogen becomes an incoherent scatterer (since $a_c^2/a_i^2 \ll 1$), and interference effects become negligible. Liquid hydrogen is 99.79% parahydrogen for equilibrium conditions at the boiling temperature (20.4°K).

V. NUMERICAL RESULTS AND DISCUSSION

Equations (29) and (30) have been programmed for computation on the IBM-7044. In particular, Fig. 1 shows the total cross section (per atom) for scattering from both orthohydrogen and parahydrogen gas at 20.4°K for initial neutron energies up to 1 eV. Notice that the parahydrogen cross section rises rapidly as E_0 approaches 0.022 eV, which is just the energy the neutron needs in the laboratory frame to cause the rotational transition $J=0 \rightarrow J=1$, from parahydrogen to orthohydrogen. Also shown in Fig. 1 are some experi-

⁹ L. Van Hove, Phys. Rev. **95**, 249 (1954).

mental points by G. Squires and A. Stewart.¹⁰ In Figs. 2-5 we show the computed double differential cross section $d^2\sigma/dE d\Omega$ for two different initial energies and scattering angles. For $E_0=1.0$ eV and $\theta=32^\circ$ one can clearly recognize the structure due to the rotational transitions superimposed on the zero- and one-phonon contributions to the cross section. The structure is more pronounced for parahydrogen than for orthohydrogen, since in the case of parahydrogen only the odd rotational levels make a significant contribution, whereas all levels contribute to the ortho cross section. The odd levels in orthohydrogen contribute roughly twice as much as the even levels as seen in Eq. (30). At a scattering angle of 55° the structure due to the rotational transitions is still present, but appears somewhat smoothed out because of the larger recoil energy. The same comments apply to the curves corresponding to $E_0=0.22$, although here only the zero-phonon term can contribute to the cross sections.

In summary then, the results given in this paper predict the scattering cross sections from H_2 and D_2 for incident neutron energies up to roughly 3 eV and all practical temperatures. For neutron energies above a few volts the calculated total cross sections for both orthohydrogen and parahydrogen tend to the free-atom limit as they should; and for low-neutron energies, so that only the first rotational transition occurs, the results are identical to those of Sarma.⁴ The calculated total cross section agrees reasonably well with the available experimental data, and as the experimental errors decrease with increasing neutron energy so does the agreement between theory and experiment improve. In Sec. IV a brief discussion of interference effects was given and the point made that for neutron energies low enough so that the rotational transitions cannot be excited, a measurement of the double differential cross section in liquid parahydrogen is directly proportional to the Fourier transform of the total Van Hove correlation function, and hence will give useful information concerning the intermolecular correlations. Also the smallness of the Debye temperature (0.007 eV) indicates that the results of this paper will be applicable to liquid hydrogen for all but very low incident neutron energies. Interference effects will always be negligible for neutron energies above 0.015 eV in parahydrogen.

Since the differential cross sections are far more sensitive to the details of the model than the total cross section, it is hoped that differential experiments planned and in progress¹¹ for various ranges of neutron energy will allow a rigorous test of the theory.

ACKNOWLEDGMENT

The authors would like to thank D. Y. Naliboff for his assistance with the numerical work.

¹⁰ G. L. Squires and A. T. Stewart, Proc. Roy. Soc. (London) **A230**, 19 (1955).

¹¹ H. Palevsky (private communication).

APPENDIX A: D_2 MOLECULE

Aside from the trivial difference in masses, the only difference between the H_2 and the D_2 molecule lies in the spin correlation. In this appendix we calculate the cross sections for the scattering of neutrons from D_2 by evaluating the effect of the spin correlations, and using the H_2 results already obtained for the vibrations, rotations, and translations.

For D_2 we find (with the same notation as in Sec.II)

$$\begin{aligned} A &= a_1 \exp(i\boldsymbol{\kappa}\cdot\mathbf{R}/2) + a_2 \exp(-i\boldsymbol{\kappa}\cdot\mathbf{R}/2) \\ &= P \cos(\boldsymbol{\kappa}\cdot\mathbf{R}/2) + iQ \sin(\boldsymbol{\kappa}\cdot\mathbf{R}/2), \end{aligned} \quad (A1)$$

where

$$P = (2a_{\text{coh}} + \sqrt{2}a_{\text{inc}}\mathbf{S}\cdot\boldsymbol{\sigma}), \quad (A2)$$

$$Q = \sqrt{2}a_{\text{inc}}\boldsymbol{\sigma}\cdot(\mathbf{I}_1 - \mathbf{I}_2), \quad (A3)$$

and $\mathbf{S} = \mathbf{I}_1 + \mathbf{I}_2$ is the total spin of the molecule. Since the deuteron has spin 1, the symmetric ("ortho") nuclear spin eigenfunctions of the D_2 molecule are those of spin 0 and 2, and these correspond to even values of J . The antisymmetric ("para") states have spin 1 and odd J values. It can readily be shown that the total spin operator \mathbf{S} has nonzero matrix elements only between states of the same total \mathbf{S} [$\mathbf{S}^2 = S(S+1)$] and that the operator Q has nonzero matrix elements only between states of total S differing by 1. It follows from this, and also from the conservation of spin angular momentum ($\Delta S = \pm 1, 0$), that the transitions $S=0 \leftrightarrow S=2$ cannot occur.

Using Eqs. (11), (12), (14), (A1), we find in this case

$$\begin{aligned} g(t)_{S=S'} &= \frac{1}{2}[8a_{\text{coh}}^2 + S(S+1)a_{\text{inc}}^2] \\ &\times \sum_J \frac{P_{JS}}{2J+1} \sum_{J'} \exp[i(E_{J'} - E_J)t] \sum_{n=0} e^{in\omega t} \\ &\times \sum_{J_z' J_z} |\langle J' J_z', n | \cos(\boldsymbol{\kappa}\cdot\mathbf{R}/2) | J J_z, n=0 \rangle|^2, \end{aligned}$$

where $\sum_{J'}$ implies that J' and J have the same parity, and for J', J both even, S is 0 or 2, while for J', J both odd S is 1.

