

## Scattering of Slow Neutrons from Methane Gas

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The intensities of neutrons scattered with discrete energy changes from a sample of room-temperature methane gas have been measured to high enough energy changes to show the effects of excitation of the lowest vibrational states. A calculation which includes the effects of excitation of these lowest vibrational states agrees qualitatively but not quantitatively with the data.

### INTRODUCTION

IN previous experiments<sup>1,2</sup> slow neutrons were inelastically scattered from methane molecules in the gaseous phase and the effects of the molecular translations and rotations were observed. These data are well fit over most of the range of momentum and energy transfers by a calculation which takes account both of coherent and incoherent scattering and treats the rotational motion exactly by quantum mechanics,<sup>3</sup> but does not include possible contributions from vibrational transitions. A second calculational method which treats the rotational motion classically and includes coherent and incoherent scattering and averages exactly over the orientations of the molecules, also shows quite good agreement with the data when the momentum transfers are large enough to correspond to excitation of many rotational quanta.<sup>4</sup> At the largest energy transfers observed, these data indicate more scattering than is predicted by either of the above calculational procedures. This suggests that the data should be extended to higher energy transfers and that calculations which take account of vibrational excitations should be made.

The present experiment extends the measurements to higher energy transfers over a limited range of momentum transfers and into the region where vibrational excitations of the methane molecule are an important contribution to the scattering. The second

calculational procedure has been extended to include the contributions of single excitations of any of the triply degenerate vibrational modes of 0.163-eV energy and the doubly degenerate modes of 0.188-eV energy.<sup>4,5</sup> However, to make the calculations tractable for an IBM-650 computer, the coherent contributions have been omitted. In the data the increased scattering caused by excitations of the lowest vibrational states is observed. The data are higher than the theoretical curve even when the theory is extended to include the effects of vibrational excitations. This suggests either that some correction to the data has been overlooked or that some modification, such as vibration-rotation interactions or a different force field, is needed in the analysis.

### EXPERIMENTAL DETAILS

The data of this experiment were obtained using the beryllium detector apparatus at the DIDO reactor.<sup>6</sup> In this apparatus an incident beam of nearly monoenergetic neutrons is produced by diffracting neutrons of the desired energy  $E_0$  from an aluminum crystal. Before this beam of monoenergetic neutrons is scattered from the sample, its intensity is determined by a thin U-235 fission chamber. The neutrons that are scattered at  $90 \pm 5^\circ$  are observed through a beryllium filter by a shielded  $\text{BF}_3$  detector. This filter is "black" for neutrons with energies greater than 0.005 eV and the scattered neutrons which are detected have a mean final energy  $E_f$  of 0.0034 eV. Thus, the neutrons that are detected have suffered an energy change  $\epsilon = E_f - E_0 = \hbar\omega$  where  $\hbar$  is Planck's constant divided by  $2\pi$  and  $\omega$  is the frequency of the vibrational mode of the molecule.

\* The experiment was performed while this author was on a one year assignment to the United Kingdom Atomic Energy Research Establishment, Harwell.

<sup>1</sup> P. D. Randolph, R. M. Brugger, K. A. Strong, and R. E. Schmunk, *Phys. Rev.* **124**, 460 (1961).

<sup>2</sup> F. J. Webb, in *Proceedings of the Chalk River Symposium on Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1963), pp. 457.

<sup>3</sup> G. W. Griffing, in *Proceedings of the Chalk River Symposium on Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1963), pp. 435. A. Rahman, *J. Nucl. Energy* **13**, 128 (1961).

<sup>4</sup> H. L. McMurry, G. W. Griffing, W. A. Hestir, and L. J. Gannon, Atomic Energy Commission Report No. IDO-16692 (unpublished).

<sup>5</sup> H. L. McMurry, L. J. Gannon, and W. A. Hestir, *Nucl. Sci. Eng.* **15**, 438 (1963).

<sup>6</sup> D. H. Saunderson and V. S. Rainey, in *Proceedings of the Chalk River Symposium on Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1963), pp. 413.

The ratio of the counting rate of the filtered detector to the counting rate of the monitor is proportional to  $(k_0/k_f)d^2\sigma/d\Omega d\epsilon \times$  (correction for non- $1/v$  dependence of the fission monitor) and this is equal to  $\exp(-\epsilon/2K_B T)S(|\kappa|, \hbar\omega)$ . Here  $d^2\sigma/d\Omega d\epsilon$  is the partial differential cross section,  $\bar{\kappa} = \bar{k}_0 - \bar{k}_f$  and  $\bar{k}_0$  and  $\bar{k}_f$  are the initial and final wave vectors of the neutrons,  $K_B$  is Boltzmann's constant,  $T$  is the absolute temperature of the sample, and  $S(|\kappa|, \hbar\omega)$  is the reduced partial differential cross section.<sup>7,8</sup> The data are presented as  $\exp(-\epsilon/2K_B T)S(|\kappa|, \hbar\omega)$  versus  $\hbar\omega$ . Since the beryllium detector apparatus was not calibrated for absolute measurements, the data of this experiment have been normalized to the absolute values of Randolph *et al.*<sup>1</sup> at an energy transfer of 0.0253 eV. This normalization compensates for a lack of knowledge of the solid angle, the flux, and the number of atoms in the sample.

