

## Scattering of Slow Neutrons by Ortho- and Parahydrogen\*

R. B. SUTTON,<sup>a</sup> T. HALL,<sup>b</sup> E. E. ANDERSON,<sup>c</sup> H. S. BRIDGE,<sup>d</sup> J. W. DEWIRE,<sup>e</sup>

L. S. LAVATELLI,<sup>f</sup> E. A. LONG,<sup>g</sup> T. SNYDER,<sup>h</sup> R. W. WILLIAMS<sup>i</sup>

*University of California, Los Alamos Scientific Laboratory, Santa Fe, New Mexico*

Measurements have been made of the scattering cross sections of ortho- and parahydrogen for neutrons of energies in the range 10°K to 30°K. Using the Schwinger-Teller theory the results have been correlated with the scattering cross section of free protons for slow neutrons and with the range of a square-well  $n-p$  potential. The measured cross sections show the energy dependence expected from the theory; they imply a free proton cross section of 19.7 barns and a force range of  $1.5 \times 10^{-13}$  cm, the first value agreeing within experimental error with the accepted figure, but the second falling significantly below the result  $2.8 \times 10^{-13}$  cm obtained by Breit, Thaxton and Eisenbud for the  $p-p$  force. The singlet state of the deuteron is again shown to be virtual, and the possibility of a neutron spin of  $\frac{3}{2}$  is eliminated since the data in this case would imply a free proton cross section of only 7 barns.

### 1. INTRODUCTION

THE values of the total scattering cross sections of ortho- and para-hydrogen give information about the range of the neutron-proton force, the dependence of this force on the relative orientation of the spins of the neutron and proton, and the binding energy of the singlet state of the deuteron; also the scattering cross section for neutrons by free protons can be obtained.<sup>1</sup>

The scattering cross section of slow neutrons by free protons can be written as the sum of two terms

$$\sigma = \frac{3}{4}\sigma_1 + \frac{1}{4}\sigma_0,$$

where  $\sigma_1 = 4\pi a_1^2$  and  $\sigma_0 = 4\pi a_0^2$ . The scattering cross section for the case of parallel spins is  $\sigma_1$  and for the case of anti-parallel spins is  $\sigma_0$ . The

scattering amplitudes,  $a_1$  and  $a_0$ , are given by

$$\begin{aligned} a_1 &= k^{-1} \sin \delta_{0,1}, & a_0 &= k^{-1} \sin \delta_{0,0}, \\ k &= (\frac{1}{2}ME_1)^{1/2}/\hbar, \\ E_1 &= \text{neutron energy (laboratory system)}, \\ M &= \text{neutron mass.} \end{aligned}$$

The  $\delta$ 's are the phase shifts of the scattered neutron wave with respect to the incident wave, and depend on the form of the neutron-proton interaction. With the assumption of a square-well form of the potential function representing the neutron-proton interaction, each phase shift can be expressed in terms of the width,  $a$  (assumed the same for the singlet and triplet interaction) and the depth of the well, or in terms of  $a$  and the binding energy of the deuteron state in question. Thus a determination of  $a_1$  will yield a value of  $a$  since the binding energy of the deuteron in the triplet state is known. The relative signs of  $a_1$  and  $a_0$  indicate whether the singlet state of the deuteron is real or virtual. The value of  $a_0$  and of  $a$  will give the binding energy in the singlet state. As is shown in reference 1, when scattering occurs from hydrogen molecules the cross section per molecule is a combination of  $a_1$  and  $a_0$  which differs depending on whether the scattering occurs from ortho- or parahydrogen. Thus a knowledge of the ortho- and parahydrogen cross sections can be used to obtain  $a_1$  and  $a_0$ .

Early experiments by Halpern, Estermann, Simpson and Stern<sup>2</sup> and by Brickwedde, Dun-

\* This paper is based on work performed under Contract No. W-7405-Eng-36 with the Manhattan Project at the Los Alamos Scientific Laboratory of the University of California.

<sup>a</sup> Now at Carnegie Institute of Technology, Pittsburgh, Pennsylvania.

<sup>b</sup> Now at University of Chicago, Chicago, Illinois.

<sup>c</sup> Now at Milwaukee-Downer College, Milwaukee, Wisconsin.

