# Neutron Scattering Cross Sections of Para- and Orthohydrogen, and of $N_2$, $O_2$, and $H_2O$*

F. G. BRICKWEDDE,** JOHN R. DUNNING,† HAROLD J. HOGE,** AND JOHN H. MANLEY,††

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Measurements were made of the transmission of liquid para- and normal hydrogen for the slow neutrons obtained from a Rn-Be source by scattering in paraffin. In some of the experiments the paraffin was at room temperatures and in others it was cooled with liquid air. Teller, and Schwinger and Teller, have shown how the spin dependence of the interaction between neutrons and protons can be investigated by such measurements. As the neutrons captured by the protons were less than 1 percent of the scattered neutrons, the scattering by the thin layers of liquid hydrogen used was determined from the transmission measurements. The scattering cross section of liquid hydrogen for slow neutrons was found to depend upon the velocity of the neutrons and on the *ortho-para* composition of the hydrogen. The following values were obtained for the neutron scattering cross sections, $\sigma$, of liquid ortho- and parahydrogen: for $\sim$300°K neutrons (paraffin at room temperatures), $\sigma$ (*ortho*) $=56\times10^{-24}$ cm$^2$ per molecule, $\sigma$ (*para*) $=29\times10^{-24}$; for $\sim$120°K neutrons (paraffin cooled with liquid air), $\sigma$ (*ortho*) $=79\times10^{-24}$ and $\sigma$ (*para*) $=18\times10^{-24}$. We were able to prove conclusively: (1) that the interaction between neutrons and protons is dependent upon the relative alignment of their spins, and (2) that the energy of the singlet state of the deuteron, in which the spins of the proton and neutron are antiparallel, is greater than the energy of these particles when far apart, that is, the singlet state of the deuteron is virtual. The scattering+capture cross sections of liquid oxygen and liquid nitrogen were measured for $\sim$300°K neutrons, and of water for $\sim$300° and $\sim$120°K neutrons. The cross sections for the 300°K neutrons were: $\sigma(O)=4\times10^{-24}$ cm$^2$ per atom, $\sigma(N)=13\times10^{-24}$ cm$^2$ per atom and $\sigma(H_2O)=91.2\times10^{-24}$ cm$^2$ per molecule. For the $\sim$120°K neutrons $\sigma(H_2O)=116\times10^{-24}$ cm$^2$ per molecule. The scattering cross sections of the protons in water, $(1/2)[\sigma(H_2O)-\sigma(O)]$, were $43.6\times10^{-24}$ cm$^2$ per proton for $\sim$300°K neutrons and $56\times10^{-24}$ cm$^2$ per proton for $\sim$120°K neutrons.

## INTRODUCTION

THE abnormally large scattering cross section of hydrogen for slow neutrons led to the assumption that the interaction between neutron and proton depends on the spins of these particles. This hypothesis, attributed to Wigner by Feenberg and Knipp,[1] had been generally accepted although it lacked direct experimental verification.[2] Teller[3] pointed out that the existence or nonexistence of a spin dependent neutron-proton interaction could be determined from a study of the scattering of slow neutrons by para- and orthohydrogen. He showed that if the interaction proved to be spin dependent it should be possible to tell from such experiments whether the singlet level of the deuteron is stable or virtual. In the detailed analyses subsequently carried out by Schwinger and Teller[4] it was shown that the effects were large enough to be readily detected experimentally, and that it might also be possible to obtain information concerning the range of the neutron-proton forces. Schwinger[5] has since pointed out that such experiments would also serve to confirm the choice of $\frac{1}{2}$ rather than $\frac{3}{2}$ for the spin of the neutron, a choice hitherto made entirely because of its simplicity.

In discussing the para- and orthohydrogen scattering cross sections for slow neutrons, briefly referred to as the *para* and *ortho* cross sections, three possibilities are to be considered:

(1) No dependence of forces on spin
(2) Spin dependent forces with
  (a) a real (stable) singlet level of the deuteron
  (b) a virtual singlet level of the deuteron.

In case (1) the only difference to be expected in the *para* and *ortho* scattering cross sections arises from the different spacing of their rotational energy levels. At room temperature only the first few levels are occupied, and for neutrons of energy comparable to the excitation energies

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** National Bureau of Standards, Washington, D. C.
† Columbia University, New York, N. Y.
†† Columbia University, New York, N. Y. Now at the University of Illinois.
[1] E. Feenberg and J. K. Knipp, Phys. Rev. **48**, 906 (1935).
[2] See reference 4 for a discussion of the evidence to be obtained from Amaldi and Fermi's measurements of the cross section for radiative capture.
[3] E. Teller, Phys. Rev. **49**, 420 (1936).
[4] J. Schwinger and E. Teller, Phys. Rev. **52**, 286 (1937).
[5] Julian Schwinger, Phys. Rev. **52**, 1250 (1937).

of these levels parahydrogen should have the larger cross section, since the closer spacing of its levels makes inelastic scattering more probable. The difference should be small however and should be negligible for neutrons either above or below this energy range. In the absence of spin forces all scattering must be of the *para→para* or *ortho→ortho* type, that is the rotational quantum number of the scattering molecule must change by 0, $\pm 2$, $\pm 4$, $\cdots$.

