

## Scattering of Slow Neutrons by Deuterated Methane\*

GEORGE W. GRIFFING

Atomic Energy Division, Phillips Petroleum Company, Idaho Falls, Idaho

(Received 15 June 1964)

The scattering of slow neutrons by gaseous deuterated methane has been computed. This case is of particular interest since contributions to the scattering from the direct scattering terms and the interference scattering terms can be comparable in magnitude. Plausible arguments are presented which suggest that an incident neutron energy of 0.1 eV is large enough so that experimental results on liquid  $\text{CD}_4$  using neutrons of that energy could be confidently compared with the calculated results of this paper for a gas, without the difficulties caused by liquid effects in interpreting the scattering at smaller incident-neutron energies.

### I. INTRODUCTION

ALTHOUGH precise formulas for the partial differential cross section for scattering of slow neutrons by molecules were presented by Zemach and Glauber<sup>1</sup> several years ago, there are few molecules for which detailed calculations have been performed that make use of these formulas. An exception is gaseous methane where extensive calculations have been performed<sup>2</sup> and the results compared with the experimental data.<sup>3</sup> In those studies it was found that it was necessary to take into account the discrete nature of the rotational levels of the molecule to obtain agreement with the experimental results. In addition, it was noted that interference scattering between different nuclei in a  $\text{CH}_4$  molecule, although small, influenced the scattering. To study interference scattering between different nuclei in a molecule, a more interesting case is gaseous deuterated methane because the scattering lengths of the deuteron are such that interference effects will be much more significant.

While the flux of neutrons from the Materials Testing Reactor was sufficient to perform scattering experiments on gaseous  $\text{CH}_4$ , it is by no means certain that this would also be possible for gaseous  $\text{CD}_4$  since the cross section of the deuteron is about an order of magnitude smaller than that of the proton. However, as will be explained later, it appears that for neutron energies suitable for the study of interference effects, the cross section for the scattering of neutrons from liquid  $\text{CD}_4$  will approximate that which would be obtained by considering the liquid to be a gas at the temperature of the liquid. Thus, it should be possible to compare experimental results on liquid  $\text{CD}_4$  with the computed results on gaseous  $\text{CD}_4$  presented in this paper. In the following section we discuss scattering of neutrons by

liquid  $\text{CH}_4$  which is intended to help make the above remarks plausible.

### II. SCATTERING BY LIQUID $\text{CH}_4$

If the incident-neutron energy is sufficiently larger than the binding energy of a molecule to the liquid, it would be expected that any influence of liquid effects of the scattering of neutrons would be small. Under such circumstances it should be permissible to compare the experimental results with computations which assume the liquid can be treated as a gas which is at the temperature of the liquid. This is analogous to scattering neutrons from free protons where, in fact, the neutrons are scattered from protons which are bound in some given homogeneous system. However, lacking a fundamental theory of liquids, it is not *a priori* obvious what incident energy is required in order to justify treating the liquid as a gas.

We can get an estimate on the lower bound of the required incident neutron energy from the heat of vaporization. Knowing that the heat of vaporization<sup>4</sup> of liquid  $\text{CH}_4$  is 2036 cal/mole, the binding energy of a  $\text{CH}_4$  molecule to the liquid is determined to be approximately 0.08 eV. Thus it would be expected the incident-neutron energy would need to be greater than 0.08 eV in order that the scattering by liquid  $\text{CH}_4$  would approximate scattering by gaseous  $\text{CH}_4$ . Some experimental results obtained at the Materials Testing Reactor on liquid  $\text{CH}_4$  will now be mentioned which gave a bearing on this approximation.

Experiments<sup>5</sup> on liquid  $\text{CH}_4$  have been conducted at incident neutron energies of 0.1, 0.07, and 0.025 eV. The temperature of the liquid was approximately 100°K and the scattering angle ranged from 15° to 150°. Necessarily there is some question on the absolute magnitude of the experimentally observed partial differential cross sections due to target thickness, counter efficiencies, and various other effects. Let us suppose that the observed partial differential cross section for liquid  $\text{CH}_4$  is normalized at some given wave length to the value of the computed partial differential cross section for gaseous  $\text{CH}_4$  at the temperature of the liquid and the shapes

\* This work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup> A. C. Zemach and R. J. Glauber, Phys. Rev. **101**, 118 (1956); **101**, 129 (1956).