<sup>d</sup> Now at Massachusetts Institute of Technology, Cambridge, Massachusetts.

<sup>e</sup> Now at Cornell University, Ithaca, New York.

<sup>f</sup> Now at Harvard University, Cambridge, Massachusetts.

<sup>g</sup> Now at University of Chicago, Chicago, Illinois.

<sup>h</sup> Now at General Electric Research Laboratories, Schenectady, New York.

<sup>i</sup> Now at Massachusetts Institute of Technology, Cambridge, Massachusetts.

<sup>1</sup> J. Schwinger and E. Teller, Phys. Rev. **52**, 286 (1937).

<sup>2</sup> J. Halpern, I. Estermann, O. C. Simpson, and O. Stern, Phys. Rev. **52**, 142 (1937).

ning, Hoge, and Manley<sup>3</sup> on the scattering of slow neutrons by various ortho-para-concentrations of liquid hydrogen showed the ortho-hydrogen cross section to be several times larger than the para-hydrogen cross section. From these results it was shown that the neutron-proton force was spin dependent and that the singlet state of the deuteron was virtual. However, the neutron energies used were high enough to cause para- to ortho-hydrogen conversion so that the elastic para-cross section could not be determined. Further, the liquid used for the scattering material may have introduced effects due to intermolecular forces. Therefore the experiment was repeated by Alvarez and Pitzer<sup>4</sup> using the time-of-flight technique to obtain very slow monochromatic neutrons, and using a 40 cm path of hydrogen gas as a scattering material. The ortho- and para-hydrogen cross sections were obtained for 20°K neutrons by measurement of the transmission through gas of normal ortho-para-concentration (75 percent, 25 percent) and through 99.8 percent pure parahydrogen; the gas in each case was kept at 20.4°K. Results of this experiment gave  $\sigma_o' = 100 \pm 3 \times 10^{-24}$  cm<sup>2</sup> and  $\sigma_p' = 5.2 \pm 0.6 \times 10^{-24}$  cm<sup>2</sup> per molecule (where the

primes refer to values not corrected for Doppler effect, and the subscripts *o* and *p* refer to ortho- and para-hydrogen respectively). Thus the results and conclusions from the previous experiments were verified. However, an analysis of these data by Schwinger<sup>5</sup> gave a value of *a* for a rectangular-well of either zero or  $8 \times 10^{-13}$  cm, and a free proton scattering cross section of  $16.6 \times 10^{-24}$  cm<sup>2</sup>. Both of these results were in disagreement with existing values. The measured neutron-proton scattering cross section is about  $20 \times 10^{-24}$  cm<sup>2</sup>; the proton-proton force range which might have been expected here is  $2.8 \times 10^{-13}$  cm. Because of these discrepancies it was decided to reinvestigate the problem.

## 2. EXPERIMENTAL METHOD AND APPARATUS

The experimental method used was that of measuring the transmission of neutrons through samples of ortho- and para-hydrogen gas at about 20°K.

The following considerations affect the choice of scatterer and of neutron energy. In order to avoid effects which might arise due to intermolecular forces in the scattering material, it is preferable to have the hydrogen in the gaseous state. The energy of the neutrons with respect to the hydrogen molecule must be below 0.023 ev (260°K) to avoid inducing para- to ortho-transitions, the cross section for this process being many times the para-elastic cross section. As a result, the scatterer used was hydrogen gas in a tube at the temperature of liquid hydrogen boiling at a pressure of about 60 cm of mercury. The range of neutron energies used was from 0.0008 ev ( $\sim 10^\circ\text{K}$ ) to 0.0025 ev ( $\sim 30^\circ\text{K}$ ).