In cases (2a) and (2b) the presence of spin forces makes possible in addition *para→ortho* and *ortho→para* scattering ($\Delta J = \pm 1, \pm 3, \cdots$). If, as is the case in liquid hydrogen, the scattering molecules are in their lowest rotational levels ($J=0$ for *para*, $J=1$ for *ortho*) spin forces should cause the *para* and *ortho* cross sections to differ markedly for neutrons of energy insufficient to excite the $0\rightarrow 1$ transition. For neutrons of this energy no inelastic scattering is possible in parahydrogen, while in orthohydrogen the scattering process involving the $1\rightarrow 0$ transition remains.

For slow neutrons whose wave-length is comparable to the distance between the protons in a hydrogen molecule interference effects are important, and it is chiefly in these that cases (2a) and (2b) differ. The phase of a neutron wave scattered by a proton depends on the sign of the binding energy involved, and changes by approximately $\pi$ when the binding energy changes sign. Hence in case (2a) the scattered waves corresponding to singlet and triplet interaction would have like phases, while in case (2b) they would have opposite phases. Case (2b) brings a possibility of obtaining destructive interference in parahydrogen with its antiparallel proton spins, for here interactions of the singlet and triplet type should occur simultaneously. The scattered waves should interfere destructively, with the result that for low energy neutrons the scattering cross section of parahydrogen should be much smaller than that of orthohydrogen, where this type of interference cannot occur. However, simple calculations based on the idea that the spin of a neutron impinging on a parahydrogen molecule is parallel to one proton spin and antiparallel to the other are not legitimate, although the trends they indicate are in the right direction. Schwinger and Teller have considered the problem more carefully and have given the necessary detailed calculations based on the use of appropriate wave functions.

Experiments to test their predictions, undertaken jointly by Columbia University and the National Bureau of Standards, have been carried out in the Bureau's Low Temperature Laboratory. Preliminary reports[6, 7] have already been published.

Considerations of intensity and geometrical arrangement render it more feasible to perform transmission than scattering experiments. Since the neutron-proton capture cross section[8, 9] is of the order of $0.3\times 10^{-24}$ cm$^2$, less than 1 percent of the average scattering cross section, the difference between the scattering+capture cross section given by transmission experiments and the true scattering cross section is inappreciable. Good geometrical conditions are exceedingly important in transmission experiments. Experiments under other conditions, employing non-parallel neutron beams, and with both source and detector close to the scattering sample have been shown to be especially unreliable with scattering samples containing hydrogen. The uncertain angular distribution of the neutrons, together with large corrections for multiple scattering, slowing down processes, back scattering, etc., make such experiments very difficult to interpret. The design of the present apparatus was such as to make the errors introduced by geometrical factors negligible.

Since the apparatus was well adapted for use with other liquefied gases, the transmissions of liquid $O_2$ and $N_2$ were also measured. As a useful check on the accuracy of the work, several measurements were also made with water in the apparatus.

## Apparatus and Procedure

The apparatus divides naturally into two parts: one the equipment for preparing and analyzing liquid hydrogen and maintaining it in the path of the neutron beam, and the other the source, detector, and amplifier system. The

[6] J. R. Dunning, J. H. Manley, H. J. Hoge, F. G. Brickwedde, Phys. Rev. **52**, 1076 (1937).
[7] J. R. Dunning, H. J. Hoge, J. H. Manley, and F. G. Brickwedde, Phys. Rev. **53**, 205 (1938).
[8] E. Amaldi and E. Fermi, Phys. Rev. **50**, 899 (1936).
[9] G. A. Fink, Phys. Rev. **50**, 738 (1936).

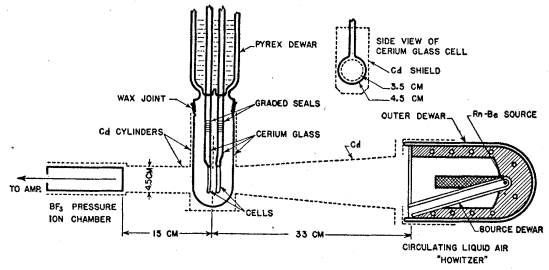


FIG. 1. Arrangement of source and detector relative to scattering sample. Scattering material was condensed in one or both of the two cells in the lower vacuum chamber of the special Dewar flask.

arrangement of this second part of the apparatus is shown in Fig. 1. The sample was contained in either or both of two cerium glass cells which formed part of a specially designed Dewar flask. The Rn-Be source of approximately 500 millicuries was contained in a "howitzer" in which the neutrons were slowed down. This howitzer[10] consisted of a mass of paraffin in the shape of a deep, very thick walled cup contained in a Dewar flask. Copper tubing for the circulation of liquid air was imbedded in the paraffin, and thermocouples were provided at two different points for temperature measurement. Immediately in front of the source was a solid copper cylinder to shield the detector from gamma-rays. The source was placed in a small source Dewar (2 cm diameter) containing a thermocouple and a heater. These were used when the howitzer was cooled with liquid air, to avoid any change in efficiency of the source due to localized condensation of the Rn. When using liquid air it was also necessary to heat the outer copper baffle at the mouth of the howitzer, to prevent the collection of moisture or frost. The effectiveness of the howitzer in cooling the neutrons will be discussed later in the paper.

The neutron beam was well diaphragmed and shielded by cadmium as shown in Fig. 1. The opening in the cadmium diaphragm placed between the two cells was 3.5 cm in diameter, 1 cm less than the diameter of the cells. The detector was an ionization chamber 3 cm in diameter and 13 cm in length, filled to a pressure of 3 atmospheres with $BF_3$ gas. The average solid angle subtended by the chamber at the cells was less than 0.02 steradian. The chamber was connected to a linear amplifier whose output was recorded by a scale-of-two thyratron counter.