<sup>2</sup> G. W. Griffing, Phys. Rev. **124**, 1489 (1961); **127**, 1179 (1962); *Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1963), Vol. I. In the last two references there is a misprint in the expressions for  $\sigma_{pp}$  and  $\sigma_{cp}$ . As given they are too large by a factor of 2.

<sup>3</sup> P. D. Randolph, R. M. Brugger, K. A. Strong, and R. E. Schmunk, Phys. Rev. **124**, 460 (1961).

<sup>4</sup> A. Frank and K. Clausius, Z. Phys. Chem. **B42**, 395 (1939).

<sup>5</sup> Y. Harker (private communication).

of the curves compared. At an incident neutron energy of 0.1 eV, it was observed that there was agreement at all scattering angles. For 0.07 eV, agreement was satisfactory for scattering angles greater than about  $30^\circ$ . Finally at 0.025 eV, there was poor agreement between the observed and computed curves at all scattering angles. On the basis of these results, we conclude that a neutron energy of 0.1 eV is sufficiently large enough for the computed scattering from gaseous  $\text{CH}_4$  to be a good approximation to the observed scattering from liquid  $\text{CH}_4$ . An example of this agreement is shown in Fig. 1.

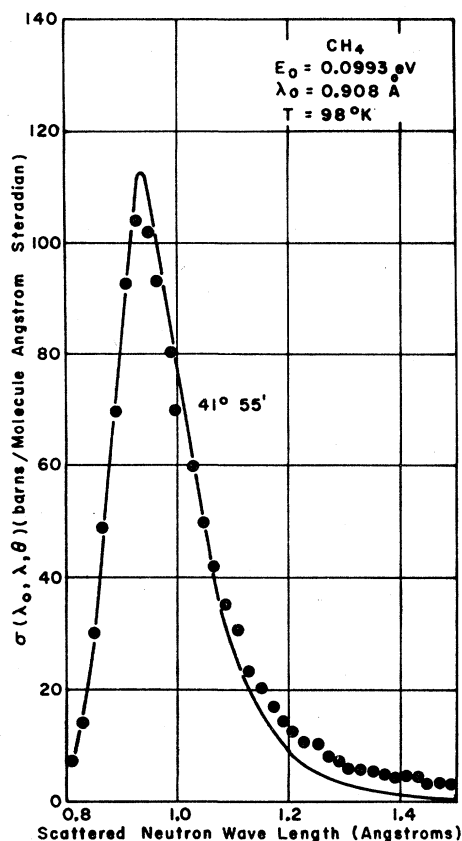


Fig. 1. Computed partial-differential cross section for gaseous  $\text{CH}_4$  shown as a function of the outgoing-neutron wave length. The experimental data for liquid  $\text{CH}_4$  are shown as solid circles and have been normalized to the calculations at  $\lambda = 0.85 \text{ \AA}$ .

Partial support for the above conclusion is given by the experiments<sup>6</sup> of Rogalska. He measured the total cross section of liquid  $\text{CH}_4$  with neutrons ranging in energy from 0.006 to 0.1 eV. Good agreement was found between the measurements and calculations for gaseous  $\text{CH}_4$  at the temperature of the liquid throughout this energy range. However, the agreement found by Rogalska to such small energies appears to us to be

<sup>6</sup> Z. Rogalska, *Physica* 29, 491 (1963); Also W. L. Whittemore, *Nucl. Sci. Eng.* 18, 182 (1964).

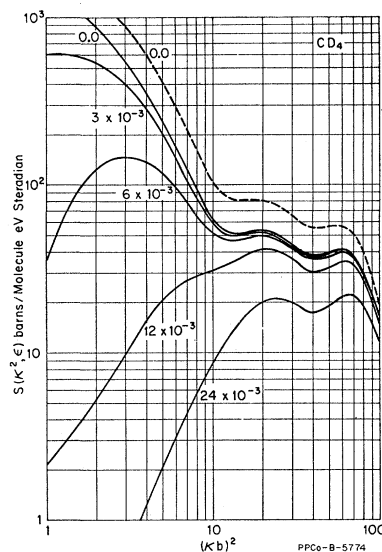


Fig. 2.  $S(k^2, \epsilon)$  shown as a function of  $(kb)^2$  for selected energy transfers. The solid line curves are for a gas temperature of  $294^\circ\text{K}$  while the broken line curve is for a temperature of  $100^\circ\text{K}$ .

fortuitous, and indicates that the total cross section is not particularly sensitive to the influence of liquid effects.