The scattering chamber assembly is shown in Fig. 1. A central brass cylinder, 4 in. diameter, 3 meters long, was sealed at the ends with 0.015 in. thick stainless steel windows and was surrounded by a concentric brass cylinder of the same length and 5 in. diameter. The central cylinder contained the scattering gas, and the shell between the two cylinders was filled with the refrigerant liquid hydrogen. This assembly was in turn mounted inside a radiation shield, consisting of a brass tube of  $7\frac{1}{2}$  in. diameter, wrapped with  $\frac{1}{2}$  in. copper tubing, through which liquid air was circulated. The outside vacuum

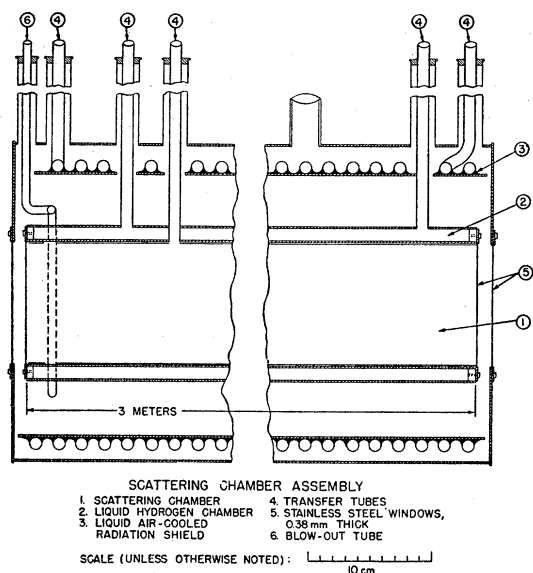


FIG. 1.

<sup>3</sup> F. G. Brickwedde, John R. Dunning, Harold J. Hoge, and John H. Manley, *Phys. Rev.* **54**, 266 (1938).

<sup>4</sup> Luis W. Alvarez and Kenneth S. Pitzer, *Phys. Rev.* **58**, 1003 (1940).

<sup>5</sup> J. Schwinger, *Phys. Rev.* **58**, 1004 (1940).

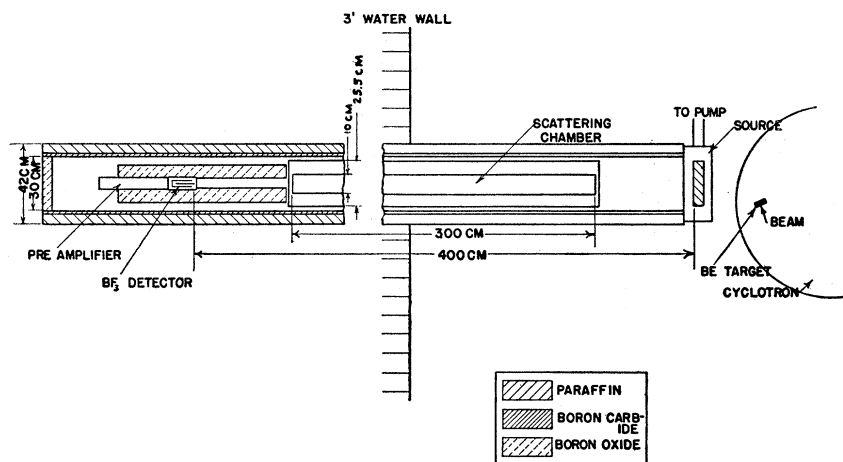


FIG. 2. Experimental arrangement. (In the legend for Fig. 2, shading lines for paraffin should be rotated through  $90^\circ$ . The shading in the figure should continue to the right-hand side of the diagram.)

case, through which the transfer and lead-in tubes passed, was a brass cylinder of 10 in. diameter. The inner containers were supported by the transfer tubes, as shown, and by appropriately placed micarta spacers. The design was very similar to that used by Alvarez and Pitzer,<sup>4</sup> but gave a much longer path length.

The thermal insulation was remarkably good. Despite the large size of the assembly, it was possible with fifty liters of liquid hydrogen to cool the scattering chamber to liquid hydrogen temperatures and to maintain very steady thermal conditions for a period of 15 to 20 hours.

Neutrons of high energy were produced by bombarding a beryllium target with deuterons in the 42-inch Los Alamos cyclotron. These neutrons were slowed down in paraffin placed as near to the target as possible (see Fig. 2). In order to increase the number of subthermal neutrons the paraffin source was cooled to liquid air temperature. This increased the yield of these neutrons by about a factor of four over the yield from the paraffin at room temperature; it also shifted the maximum of the number-velocity distribution from 3300 meters per second to 2000 meters per second.<sup>5a</sup> The paraffin moderator was 5 cm thick and was supported below a liquid air chamber; copper cooling vanes attached to the chamber passed through and

around the paraffin. The assembly was mounted in a vacuum chamber. This thickness of paraffin has been found to give maximum slow neutron intensity and have only about 100  $\mu$ sec. neutron half-life when at room temperature. It was assumed that the lifetime could be only lower when the paraffin was at liquid air temperature.