All glass in the path of the neutron beam was a special cerium glass, whose exceptionally low boron content results in very small neutron absorption. Total glass in the path of the beam was about 6 mm, the cell walls being slightly less than a millimeter thick and the Dewar walls slightly more. Total absorption by empty cells, Dewar walls, and silvered surfaces was about 6 percent for room temperature ($\sim$300°K) neutrons. Cell volumes were determined by weighing mercury and diameters by direct measurement. These data gave the mean internal cell thicknesses, which appeared very uniform over the entire area of a cell. In any case only a difference in the mean thickness of the exposed (60 percent) and shielded (40 percent) area of a cell would cause appreciable error. Transmission measurements with water in the cells were used to check the cell thicknesses. Satisfactory agreement was found in all cases except one. In this case (0.234 cm cell) the transmission data were used to determine the thickness of the cell, resulting in a small reduction of the value originally found.

Figure 2 is a diagram of the apparatus used in the preparation and analysis of the liquid hydrogen samples. Glass tubes leading to the cells passed through liquid hydrogen in the upper part of the Dewar. Hydrogen of the desired composition was condensed in these tubes and ran down into the cells. This arrangement permitted the amount of hydrogen in the cells to be kept constant during the course of the measurements, for any hydrogen lost by evaporation was simply recondensed and returned to the cells. Since the level of the liquid in the cells could not be observed, the volume of gas condensed was measured. This was done either by taking the hydrogen from a cylinder or tank of known volume and measuring the fall in pressure, or by using a Toepler pump of known capacity. A considerable margin of safety was allowed, sufficient to bring the level of the liquid well up into the connecting tubes.

Hydrogen to be condensed in the cells was taken from a supply tank of especially prepared electrolytic hydrogen, the purity of which was

[10] P. N. Powers, H. Carroll, J. R. Dunning, Phys. Rev. **51**, 1112 (1937).

believed to be in excess of 99.98 mole percent. This hydrogen was passed over hot copper (600°C) and through a liquid-air trap to remove oxygen, which is an active *ortho-para* conversion catalyst in liquid hydrogen.[11] Passage over hot copper also insured attainment of the high temperature equilibrium concentration of 75 percent *ortho* 25 percent *para* (normal hydrogen). When normal hydrogen was desired in a cell, this gas was condensed directly into it. When parahydrogen was desired, the normal hydrogen was first condensed in a tube containing a conversion catalyst in granular form. The catalyzing chamber was kept at 20.4°K by surrounding it with liquid hydrogen. At this temperature the equilibrium concentration is 99.8 percent parahydrogen.[12]

Hydrogen from the catalyzing chamber was either condensed directly in the cells or stored until needed in a tank which could be filled at liquid-air temperature and emptied at room temperature.

A promoted iron catalyst[13] of the kind used in the synthesis of ammonia was placed at our disposal by Dr. P. H. Emmett. The catalyst, received in the form of an oxide, was prepared for use by reducing in hydrogen at 500°C, outgassing while hot and cooling to 20.4°K before admitting hydrogen again. Liquid hydrogen condensed on the catalyst was allowed to remain in contact with it 15 minutes or longer before use.

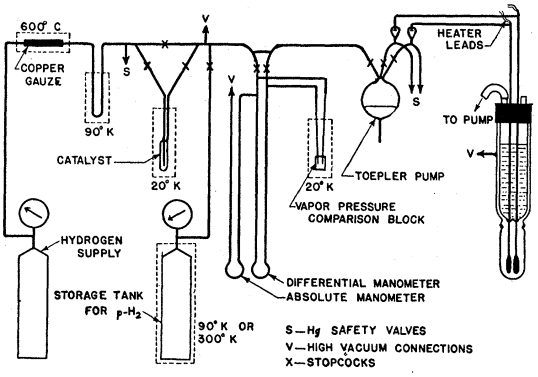


FIG. 2. Apparatus used in the preparation and analysis of the liquid hydrogen samples.

[11] R. B. Scott, F. G. Brickwedde, H. C. Urey, and M. H. Wahl, J. Chem. Phys. **2**, 454 (1934).
[12] R. W. Harkness and W. E. Deming, J. Am. Chem. Soc. **54**, 2850 (1932).
[13] P. H. Emmett and R. W. Harkness, J. Am. Chem. Soc. **57**, 1624 (1935).

The *ortho-para* composition of the contents of any cell was determined immediately after transmission measurements on it had been completed. Brickwedde and Scott[14] have measured the vapor pressures of normal hydrogen, parahydrogen, and mixtures of several intermediate concentrations, using a differential oil manometer to measure the pressure differences. With the aid of their results the composition of an unknown sample of hydrogen may be determined by comparing its vapor pressure with that of normal hydrogen. In the neighborhood of their boiling points the vapor pressures of para- and normal hydrogen differ by about 26 mm Hg; that of *para* is higher. This is approximately 1 mm fall in vapor pressure for each 3 percent increase in *ortho* content. In making an analysis about one-fourth of the contents of a cell was removed; then a sample from the cell and a sample of freshly prepared normal hydrogen were condensed in separate compartments in the vapor pressure comparison block. Their vapor pressure difference was then read on the differential mercury manometer, the total pressure on the normal hydrogen being given by the absolute manometer. The copper comparison block was connected to the glass system by 3 mm copper-nickel tubes.