The heat of vaporization<sup>7</sup> of liquid  $\text{CD}_4$  has been measured as 2025 cal/mole. Thus the binding energy of a  $\text{CD}_4$  molecule to the liquid is approximately the same as that for a  $\text{CH}_4$  molecule. From the above discussion on liquid  $\text{CH}_4$ , it appears plausible that all of the interesting features resulting from interference contributions presented in this paper on gaseous  $\text{CD}_4$  could be compared with experimental observations on neu-

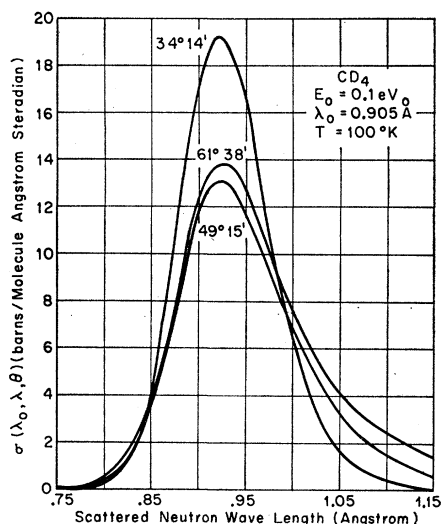


Fig. 3. Computed partial-differential cross sections shown as a function of the outgoing-neutron wave length for selected scattering angles.

<sup>7</sup> J. H. Colwell, E. K. Gill, and J. A. Morrison, *J. Chem. Phys.* 39, 635 (1963). In a note attached to the reprint, the authors mentioned that their measured heat of vaporization may be low by about 2%. This is of no consequence in our arguments.

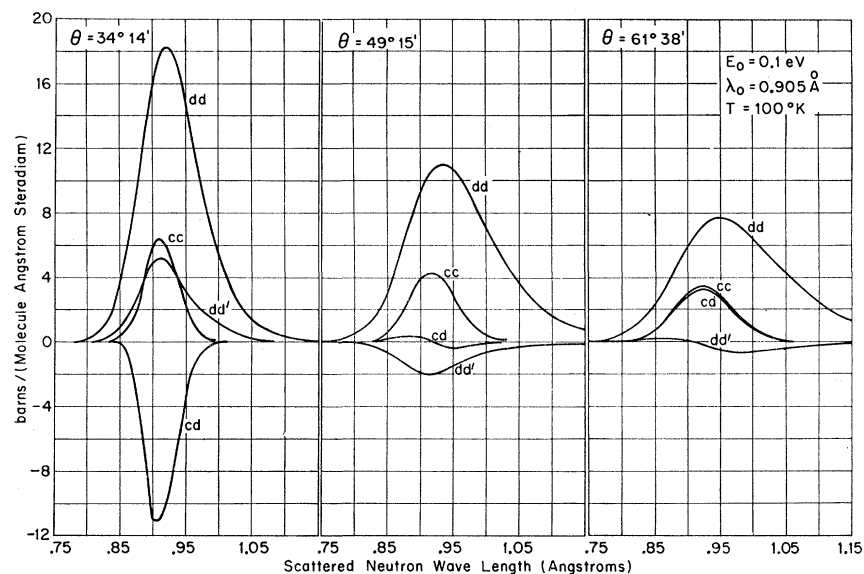


FIG. 4. Contributing terms which determine the partial-differential cross section in Fig. 3 for the selected scattering angles. The symbol  $dd$  is for  $4\sigma_{dd}(E_0, \epsilon, \theta)$ . By reference to Eq. (1), the remaining symbols are evident.

trons with an incident energy of 0.1 eV scattered from liquid  $CD_4$ .

### III. SCATTERING BY $CD_4$

Since  $CD_4$  has the same molecular structure as  $CH_4$ , the theory as discussed in Ref. 2 is applicable. The values for the parameters used in the computations are given in the Appendix. To help clarify the discussion, we mention that the partial differential cross section of  $CD_4$  at a temperature  $T$  for an incident energy  $E_0$ , energy transfer  $\epsilon$ , and a scattering angle  $\theta$  is given by

$$\sigma(E_0, \epsilon, \theta) = 4\sigma_{dd}(E_0, \epsilon, \theta) + 12\sigma_{dd'}(E_0, \epsilon, \theta) + 8\sigma_{cd}(E_0, \epsilon, \theta) + \sigma_{cc}(E_0, \epsilon, \theta). \quad (1)$$