As shown in Fig. 2, the paraffin source was at one end of a collimator, inside of which the scattering chamber and detector were placed. The purpose of the collimator was to make certain that the only neutrons reaching the detector came from the source and went through the scatterer. The collimator was constructed of a layer of  $B_4C$  (1.3 g per  $cm^2$ ) surrounded by 5 cm of paraffin. To prevent neutrons which were scattered in the liquid hydrogen surrounding the scattering chamber from reaching the detector, the scattering chamber was lined with 0.030-inch cadmium sheet. Further collimation was provided by a cadmium diaphragm between the source and scattering tube; the opening in the diaphragm was slightly smaller than the windows in the scattering tube. The detector was surrounded by a  $B_2O_3$  collimator of  $3\frac{1}{2}$  in. wall thickness. To demonstrate the adequacy of the shielding, the exit window of the scattering tube was covered with cadmium. No neutrons in the  $10^\circ K$  to  $30^\circ K$  region were observed.

The time of flight of the neutrons was measured by a modulation method, similar to that described by Baker and Bacher,<sup>6</sup> by Bacher,

<sup>5a</sup> The characteristics of liquid hydrogen as a moderator were also investigated. By the use of this the neutron intensity in the  $20^\circ K$  region was increased by about a factor of ten over the intensity from room temperature paraffin. The maximum of the number-velocity curve occurred at about 1540 meters per second.

<sup>6</sup> C. P. Baker and R. F. Bacher, Phys. Rev. **59**, 332 (1941).

Baker and McDaniel,<sup>7</sup> and by B. D. McDaniel.<sup>8</sup> The modulation equipment and BF<sub>3</sub> detector were constructed at Cornell University and are described in reference 8.

The beam modulation required for the time-of-flight measurement was obtained by applying the voltage to the cyclotron arc in the form of square pulses. Even when this voltage was off, ions were accelerated in the cyclotron. This resulted in a neutron background of uniform intensity around the modulation cycle. Although this intensity was not serious for measurements on room temperature neutrons, where the neutron intensity was high, it was necessary to remove this background in the present experiment since the intensity of 20°K neutrons was quite low. This was accomplished by also modulating the cyclotron dee-voltage so that acceleration could take place only during a 300 μsec. time interval which began at the time the arc was turned on.

In an experiment such as this one there is possibility of error arising from the counting of scattered neutrons. The amount of the effect will depend on the geometry of the experiment. Two types of such error were considered. One may be due to neutrons which scatter elastically through a very small angle and hence reach the detector at their proper time. Another may be due to neutrons initially of energy too high to arrive at the detector at a time such that they would be counted, but which are slowed down by an inelastic collision to such a velocity that they may arrive at a time when pulses are being recorded. Calculation showed that the first type of effect gives a ratio of elastically scattered to transmitted intensity of about 10<sup>-3</sup> for the geometry used, and the second type gives a ratio of about 10<sup>-6</sup>.

The transmission of the scattering gas for any one of the time-of-flight (energy) intervals was obtained by the following method. The number of counts  $C_A$  occurring in the given energy interval and the total number of counts  $T_A$  over the whole cycle (i.e., for all energies) were each recorded for a number of runs with the scattering gas in the chamber. Similar readings,  $C_B$  and  $T_B$ ,

were taken for runs with no scattering gas in the chamber.

The transmission of the gas was then given by

$$\frac{C_A}{T_A} \div \frac{C_B}{T_B} \times N,$$

where  $N$  is a normalization factor equal to the ratio of  $T_A$  to  $T_B$  each for the same neutron flux incident on the scatterer. This method thus eliminates effects of changes in sensitivity of the BF<sub>3</sub> counting system during a run.