When parahydrogen (99.8 percent) is condensed in a cell there is no change in composition with time since the material is in equilibrium. With normal hydrogen this is not true, for even with no catalyst present the *ortho* content decreases at the rate of about 1 percent per hour.[11] When analysis of a sample from a cell filled with normal hydrogen showed a composition of, for example, 72 percent orthohydrogen, it was assumed that the composition had changed linearly with time from 75 percent at the time of condensation until the time of analysis. The compositions given in Table I correspond to the mean time of transmission measurement. Strictly the change with time is not quite linear since it depends on collisions between two orthohydrogen molecules. Because the frequency of this type of collision is proportional to the square of the orthohydrogen concentration the corrections were negligible for all "*para*" samples.

It was necessary to reduce the pressure above the liquid hydrogen bath until its temperature

[14] To be published.

fell almost to the triple point before liquid hydrogen would collect in the cells. With the bath at this temperature and the gas pressure in the cell slightly above one atmosphere rapid condensation would begin after 15 or 20 minutes. Liquid was kept in the cells under a pressure of 200–600 mm Hg, the mean temperature of the samples being taken as 18.4°K. To remove liquid a small heating coil of No. 38 constantan wire (50 ohms) was placed in the bottom of each cell, below the edge of the cadmium diaphragm.

## Measurements

Each transmission value was computed from the results of from 3 to 6 ten minute counts. Roughly one-third of the counts were taken with a cadmium sheet 0.08 cm thick placed just in front of the howitzer, so that the effect of the high energy cadmium-penetrating neutrons could be determined. After a set of counts a cell was either filled or emptied and another set taken. The normal order was: (1) both cells empty, (2) first (thicker) cell full, (3) both cells full, (4) second cell full (first empty), (5) both cells empty. With both cells empty the approximate number of neutrons counted in 10 minutes was 2500, of which all but approximately 18 percent were cut off by interposing the cadmium sheet. After a series of cell fillings with normal hydrogen a similar series was taken with parahydrogen,

Table I. *Transmissions and cross sections (scattering+capture) of para- and normal hydrogen for neutrons of four different energy distributions. T is the neutron temperature; C is the composition in percent of orthohydrogen of the scattering medium; x is the thickness; $I/I_0$, the transmission; $\sigma$, the cross section per molecule.*

| $T$ | $C$ % $o-H_2$ | $x$ | $I/I_0$ | $-\ln I/I_0 = n\sigma x$ | $\sigma$(obs.) | $\sigma$(obs.) $-\sigma$(calc.)§ | Cross Sections of Pure Components (by Extrapolation) |
|---|---|---|---|---|---|---|---|
| | | cm | | | $10^{-24}$ cm$^2$ | $10^{-24}$ cm$^2$ | $10^{-24}$ cm$^2$ |
| ~300°K | 74.2 | 0.234 | 0.780 | 0.249 | 48.6 | −0.2 | $\sigma_{ortho}=55.8$ |
| | 72.5 | 0.32 | 0.705 | 0.351 | 50.1 | +1.7 | |
| | 73.9 | 0.35 | 0.695 | 0.363 | 47.4 | −1.4 | |
| | 74.0 | 0.58 | 0.538 | 0.620 | 48.7 | −0.1 | |
| | 73.3 | 0.93 | 0.371 | 0.991 | 48.6 | 0.0 | |
| | 1.7 | 0.234 | 0.871 | 0.139 | 27.1 | −2.3 | $\sigma_{para}=29.0$ |
| | 1.4 | 0.32 | 0.809 | 0.212 | 30.1 | +0.7 | |
| | 3.0 | 0.58 | 0.671 | 0.40 | 31.4 | +1.6 | |
| | 2.5‡ | 0.814 | 0.593 | 0.521 | 29.3 | (−0.4) | |
| ~120°K | 74.5 | 0.234 | 0.720 | 0.329 | 64.2 | +1.1 | $\sigma_{ortho}=78.7$ |
| | 73.8‡ | 0.32 | 0.656 | 0.422 | 60.1 | (−2.6) | |
| | 72.6 | 0.58 | 0.449 | 0.80 | 63.0 | +1.1 | |
| | 74.0 | 0.814 | 0.34 | 1.079 | 60.6 | −2.2 | |
| | 1.4 | 0.234 | 0.911 | 0.092 | 18.0 | −0.4 | $\sigma_{para}=17.6$ |
| | 4.1 | 0.32 | 0.879 | 0.128 | 18.3 | −1.8 | |
| | 6.5 | 0.58 | 0.740 | 0.301 | 23.6 | +2.0 | |
| | 5.0 | 0.814 | 0.690 | 0.371 | 20.8 | +0.2 | |
| | 5.9, 74.3 } 25.6 | 0.58, 0.234 } 0.814 | 0.50* | 0.693 | 38.9 | +5.7 | |
| | 73.7‡, 16.2 } 57.2 | 0.58, 0.234 } 0.814 | 0.39* | 0.941 | 52.7 | +0.2 | |
| ~120°K filtered through 0.58 cm $n-H_2$ | 74.2, 73.4 | 0.58, 0.234 | 0.769† | 0.263 | 51.1 | — | $\sigma_{ortho}=63.0$ |
| | 73.7, 16.2 | 0.58, 0.234 | 0.878† | 0.130 | 25.4 | — | $\sigma_{para}=18.1$ |
| ~120°K filtered through 0.58 cm $p-H_2$ | 5.9, 74.3 | 0.58, 0.234 | 0.671† | 0.399 | 77.6 | — | $\sigma_{ortho}=100.1$ |
| | 6.5, 1.4 | 0.58, 0.234 | 0.932† | 0.070 | 13.8 | — | $\sigma_{para}=12.6$ |

* Transmission of both cells considered as a unit, one containing $p-H_2$, the other $n-H_2$.
† Transmission of second cell for filtered beam from first cell.
‡ Estimated.
§ Deviations from linear equations of Fig. 3.