Similarly, Eqs. (17), (19), and (A3) result in

$$\begin{aligned} g(t)_{S \neq S'} &= (a_{\text{inc}}^2/2)[8 - S(S+1)] \\ &\times \sum_J \frac{P_{JS}}{2J+1} \sum_{J'} \exp[i(E_{J'} - E_J)t] \sum_n e^{in\omega t} \\ &\times \sum_{J_z J_z'} |\langle J' J_z', n | \sin(\boldsymbol{\kappa}\cdot\mathbf{R}/2) | J J_z, n=0 \rangle|^2, \end{aligned}$$

where $\sum_{J'}$ implies that J' and J have opposite parity, and for J even, S is 0 or 2, while for J odd, S is 1.

The above results now give, in complete analogy with the way Eqs. (29) and (30) were obtained, the

“self” cross section for orthodeuterium:

$$\begin{aligned} \frac{d^2\sigma_s}{d\Omega d\epsilon} \Big|_{\text{ortho}} &= \frac{k}{k_0} \left(\frac{M_d}{\pi\kappa^2 T} \right)^{1/2} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M_d\omega} \right)^n \sum_{J=0,2,4,\dots} P_J \\ &\times \left[(a_{\text{coh}}^2 + \frac{5}{8}a_{\text{inc}}^2) \sum_{J'=0,2,4,\dots} + \frac{3}{8}a_{\text{inc}}^2 \sum_{J'=1,3,5,\dots} \right] \\ &\times (2J'+1) \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M_d} \right)^2 / \left(\frac{\kappa^2 T}{M_d} \right) \right\} \\ &\times \sum_{l=|J'-J|}^{J'+J} C^2(JJ'l; 00) |A_{nl}|^2. \quad (\text{A4}) \end{aligned}$$

For paradeuterium, we find

$$\begin{aligned} \frac{d^2\sigma_s}{d\Omega d\epsilon} \Big|_{\text{para}} &= \frac{k}{4k_0} \left(\frac{M_d}{\pi\kappa^2 T} \right)^{1/2} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M_d\omega} \right)^n \sum_{J=1,3,5,\dots} P_J \\ &\times \left[3a_{\text{inc}}^2 \sum_{J'=0,2,4,\dots} + (4a_{\text{coh}}^2 + a_{\text{inc}}^2) \sum_{J'=1,3,5,\dots} \right] \\ &\times (2J'+1) \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M_d} \right)^2 / \left(\frac{\kappa^2 T}{M_d} \right) \right\} \\ &\times \sum_{l=|J'-J|}^{J'+J} C^2(JJ'l; 00) |A_{nl}|^2, \quad (\text{A5}) \end{aligned}$$

where M_d is the deuteron mass. The energy levels for D_2 are given by

$$E_J = \hbar^2 J(J+1) / 4M_d a^2 = 0.0071 [J(J+1)/2] \text{ eV},$$

and thus for low temperatures ($\lesssim 0.02$ eV) spin correlations will be important for deuterium.

APPENDIX B

Here the values of the Clebsch-Gordon coefficients $C^2(JJ'l; 00)$ are given for $J=0, 1, 2, 3, 4$. The analytic expression for $C(JJ'l; 00)$ is given in Ref. 7. For simplicity in notation, we define the quantity $(JJ'|l)$ so that

$$(JJ'|l) = (2J'+1)C^2(JJ'l; 0).$$

The results are:

$J=0$

$$(0J'|l) = 2J'+1,$$

$J=1$

$$(1J'|J'+1) = J'+1,$$

$$(1J'|J'-1) = J',$$

$J=2$

$$(2J'|J'+2) = \frac{3(J'+2)(J'+1)}{2(2J'+3)},$$

$$(2J'|J') = \frac{J'(J'+1)(2J'+1)}{(2J'+3)(2J'-1)},$$

$$(2J'|J'-2) = \frac{3J'(J'-1)}{2(2J'-1)},$$

$J=3$

$$(3J'|J'+3) = \frac{5(J'+3)(J'+2)(J'+1)}{2(2J'+5)(2J'+3)},$$

$$(3J'|J'+1) = \frac{3(J'+2)(J'+1)J'}{2(2J'+5)(2J'-1)},$$

$$(3J'|J'-1) = \frac{3(J'+1)J'(J'-1)}{2(2J'+3)(2J'-3)},$$

$$(3J'|J'-3) = \frac{5J'(J'-1)(J'-2)}{2(2J'-1)(2J'-3)},$$

$J=4$

$$(4J'|J'+4) = \frac{35(J'+4)(J'+3)(J'+2)(J'+1)}{8(2J'+7)(2J'+5)(2J'+3)},$$

$$(4J'|J'+2) = \frac{5(J'+3)(J'+2)(J'+1)J'}{2(2J'+7)(2J'+3)(2J'-1)},$$

$$(4J'|J') = \frac{9(2J'+1)(J'+2)(J'+1)(J'-1)J'}{4(2J'+5)(2J'+3)(2J'-1)(2J'-3)},$$

$$(4J'|J'-2) = \frac{5(J'+1)J'(J'-1)(J'-2)}{2(2J'+3)(2J'-1)(2J'-5)},$$

$$(4J'|J'-4) = \frac{35J'(J'-1)(J'-2)(J'-3)}{8(2J'-1)(2J'-3)(2J'-5)}.$$