$C$  (for each of the twelve channels) and  $T$  were recorded for each of several runs of about one-half to one hour duration with the scattering gas in. The number of counts from a stable neutron monitor (fission detector) was also recorded for each run. The gas was pumped out and several blank runs were taken. If the sensitivity of the BF<sub>3</sub> system was stable,  $N$  could be obtained from the total counts and the monitor counts. In order to eliminate effects of possible changes in sensitivity in the counting system  $N$  was also obtained by measuring the ratio of total counts to monitor counts for several two-minute intervals, and measuring the sensitivity of the BF<sub>3</sub> system between each interval with an  $\alpha$ -Be source. This was done with the scatterer in and with the scatterer out. If we call the ratio

$$\frac{(\text{Total counts/Monitor counts})}{\text{Total counts per sec.}}$$

(the counts in the numerator being due to the beam, those in the denominator due to the  $\alpha$ -Be source)  $A$  for scatterer in and  $B$  for scatterer out, then  $N$  is given by  $A/B$ .

The transmission is related to cross section in the following way:

$$T = e^{-nl\sigma_t} \quad (1)$$

where  $n$  = number of hydrogen molecules per cc,  $l$  is the length of the scattering path and  $\sigma_t$  is given by

$$\sigma_t = f_o\sigma_o + f_p\sigma_p + \sigma_c, \quad (2)$$

$f_o$  is the fraction of hydrogen molecules in the ortho-state,  $f_p$  the fraction in the para-state,  $\sigma_o$  and  $\sigma_p$  are the scattering cross sections of ortho- and para-hydrogen respectively, and  $\sigma_c$  is the capture cross section per molecule. Thus in order to determine  $\sigma_o$  and  $\sigma_p$  it is necessary to

<sup>7</sup> R. B. Bacher, C. D. Baker, and B. D. McDaniel, Phys. Rev. **69**, 443 (1946).

<sup>8</sup> B. D. McDaniel, Phys. Rev. **70**, 832 (1947).

know  $f_o$  and  $f_p$ , for two different concentrations of scattering gas, and  $\sigma_o$ ,  $\sigma_e$  at the neutron energies used was obtained by extrapolation of the measured value for 0.025 ev, assuming  $\sigma_e$  varied inversely with neutron velocity. The value used for 0.025 ev neutrons was  $0.62 \times 10^{-24}$  cm<sup>2</sup> per molecule.<sup>9</sup>

One scattering sample was 99.9 percent parahydrogen. To obtain this sample, liquid hydrogen was condensed over activated charcoal for a period of about 48 hours. Measurements of the thermal conductivity of the gas evaporated from this liquid were made at intervals of time, by the method described in Farkas and Farkas.<sup>10</sup> When the conductivity showed no further change with time it was assumed that equilibrium had been reached. At this point  $f_p = 0.999$  and  $f_o = 0.001$ . Samples of gas were taken when the parahydrogen had been introduced into the scattering chamber, and also when the scattering runs were completed. These samples showed the same conductivity as a sample taken from the converter. Thus it was concluded that the

concentration had remained unchanged during the experiment.

For the second concentration normal hydrogen gas was used. The equilibrium concentration at room temperature in this case is  $f_o = 0.75$  and  $f_p = 0.25$ . It was apparent that when the gas was kept in the scattering chamber at liquid hydrogen temperatures, conversion from ortho- to parahydrogen took place. The rate of conversion was such that over a period of about six hours  $f_o$  went from 0.75 to 0.45. Several runs, each of about one-half hour duration, were taken with each filling of scattering gas.  $N$  as a function of time was obtained by measuring the ratio  $A$  (defined above) at several times while each filling was in the scattering chamber. The value of  $N$  used for each run was that corresponding to the time half-way through the run in question.