TABLE II. *Transmissions of liquid $O_2$ and $N_2$ and cross sections (scattering+capture) per atom for ~300°K neutrons. The thickness is designated by x. $I/I_0$ is the transmission and $\sigma$ is the cross section.*

| MATERIAL | $x$ | $I/I_0$ | $-\ln I/I_0 = n\sigma x$ | $\sigma$ | AVERAGES |
|---|---|---|---|---|---|
| | cm | | | $10^{-24}$ cm² | $10^{-24}$ cm² |
| Liquid $O_2$ | 0.35 | —— | —— | —— | $\sigma(0)=4.05$ |
| | 0.58 | 0.905 | 0.995 | 3.98 | |
| | 0.93 | 0.847 | 0.166 | 4.12 | |
| Liquid $N_2$ | 0.35 | 0.870 | 0.140 | 11.5 | $\sigma(N)=12.7$ |
| | 0.58 | 0.761 | 0.273 | 13.5 | |
| | 0.93 | 0.657 | 0.420 | 13.0 | |

all with the howitzer at room temperature. Then the howitzer was cooled with liquid air to approximately 100°K and the process repeated. In this condition the transmission was also measured with parahydrogen in the first cell, normal hydrogen in the second, and vice versa. Data on the cadmium penetrating background have not been included in the tables. This background has in all cases been subtracted to give the transmission values reported. Data for hydrogen are given in Table I. In calculating the cross sections, $n$ the number of molecules per cm³ was taken from the results of Scott and Brickwedde.[15] It is interesting to note that because of the large thermal expansion of liquid hydrogen this number is approximately 3 percent greater at 18.4°K than at 20.4°K, the boiling point of normal hydrogen.

The procedure with liquid $O_2$, $N_2$, and $H_2O$ was similar to that with hydrogen except that no measurements on $O_2$ and $N_2$ were made while the howitzer was cooled with liquid air. Data for liquid $O_2$ and $N_2$ are given in Table II; for water in Table III.

## DISCUSSION OF RESULTS

The dependence of cross section on composition and on neutron energy is shown in Fig. 3, where cross section per hydrogen molecule has been plotted against percent orthohydrogen in the scattering layer. Each of the four straight lines refers to neutrons of a different energy distribution. The equations of the lines were determined simply to make the sum of the deviations zero for the *para* group of points and likewise zero for the points taken with normal hydrogen. The intercepts of these lines on the

[15] R. B. Scott and F. G. Brickwedde, J. Chem. Phys. **5**, 736 (1937).

TABLE III. *Transmission of water and the cross sections (scattering+capture) of (1) water per molecule and of (2) the protons of water per proton, for neutrons of ~300°K. and ~ 120° K. T is the neutron temperature; x is the thickness; $I/I_0$ is the transmission; $\sigma_p=\frac{1}{2}(\sigma(H_2O)-\sigma(0))$. Average for "room temperature" neutrons $\sigma_p=42.2\times10^{-24}$ cm². The 0.93 cm thickness was not used in obtaining this value.*

| $T$ | $x$ | $I/I_0$ | $-\ln I/I_0 = n\sigma x$ | $\sigma(H_2O)$ | $\sigma_p$ |
|---|---|---|---|---|---|
| | cm | | | $10^{-24}$ cm² | $10^{-24}$ cm² |
| ~300°K | $0.23_4$ | 0.495 | 0.703 | .... | .... |
| ~270°K | 0.32 | 0.370 | 0.994 | 92.0 | 44.0 |
| ~300°K | 0.35 | 0.349 | 1.05 | 89.1 | 42.5 |
| " | 0.58 | 0.190 | 1.66 | 85.0 | 40.5 |
| " | 0.58 | 0.170 | 1.77 | 90.5 | 43.2 |
| " | 0.93 | 0.079 | 2.54 | 81.2 | 38.6 |
| ~120°K | $0.23_4$ | 0.400 | 0.915 | 116.0 | 56.0 |

0 and 100 percent axes give the cross sections of pure para- and pure orthohydrogen for neutrons of each energy distribution. The values of these intercepts are given in the last column of Table I. The use of linear extrapolation requires some justification. For very thin layers a linear relation between cross section and composition is to be expected, since this requires only that the cross section of an individual molecule be independent of whether its neighbors are para- or ortho-molecules. For thick layers a nonlinear relation is to be expected because of multiple scattering. The geometry of the present apparatus was such as to minimize any complications due to multiple scattering, since a neutron was effectively removed from the beam by its first collision, its subsequent history being of no importance. The transmission curves for water (Fig. 4) which has approximately the same proton density as liquid hydrogen showed a reasonably smooth exponential decrease which is experimental evidence that multiple scattering was not significant.