In this equation,  $d$  denotes a deuteron,  $d'$  a different deuteron in the same molecule and  $C$  the carbon nucleus. Analytical expressions for these quantities are given in Ref. 2. Multiplying (1) by  $(k_0/k) \exp(\frac{1}{2}\epsilon/T)$  defines the function

$$S(\kappa^2, \epsilon) = S_{dd}(\kappa^2, \epsilon) + S_{dd'}(\kappa^2, \epsilon) + S_{cd}(\kappa^2, \epsilon) + S_{cc}(\kappa^2, \epsilon), \quad (2)$$

where

$$S_{dd}(\kappa^2, \epsilon) = 4(k_0/k) \exp(\frac{1}{2}\epsilon/T) \sigma_{dd}(E_0, \epsilon, \theta)$$

and the remaining terms are similarly defined. The initial and final neutron momenta are denoted by  $\mathbf{k}_0$  and  $\mathbf{k}$ , and  $\kappa$  is the momentum transfer. By introducing  $S(\kappa^2, \epsilon)$ , the results can be presented in a more compact manner.

In Fig. 2,  $S(\kappa^2, \epsilon)$  for gaseous  $CD_4$  is plotted as a function of  $(\kappa b)^2$  for a few selected values of the energy transfer  $\epsilon$ . The distance from the carbon nucleus to the deuteron is denoted by  $b$ . For a gas temperature of 294°K the energy transfer  $\epsilon$  ranges from 0.0 to 0.024 eV, while for a temperature of 100°K only the curve for  $\epsilon=0.0$  is shown. Curves for other values of the

energy transfer  $\epsilon$  for 100°K will be similar in shape to the corresponding curves for 294°K. Confining our attention to the case of an energy transfer of  $\epsilon=0.0$ , it will be noted that there are minima at  $(\kappa b)^2$  equal to 15 and 40 with maxima at  $(\kappa b)^2$  equal to 20 and 60. This feature is similar to that observed in scattering neutrons from liquid sodium.<sup>8</sup> In that case the observed structure was due to contributions of interference effects to the scattering from a sodium atom by scattering from sodium atoms in the first and second coordinate shells. While a thorough discussion of the structure for  $CD_4$  would be complicated, it is perhaps worth pointing out that a qualitative understanding is possible by considering only the zero order Bessel function  $j_0(\kappa b)$  appearing in the terms contributing to  $S(\kappa^2, \epsilon)$ . For convenience we mention that  $j_0(\kappa b)$  has its first minimum at  $(\kappa b)^2 \approx 20$ , a value of zero at  $(\kappa b)^2 \approx 40$ , and a second maximum at  $(\kappa b)^2 \approx 60$ . These correspond to scattering angles of 34°14', 49°15' and 61°38', respectively, for  $\epsilon=0.0$  and  $E_0=0.1$  eV. The partial differential cross section  $\sigma(\lambda_0, \lambda, \theta)$  is plotted as a function of the scattered-neutron wave length  $\lambda$  with the gas at a temperature of 100°K for these scattering angles in Fig. 3. As would be expected from the results shown in Fig. 2, the quasielastic peak for a scattering angle of 49°15', is smaller than that at 61°38'. This is due to interference scattering. In order for the reader to better understand Fig. 3, the separate contributions to  $\sigma(\lambda_0, \lambda, \theta)$  for these scattering angles are plotted in Fig. 4.

### ACKNOWLEDGMENT

The transmittal of the experimental data on liquid  $CH_4$  by Y. Harker prior to publication is gratefully acknowledged. The author is further indebted to him

<sup>8</sup> P. D. Randolph, Phys. Rev. 134, A1238 (1964).

for calling attention to the misprint mentioned in Ref. 2.

#### APPENDIX

Using the same notation as in Ref. 2, the value of the various parameters<sup>9-11</sup> used in the computations were  $C_C=0$ ,  $A_C=0.64\times 10^{-12}$  cm,  $C_d=-0.41\times 10^{-12}$

<sup>9</sup> N. K. Pope, Can. J. Phys. **30**, 597 (1952).

<sup>10</sup> G. E. Bacon, *Neutron Diffraction* (Oxford University Press, London, 1955) p. 125.

<sup>11</sup> T. Y. Wu, *Vibrational Spectra and Structure of Polyatomic Molecules* (Edwards Brothers, Inc., Ann Arbor, Michigan, 1946).

cm,  $A_d=0.65\times 10^{-12}$  cm,  $b=1.093\times 10^{-8}$  cm,  $I=10.66\times 10^{-40}$  g cm<sup>2</sup>,  $\gamma_{dd}=4.02\times 10^{-19}$  cm<sup>2</sup>,  $\gamma_{CC}=3.57\times 10^{-20}$  cm<sup>2</sup>, and  $M=53.51\times 10^{-24}$  g.