The rate of conversion from ortho- to parahydrogen could be obtained in two ways. First, samples of gas were taken from the scattering chamber at the conclusion of the series of runs and  $f_o$  is measured. (The concentration at the

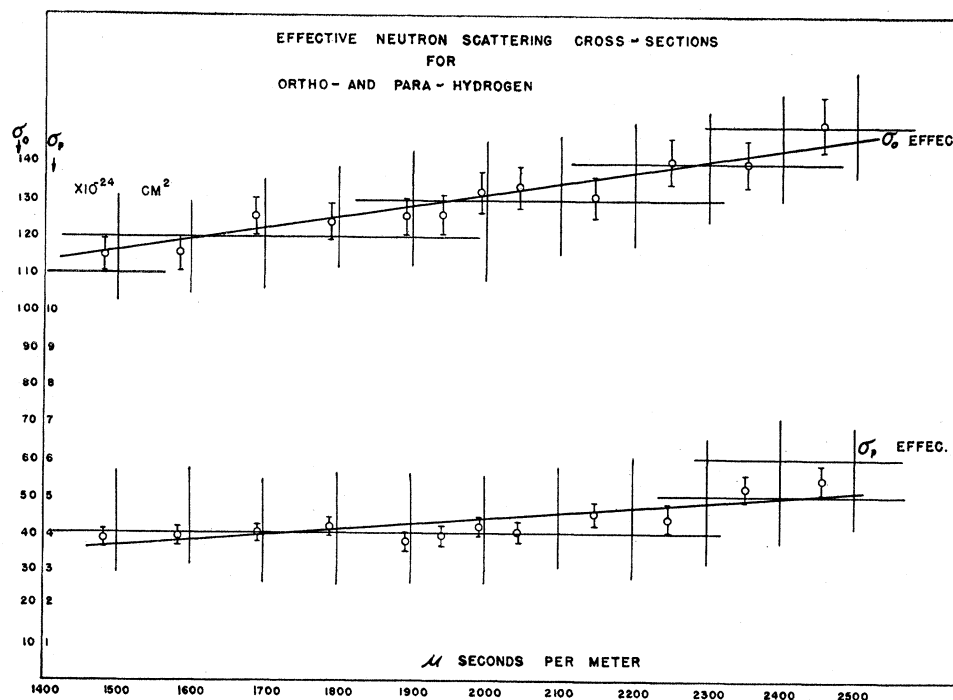


FIG. 3.

<sup>9</sup> This value was obtained from work done on the Manhattan Project.

<sup>10</sup> A. Farkas and L. Farkas, *Orthohydrogen, Parahydrogen and Heavy Hydrogen* (Cambridge University Press, 1935).

time of filling the scattering tube was  $f_o = 0.75$ .) Since conversion is expected to occur at an exponential rate,<sup>10</sup>  $f_o$  at any time could be found. A second method for determining  $f_o$  vs. time was possible by making use of the variation of the transmission with time. The experimental results indicated that  $\log \sigma_t$  vs. time was a straight line (actually the sum of the values of  $\log \sigma_t$  for each of the twelve time-of-flight regions was plotted). This is to be expected since  $\sigma_p$  is so much smaller than  $\sigma_o$ . Since Eq. (2) can be written as

$$d(\log f_o)/dt = (1 + (\sigma_p + \sigma_c)/(\sigma_o - \sigma_p)f_o)d(\log \sigma_t)/dt \quad (3)$$

the slope of  $\log f_o$  vs. time could be obtained. (Since the term on the right-hand side of the equation contains  $f_o$  and since  $\log f_o$  vs. time should be a straight line, it is not accurately true that  $\log \sigma_t$  vs. time should be a straight line. However, the term  $(\sigma_p + \sigma_c)/(\sigma_o - \sigma_p)f_o$  has a value only about 0.09, so that changes in  $f_o$  such as occurred from the first to the last run in a series made on one gas filling can change the value of the term in the brackets by only about 5 percent. This leads to a maximum error of 2 percent in  $f_o$  obtained from the curve of  $f_o$  vs. time in this manner.)

Unfortunately the two determinations did not agree; in fact the values of  $f_o$ , corresponding to seven hours in the scattering chamber, given by the two methods differed by as much as 20 percent.

The values used for  $f_o$  were obtained from the transmission data. This method was assumed to give more nearly correct results since not only did this method give the expected time variation of  $f_o$  (whereas only the end points could be obtained by the conductivity method) but also  $f_o$  was essentially determined from the entire

sample of gas, whereas the thermal measurements were made on a small sample taken from the chamber. These samples could lead to error if, for example, the ortho-para-concentration was not uniform in the scattering chamber.