The energy distribution of the neutrons is the most uncertain factor in the interpretation of the results. This depends on the effectiveness of the howitzer in cooling down the neutrons from their high energy and establishing something of the nature of a Maxwellian distribution. A howitzer similar to the one used in these experiments had previously been tested with a velocity selector.[16–18] With the howitzer at room tempera-

[16] J. R. Dunning, G. B. Pegram, G. A. Fink, D. P. Mitchell, E. Segrè, Phys. Rev. **48**, 704 (1935).
[17] G. A. Fink, J. R. Dunning, G. B. Pegram, D. P. Mitchell, Phys. Rev. **49**, 103 (1936).
[18] G. A. Fink, Phys. Rev. **50**, 738 (1936).

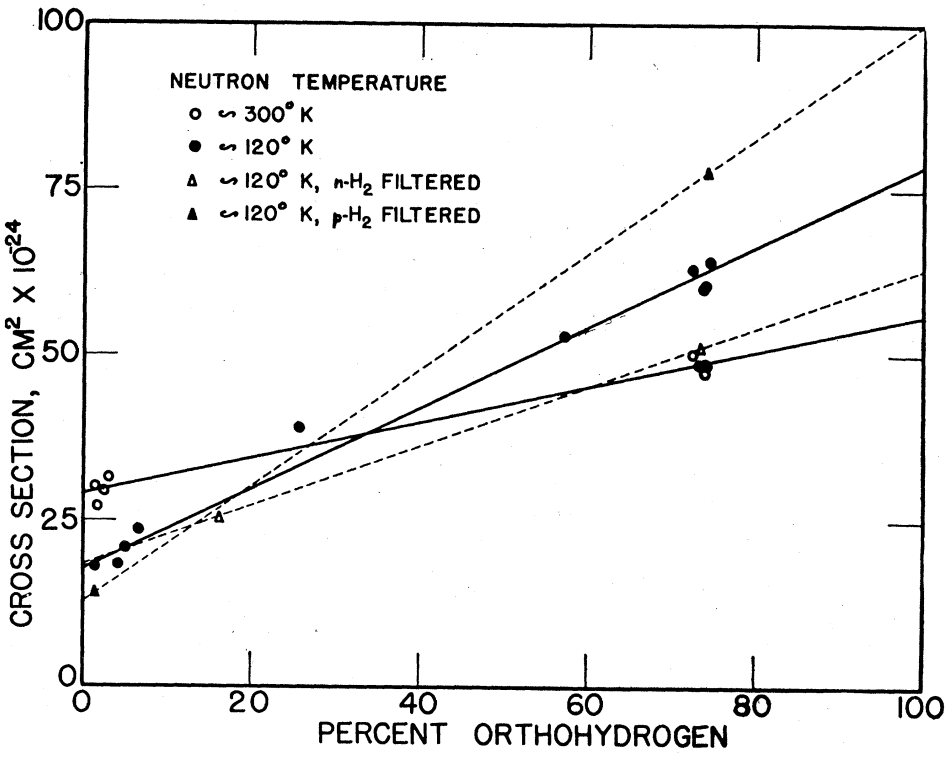


FIG. 3. Dependence of neutron cross section (scattering + capture) per molecule on *ortho-para* composition of the liquid hydrogen scattering sample. Each straight line refers to neutrons of a different energy distribution. The indicated temperatures are only approximate.

ture the emergent neutrons absorbable in cadmium were found to possess a distribution whose maximum corresponded approximately to thermal equilibrium with the paraffin. However, it was shown that more high energy neutrons were present than in a true Maxwellian distribution. The effect of all high energy neutrons above the cadmium absorption limit (about 0.4 ev[19, 20]) is eliminated by the cadmium difference method, but there still remains a long high energy "tail" extending up to about 0.4 ev.

The obtaining of a beam of neutrons with a Maxwellian distribution of velocities characteristic of liquid-air temperatures is of greater difficulty than for room temperatures.[16–18, 21, 22] The cooling efficiency of the howitzer used in these experiments was investigated by Mr. P. N. Powers of Columbia University, who measured the absorption of the emerging neutrons in boron. Using the cadmium difference method, he found that the absorption cross section of the neutrons in boron was 1.6 times greater when the howitzer was cooled with liquid air than when it was at room temperature.

Experiments have indicated that absorption in boron follows the $1/v$ law[23, 24] hence, if $T_1$ and $T_2$ are the "effective" temperatures of the neutron beams obtained when the howitzer is at room temperature and when it is cooled with liquid air, respectively, we have

$$(T_1/T_2)^{\frac{1}{2}}=1.6. \qquad (1)$$

The two imbedded thermocouples indicated that the average temperature of the paraffin was approximately 105°K while liquid air was being circulated. In the ideal case in which the neutron

[19] O. R. Frisch and G. Placzek, Nature **137**, 357 (1936).
[20] J. G. Hoffman and M. S. Livingston, Phys. Rev. **52**, 1228 (1937).
[21] O. R. Frisch, H. von Halban, Jr., Jørgen Koch, Nature **139**, 922 (1937).
[22] P. B. Moon and J. R. Tillman, Proc. Roy. Soc. **A153**, 476 (1936).
[23] F. Rasetti, E. Segrè, G. Fink, J. R. Dunning, G. B. Pegram, Phys. Rev. **49**, 104 (1936).
[24] F. Rasetti, D. P. Mitchell, G. A. Fink, G. B. Pegram, Phys. Rev. **49**, 777 (1936).