The sample of 99+% purity methane gas at a pressure of 114 psig and room temperature was contained in a cylindrical sample can 3 in. high by 2 in. diam with 0.030-in. walls. This sample has a transmission of about 0.82 for 0.028-eV neutrons and of 0.91 for 0.18-eV neutrons.

To obtain the data, a cadmium shutter which is located after the fission monitor but before the sample, is oscillated into the beam at 10–50-min intervals. The difference between the counting rates of the filtered detector is the effect of scattering the sub-cadmium neutrons which are in the beam. For each incident energy the ratio of this difference to the rate of the monitor was obtained for runs of 15–75 h. Then the aluminum monochromating crystal was rotated off the Bragg angle and a similar run was made to see if any correction needed to be made for thermal neutrons passing through the cadmium or fast neutrons being stopped by the cadmium. Only at the highest incident energies were corrections necessary and made. This off Bragg measurement also provided a background correction for the fission monitor.

At each incident energy a measurement of the order contamination of the beam was made by measuring the transmission of gold. At  $E_0 = 0.028, 0.075,$  and  $0.1798$  eV the amounts of 1st order in the beam were 98.6, 100, and 90%. Since the 2nd order in the beam was small, no correction for it has been applied to the data.

The filter has a transmission of 1/2000 for neutrons above 0.005 eV when the transmission is measured with the detector and beryllium filter in their shielding. Using the theory of this paper, spectra were calculated for each incident neutron energy. The area of these spectra at neutron energies above 0.005 eV were multi-

<sup>7</sup> R. M. Brugger, U. S. Atomic Energy Commission Report, IDO-16694, Rev., 1962 (unpublished). This report is also designated as TNCC-US-21 (Rev.) and EANDC-US-25.

<sup>8</sup> P. A. Egelstaff, in *Proceedings of the Chalk River Symposium on Inelastic Scattering of Slow Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1963), pp. 25.

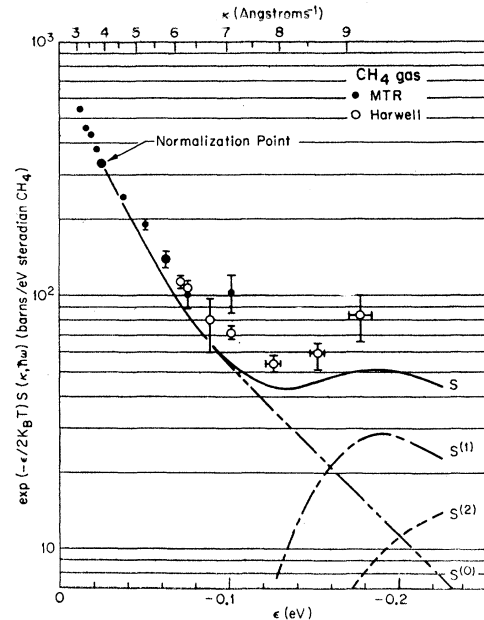


FIG. 1. The scattering intensity versus energy change for the scattering of neutrons from methane gas. The open circles are the data of this experiment and the solid circles are the data of Randolph *et al.* (Ref. 1). The lines represent the predicted scattering as defined in the text.

plied by 1/2000 and compared to the area of the spectra below 0.005 eV. The ratio of the first areas  $\times 1/2000$  to the second areas gave corrections for the finite transmission of the filter for neutrons with energies greater than 0.005 eV. This correction lowered the points by about 15% at the highest energy changes. Similar calculations were made to determine how much difference in  $\exp(-\epsilon/2K_B T)S(|\kappa|, \hbar\omega)$  would occur if the final energy were 0.005 eV instead of 0.0034 eV, or if the incident energy were 0.96 of its stated value. Less than 5% changes were obtained from these calculations indicating that energy resolution of the final or incident neutrons introduces only a small error into the data.

## RESULTS

Figure 1 shows the data obtained in this experiment and corresponding points obtained by Randolph *et al.*<sup>1</sup> The errors on the present data include an estimate of counting statistics, and uncertainties in order and background corrections. The errors on the Randolph data give an indication of the spread of their points. The two sets of data are in agreement.