The values for  $\gamma_{dd}$  and  $\gamma_{CC}$  were obtained by using the results of Ref. 9. The bound coherent scattering length  $A_d$  and the bound incoherent scattering length  $C_d$  were obtained by using the scattering lengths of Ref. 10 in their definitions.<sup>1</sup> It is perhaps worth mentioning that with an incident-neutron energy of 0.10 eV, there will be no vibrational states excited since the lowest vibrational state<sup>11</sup> of CD<sub>4</sub> is about 0.12 eV.

## Excitation of Ion-Acoustic Waves

R. W. GOULD\*

*Institut für Plasmaphysik, Garching bei München, Germany*

(Received 11 May 1964)

The excitation of ion-acoustic waves by a pair of idealized grids in a collisionless plasma is examined. It is shown that in a limited region, neither too close nor too far from the source, the disturbance closely approximates an exponentially damped (spatially) ion-acoustic wave. Far from the source a weak electron "wave" with less damping and larger wavelength dominates; near the source the potential varies as  $1/z$ .

### I. INTRODUCTION

THE velocity and damping of ion-acoustic waves in cesium and potassium plasmas have been measured by Wong, D'Angelo, and Motley.<sup>1</sup> Since this is the only experiment to date which seems to confirm the existence of Landau damping<sup>2</sup> it is important to examine the interpretation of the experimental results carefully. In this note we examine the excitation of ion-acoustic waves by a source in a collisionless unbounded plasma. We show that, *in general*, ion waves cannot be interpreted by simply examining a single root of the dispersion relation. However, we find that in a region of a few wavelengths in extent, neither too close to the source nor too far, the disturbance from an idealized sinusoidally driven pair of grids has a wavelike character with approximately exponential damping.

In obtaining this result we make use of the Fourier transform (or superposition) method together with a frequency- and wave-number-dependent complex dielectric constant.

### II. DISPERSION EQUATION FOR ION-ACOUSTIC WAVES

The complex dielectric constant for small amplitude longitudinal waves in a hot collisionless plasma may be

\* On leave from the California Institute of Technology, Pasadena, California.

<sup>1</sup> A. Y. Wong, N. D'Angelo, and R. W. Motley, Phys. Rev. Letters **9**, 415 (1962); Phys. Rev. **133**, A436 (1964).

<sup>2</sup> L. D. Landau, J. Phys. (USSR) **10**, 25 (1946).

written<sup>3</sup>

$$K(\omega, k) = \frac{1}{k^2} \int \frac{\mathbf{k} \cdot \nabla_v (\omega_{pi}^2 F_0 + \omega_{pe}^2 f_0)}{(\omega - \mathbf{k} \cdot \mathbf{v})} d^3v, \quad (1)$$

where a space and time dependence of  $e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}$  is assumed.  $F_0$  and  $f_0$  are the normalized ion and electron velocity distribution functions, respectively, and  $\omega_{pi} = (Ne^2/\epsilon_0 M_i)^{1/2}$  and  $\omega_{pe} = (Ne^2/\epsilon_0 M_e)^{1/2}$  are the ion and electron plasma frequencies, respectively. The velocity integrals are to be evaluated assuming  $\omega$  to have a *small positive* imaginary part (corresponding to having "turned on" the sources in the past) and  $k$  to be real (Fourier transform in space). It is furthermore convenient to regard the dielectric constant as functions of the *complex* variables  $\omega$  and  $k$  in which case analytic continuation, as discussed by Landau,<sup>2</sup> is implied.

Upon introducing the Maxwell velocity distribution functions

$$F_0 = (\pi^{1/2} v_i)^{-3} \exp(-v^2/v_i^2)$$

and

$$f_0 = (\pi^{1/2} v_e)^{-3} \exp(-v^2/v_e^2),$$

with  $v_i = (2\kappa T_i/M_i)^{1/2}$  and  $v_e = (2\kappa T_e/M_e)^{1/2}$ , the ion and electron thermal speeds, respectively, one obtains

$$K(\omega, k) = 1 - \frac{\omega_{pi}^2}{k^2 v_i^2} Z' \left( \frac{\omega}{k v_i} \right) - \frac{\omega_{pe}^2}{k^2 v_e^2} Z' \left( \frac{\omega}{k v_e} \right), \quad (2)$$

<sup>3</sup> B. D. Fried and R. W. Gould, Phys. Fluids **4**, 139 (1961).