In the case of the para-measurement the results given were obtained in five runs on one filling of para-hydrogen, and in two runs with no gas in the chamber. With gas in, the lowest number of counts in one channel was 1000, the highest 14,000. With gas out, these were 800 and 10,800 respectively. The pressure of gas used was about 55 cm Hg. The transmission measured varied from 0.58 to 0.45 over the energy region used. The cross section is shown in Fig. 3. The errors shown are made up of the probable errors obtained from the number of counts and the errors in  $A$  and  $B$ , obtained from the mean square deviations in the separate values (2.9 percent). The value of  $\sigma$  plotted have not been corrected for Doppler effect due to the thermal motion of the  $H_2$  molecules.

In the ortho-measurement, two fillings of the scattering chamber were used. On one filling three runs were taken, the lowest and highest number of counts recorded being 640 and 10,000 respectively. On the other filling five runs were taken, the corresponding counts being 880 and 13,000. Six runs were taken with no gas in the chamber. The lowest and highest number of counts were 2000 and 25,000 respectively. Gas pressures of about 7 cm Hg were used; the transmissions were about 0.40. The cross sections obtained from the two sets of runs differed by only 2 percent. Figure 3 shows the average of both sets. The errors shown consist of the probable errors from the number of counts, the errors in  $A$  and  $B$  (1.5 percent), and an estimated error of 2 percent in  $f_o$ .

TABLE I. Theoretical dependence of effective cross section on energy.

$$\sigma'_{ortho} = (2k'_{1,1} + k'_{1,0})(a_1 - a_0)^2 + k'_{1,1}(3a_1 + a_0)^2;$$

$$\sigma'_{para} = k'_{0,0}(3a_1 + a_0)^2.$$

Neutron energy, ev	$k'_{1,1}$	$k'_{0,0}$	$k'_{1,0}$	$2k'_{1,1} + k'_{1,0}$
0.0008673	7.702	7.696	2.158	17.563
0.001250	6.953	6.946	1.853	15.760
0.001463	6.685	6.675	1.741	15.112
0.001723	6.435	6.423	1.637	14.506
0.001830	6.351	6.338	1.601	14.302
0.002386	6.012	6.000	1.460	13.484

### 3. INTERPRETATION OF RESULTS

The theory of slow neutron scattering in ortho- and para-hydrogen is given in reference (1), where it is shown that

$$\begin{aligned} \sigma_{000;000} &= k_{0,0}(3a_1 + a_0)^2, \\ \sigma_{000;101} &= k_{1,0}(a_1 - a_0)^2, \\ \sigma_{101;101} &= k_{1,1}[(3a_1 + a_0)^2 + 2(a_1 - a_0)^2], \end{aligned} \quad (4)$$

where  $\sigma_{J'v's';Jvs}$  is the cross section for the

TABLE II. Values of free proton cross section  $\sigma_f$  and force range  $a$  implied by observed values  $\sigma'_{\text{ortho}}$  and  $\sigma'_{\text{para}}$ .

Neutron energy, ev	Obs. $\sigma'_{\text{ortho}}$ , barns	Obs. $\sigma'_{\text{para}}$ , barns	$(3a_1+a_0)^2$ , barns	$(a_1-a_0)^2$ , barns	$-a_1$ , $10^{-12}$ cm	$a_0$ , $10^{-12}$ cm	$\sigma_f$ , barns	$a$ , $10^{-12}$ cm
0.0008673	145	4.98	0.647	7.97	0.505	2.32	19.3	1.33
0.001250	133	4.40	0.634	8.10	0.510	2.33	19.6	1.43
0.001463	128	4.19	0.628	8.19	0.518	2.34	19.8	1.58
0.001723	124	3.97	0.618	8.20	0.520	2.34	19.8	1.61
0.001830	122	3.90	0.615	8.25	0.522	2.35	19.9	1.65
0.002386	116	3.58	0.597	8.26	0.526	2.35	19.9	1.72

process in which the  $\text{H}_2$  molecule goes from rotational level  $J$  to  $J'$ , vibrational level  $v$  to  $v'$  and spin state  $s$  to  $s'$ ; the  $k_{v',i}$  are functions of the neutron energy given explicitly in reference (1), Eqs. (38) and (40), and  $a_1$  and  $a_0$  are the amplitudes of the triplet and singlet, scattered waves, respectively.