## THEORY

Values of  $S(|\kappa|, \hbar\omega)$  are calculated as the sum of parts  $S^{(0)}$ ,  $S^{(1)}$ , and  $S^{(2)}$  which correspond to no vibrational excitations, excitations of the 0.163-eV modes, and excitation of the 0.188-eV modes, respectively. Figure 1 shows the individual values of  $S^{(0)}$ ,  $S^{(1)}$ , and  $S^{(2)}$  and their sum. The calculated and measured values

of  $\exp(-\epsilon/2K_B T)S(|\kappa|, \hbar\omega)$  agree at low  $\epsilon$  but as  $\epsilon$  increases, the experimental values rise above the calculated ones. The discrepancy is rather small in the region where the  $S^{(1)}$  and  $S^{(2)}$  contributions are negligible. But where these contributions dominate, the calculated values are only about  $\frac{2}{3}$  the measured ones.

This discrepancy between the experimental and calculated values suggests that either an appreciable correction to the data has been overlooked or that the computations fail to deal adequately with the vibrational excitations. The contributions from excitation of the higher energy modes near 0.36 eV, and from multiple excitations of the low-energy modes, have been examined and found to be negligible. Therefore, the computational errors must be sought in the procedures used in calculating  $S^{(1)}$  and  $S^{(2)}$ . When rotation-vibration coupling and anharmonic effects are neglected, the vibrational intensities are related to the expectations  $\langle \Delta r_{\nu\tau}^2 \rangle$ . Here  $\Delta r_{\nu\tau}$  is the displacement of the scattering atom  $\nu$  during the  $\tau$  normal vibration. These expectations depend on the force field used in the normal

coordinate analysis. Since more than one force field may give the correct vibrational frequencies but different normal modes, there is some possibility for altering  $S^{(1)}$  and  $S^{(2)}$  by changing the force field. Rotation-vibration coupling and anharmonic effects will cause perturbations which will produce some modification in the  $\langle \Delta r_{\nu\tau}^2 \rangle$ . The possibility that the proper treatment of one or more of these factors will resolve the discrepancy between calculated and observed results is being studied, but the work has not progressed to where definite conclusions can be drawn.

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### Characteristic X-Ray Production in the $M_V$ Shell in Ytterbium by 30–100-keV Protons\*

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Characteristic  $M$ -shell x rays produced when protons of 30–100-keV energy are stopped in a thick target of ytterbium were observed by proportional-counter detection. By use of an aluminum absorber, radiation originating from the filling of  $M_V$ -subshell vacancies can be isolated. Thick-target yields of the  $M_V$ -shell and ( $M_I + M_{II} + M_{III} + M_{IV}$ )-shell x rays were measured separately. With extrapolated values of the fluorescence yield (0.06), the mass absorption coefficient (1500 cm<sup>2</sup>/g) and the stopping power, estimates of the ionization cross section for the  $M_V$  shell have been made (neglecting contributions from other subshells by Coster-Kronig transitions). These vary from  $6 \times 10^{-26}$  cm<sup>2</sup> at 30 keV to  $2 \times 10^{-22}$  cm<sup>2</sup> at 100 keV.

#### INTRODUCTION

THE investigation of ionization of atoms by proton bombardment has long been in the domain of nuclear physics. It has been mainly concerned with the evaluation of stopping powers and background subtraction in Coulomb excitation and other nuclear reaction experiments.<sup>1</sup> As time has passed and more detailed data have become available, many questions of a purely atomic nature have arisen.<sup>2</sup> The answers to these involve knowledge of the transition probabilities for the radiative and nonradiative reorganization

processes taking place in the atom following an ionizing event.<sup>3</sup> Description of the total event from the ionization event to the observation of x rays in a detector places a severe test upon our understanding of atomic processes.

The phenomena can be separated into two parts; the production of "initial" vacancies in the inelastic scattering process and the reorganization of the atom filling all vacancies. During this reorganization the "initial" vacancies may be redistributed among the subshells of any given shell (Coster-Kronig transitions), e.g.,  $M_I \cdots M_V$  subshells.<sup>3</sup> In cases where the quantum energy of the radiative process is low ( $< 5$  keV), few ( $< 0.1$ ) of the total number of vacancies are filled by photon emission. Given an accurate description of either the ionization event or the reorganization

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<sup>2</sup> J. M. Khan, D. L. Potter, and R. D. Worley, *Phys. Rev.* 134, A316 (1964).

<sup>3</sup> E. H. S. Burhop, *The Auger Effect and other Radiationless Transitions* (Cambridge University Press, Cambridge, 1952).