beams would be in thermal equilibrium with the paraffin at either temperature and the $BF_3$ receiver counted all neutrons passing through it, this ratio would be $(300°/105°)^{\frac{1}{2}}=1.69$. Eq. (1) cannot be used to calculate $T_1$ or $T_2$ unless some assumption is made concerning the relative effectiveness of paraffin at 300°K and at 105°K in reducing the effective temperatures of the neutrons. The effective temperatures must be higher than the temperature of the paraffin. However, it does not seem probable that they are very much higher since with Eq. (1) this would lead to a much larger difference in neutron temperatures than the change in the temperature of the paraffin. Since $T_1$ cannot be less than 300°, $T_2$ cannot be less than 117° which corresponds to a mean energy of 0.015 ev. This difference in temperature will make a considerable difference in the fraction of the neutrons having energies greater than 0.023 ev, which is the minimum energy required to excite the O→1 transition, giving rise to inelastic scattering in $p-H_2$. We are therefore referring to the neutrons as ~300°K and ~120°K neutrons to emphasize the fact that the effective temperature of the cold neutrons is above the temperature of the howitzer.

Since the absorption of neutrons in boron depends upon their velocity, the fraction of the neutrons entering the $BF_3$ receiver that are counted depends upon the velocity distribution of the neutrons. The receiver used in this investigation would absorb and count about 40 percent of an ~300°K neutron energy distribution. In accordance with the $1/v$ law a larger percentage of the slower neutrons than of the faster ones should be counted. Since in each transmission measurement counts are taken both with and without the scattering sample in the beam, the detector will introduce an error only if the energy distribution of the neutrons is altered by passage through the scattering sample. Since the scattering is to some extent selective this will necessarily be the case. The calculation of this correction is difficult even when the velocity distribution of the neutrons is known. In our experiments the velocity distribution was not exactly known and no correction of this kind has been applied to the measurements. The correction, however, is believed to be small.

This uncertainty in energy distribution is unfortunately characteristic of experiments involving slow neutrons, and is almost always the chief limiting factor in the interpretation of results. It seems probable that the possibility of using parahydrogen as a neutron filter brought out by the present experiments may help to clear up this situation. The use of two separate cells for liquid hydrogen (suggested by Professor Teller) made it possible to study the effect of filtration. By placing parahydrogen in the first cell, a filtered beam was obtained which could then be investigated by placing scattering material in the second cell. As a matter of interest measurements were also made using a normal hydrogen filter, so that in all four different neutron energy groups were investigated. These are designated as ~300°K, ~120°K, ~120°K *para*-filtered, and ~120°K normal-filtered.

It is evident from Fig. 3 that the scattering cross section is sensitive both to hydrogen composition and to neutron temperature. The dependence of cross section on composition increases with decreasing neutron energy. The slope of the ~300°K neutron curve is least, that of the ~120°K neutron curve is greater, while the slope of the *para*-filtered neutron curve is still greater. To see that this last result fits into the picture it should be remembered that according to Schwinger and Teller's theory parahydrogen should be practically transparent for neutrons of less than 0.023 ev. Hence the effect

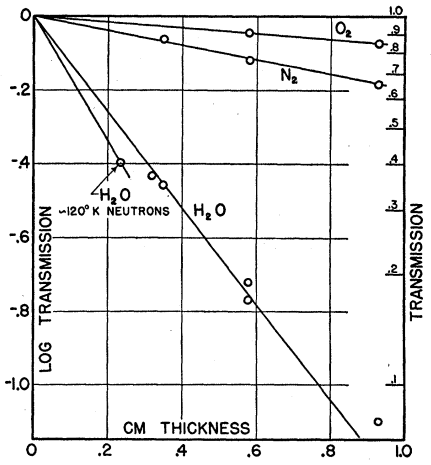


FIG. 4. Transmission of neutrons by water, liquid $O_2$ and liquid $N_2$. Neutron temperature ~300°K except as noted.

of *para*-filtration should be to remove high energy neutrons from the ~120°K beam and effectively cool it still further. The curve for normal-hydrogen-filtered neutrons is less interesting and more difficult to interpret. The 25 percent of parahydrogen in the filtering layer would tend to remove high energy neutrons from the beam, while the 75 percent of orthohydrogen would work somewhat in the opposite direction. Less weight is to be given to the dotted curves than to those for ~300°K and ~120°K neutrons, both because of the fewer experimental points involved, and because of the larger number of measurements required to determine a single point.

Figure 3 shows that the *para* cross section is consistently smaller than the *ortho* cross section. This one fact is sufficient to eliminate the possibility of an interaction independent of spin (case 1 discussed in the introduction) for if there were no spin dependence the differences should be in the opposite direction. The problem becomes therefore one of distinguishing between the two cases involving spin dependent forces, (2a) a real singlet deuteron level, and (2b) a virtual singlet deuteron level. Schwinger and Teller[4] have shown that the dependence of the *para* cross section on neutron energy will be very different in the two cases. If the singlet level is real the *para* cross section should be slightly greater (about 30 percent) for "liquid air" (0.012 ev) neutrons than for "room temperature" (0.037 ev) neutrons, while if the singlet level is virtual the cross sections for "liquid air" neutrons should be ~100 times smaller. Moreover if the singlet state should be real the orthohydrogen cross section would be only about 40 percent greater than the parahydrogen cross section for liquid air neutrons, whereas for a virtual state the difference may be very large. Fig. 3 shows (1) that there is a marked decrease in the *para* cross section with decreasing neutron energy and (2) that the ratio of the *ortho* to *para* cross sections for the ~120°K neutron distribution was 4 : 1. Our results therefore prove conclusively that the singlet state of the deuteron is virtual. This confirms the work of Halpern, Estermann, Simpson and Stern.[25] Taking a more general view of the matter it may be said that the virtual level hypothesis is the only one which leads to large differences in the *para* and *ortho* cross sections, and that none of the other possibilities can account for differences as large as those observed.

As Schwinger[5] pointed out, these results also require that the neutron spin be $\frac{1}{2}$ rather than $\frac{3}{2}$.

The presence of the tail of high energy neutrons unfortunately makes it impossible to gain any information concerning the range of the neutron-proton forces from our experiments. The fact that the observed cross section of parahydrogen for ~120°K neutrons is not as small as the theory predicts is to be attributed to these high energy neutrons, whose inelastic scattering completely masks the small elastic *para* scattering (0→0) which depends sensitively on the range of the neutron-proton forces.

It should be borne in mind that Schwinger and Teller's calculations give cross sections not as a function of temperature, but as a function of the energy of a monochromatic neutron beam. Also their calculations are strictly applicable only to hydrogen in the gaseous state, whereas our measurements apply to the liquid. These facts, together with the excess of high energy neutrons in the distributions with which we worked, are sufficient to account for the differences between our results and the predictions of Schwinger and Teller. Specifically, they are sufficient to account for the fact that the *ortho* cross section increases somewhat more rapidly with decreasing neutron "temperature" than was predicted, while the *para* cross section does not decrease as rapidly.

The possibility referred to earlier of using parahydrogen as a filter to obtain slow neutrons does not at first sight appear extremely promising. The ratio 29.0/17.6 found for the cross sections of parahydrogen for ~300°K and ~120°K neutrons is far from the 100 to 1 ratio[26] predicted by Schwinger and Teller for 0.037 and 0.012 ev neutrons. However, the mean energy ($3KT/2$) corresponding to 120°K is not 0.012 but 0.015 ev, and it must be remembered that 300°K and 120°K are to be interpreted not as actual temperatures but as lower limits. It seems reasonable

[25] J. Halpern, I. Estermann, O. C. Simpson and O. Stern, Phys. Rev. **52**, 142 (1937).

[26] As the authors point out this figure cannot be considered as a precise estimate, since it depends on the assumed range of the force of interaction.

therefore that a considerable number of the ~120°K neutrons should have energies greater than 0.023 ev, which is the point at which 0→1 scattering begins to take place and the *para* cross section begins to increase rapidly. It seems probable therefore that the ratio of *para* cross sections for 0.037 and 0.012 ev neutrons is actually much larger than 29.0/17.6. A layer of parahydrogen of suitable thickness should therefore transmit neutrons in the low energy range of a Maxwell distribution, and cut off rather sharply at an energy of 0.023 ev. Such a low energy beam should be quite useful in experimental work.

In the actual transmission measurements and calculations of cross sections, the chief errors involved are in cell thickness, composition of the hydrogen, and statistical fluctuations of the counts. Of these the largest is probably in the determination of cell thicknesses. Although this will affect the absolute values of the cross sections it will have very little influence on the relative values for different *ortho-para* compositions. Boiling may reduce the effective thickness of material in a cell. An estimate of the rate of heat transfer to the cells by radiation indicated that the amount of gas produced by evaporation in one second would not fill more than 2 percent of the cell volumes. We have observed in other experiments that the bubbles in liquid hydrogen are small and rise rapidly. Errors due to the presence of bubbles should be of the order of 1 percent. Uncertainty in the measurement of the composition is 1 percent. The number of counts was sufficient to make the probable error due to statistical fluctuations less than 2 percent. The combined error due to all the above factors certainly cannot be greater than 10 percent.

The results of the measurements on liquid $N_2$, $O_2$, and $H_2O$ are in substantial agreement with previous values.[8, 9, 27] Log transmission has been plotted against scattering thickness in Fig. 4. In obtaining proton cross sections from the water measurements the value $4.0\times10^{-24}$ cm$^2$ for the cross section of oxygen was taken from Table II. The same value was used for both the ~300°K and ~120°K neutrons since no information on the temperature dependence was available. The ratio of the proton cross sections in water thus obtained is $\sigma_{120}/\sigma_{300}=1.29$. Errors in these measurements are substantially the same as in the hydrogen measurements.

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[27] M. Goldhaber and G. H. Briggs, Proc. Roy. Soc. **A162**, 127 (1937).

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# The Isotopic Constitution of Strontium, Barium, Bismuth, Thallium and Mercury

ALFRED O. NIER*
*Research Laboratory of Physics, Harvard University, Cambridge, Massachusetts*

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A mass-spectrographic study of the isotopic constitution of Sr, Ba, Bi, and Tl has been made. Although the sensitivity and resolving power of the apparatus were sufficient to detect extremely rare isotopes, no new ones were found. Relative abundances of the isotopes of Sr, Ba, and Tl were determined. It was possible to show that if $Hg^{203}$ exists it must be with an abundance less than one-tenth of that at one time assigned to it by Aston.

ALTHOUGH a knowledge of the existence or nonexistence of rare stable isotopes is of great value in the study of nuclear physics, relatively few elements have been investigated with apparatus capable of detecting isotopes present in only small amounts. The present paper is a report on the continuation of a sys-

* National Research Fellow.