However, because of the thermal motion of the  $\text{H}_2$  molecules, one measures not the actual cross section at the energy of the neutron beam, but an average of the cross section weighted over an energy range; i.e., for a beam of neutrons of velocity  $v$  the measured cross section  $\sigma'$  is

$$\sigma'(v) = \frac{1}{v} \int_{\underline{u}} |v-\underline{u}| \sigma(|v-\underline{u}|) N(\underline{u}) d\underline{u}, \quad (5)$$

where  $N(u)$  is the distribution of the molecular velocity  $u$ . Hamermesh and Schwinger<sup>11</sup> have obtained expressions for the quantities  $\sigma'$  by expanding the functions  $k_{v',i}$  as power series in the neutron energy  $E_1$ , substituting the early terms of the series into (5) and integrating exactly. They thus obtain a new set of functions  $k'_{v',i}$  which, replacing the  $k_{v',i}$  in (4) will give the *effective* cross section  $\sigma'_{J',v',s';Jv s}$ . The coefficients  $k'_{v',i}$  have been calculated for the energies of this experiment using the Schwinger-Hamermesh expressions; the values are listed in Table I. The temperature of the  $\text{H}_2$  gas was taken to be 19.5°K and the average separation  $\bar{r}$  of the protons in the  $\text{H}_2$  molecule to be  $0.765 \times 10^{-8}$  cm.<sup>12</sup> We are grateful to Drs. Hamermesh and

Schwinger, who provided us with the expressions described in reference (11) and in addition recalculated the coefficients at neutron energy 20°K, and to Mr. Benjamin Sussholz who carried out an independent evaluation of the quantities  $k'_{0,0}$ , using expressions of a different form, thus insuring the results of a rather involved calculation.

In this experiment, the  $\text{H}_2$  gas, at the low temperature of 20°K, is all initially at the zero vibrational level and zero or first rotational level, the zero rotational level occurring only with the para spin state and the first rotational level only with the ortho spin state. In addition, the neutrons to be counted are all too slow to produce the rotational or vibrational level changes  $0 \rightarrow 1$  or the rotational change  $1 \rightarrow 2$ . For these reasons the only possible collision processes are those of Eq. (4). Accordingly for these measurements

$$\begin{aligned} \sigma'_{\text{para}} &= \sigma'_{000;000} = k'_{0,0}(3a_1+a_0)^2, \\ \sigma'_{\text{ortho}} &= \sigma'_{000;101} + \sigma'_{101;101} \\ &= (2k'_{1,1} + k'_{1,0})(a_1-a_0)^2 + k'_{1,1}(3a_1+a_0)^2. \end{aligned} \quad (6)$$

Substitution of the experimental results into (6) yields values of  $(a_1-a_0)^2$  and  $(3a_1+a_0)^2$ . These imply a value for the free proton cross

TABLE III. Values of  $\sigma'_{\text{ortho}}$  and  $\sigma'_{\text{para}}$  implied by  $\sigma_f = 20.8$  barns and  $a = 2.8 \times 10^{-12}$  cm.\*

Neutron energy, ev	Implied $\sigma'_{\text{para}}$ , barns	Implied $\sigma'_{\text{ortho}}$ , barns
0.0008673	2.86	155
0.001250	2.58	139
0.001463	2.48	134
0.001723	2.39	128
0.001830	2.36	127
0.002386	2.23	119

\* Implied values are:  $-a_1 = 0.584 \times 10^{-12}$  cm;  $a_0 = 2.362 \times 10^{-12}$  cm;  $(a_1-a_0)^2 = 8.679$  barns;  $(3a_1+a_0)^2 = 0.372$  barns;  $\sigma'_{\text{ortho}} = 8.679 \times (2k'_{1,1} + k'_{1,0}) + 0.372k'_{1,1}$ ;  $\sigma'_{\text{para}} = 0.372k'_{0,0}$ .

<sup>11</sup> M. Hamermesh and J. Schwinger, Phys. Rev. **55**, 679 (1939).

<sup>12</sup> The exact procedure for calculating the  $k'_{v',i}$  is to substitute average values for  $r$ ,  $r^2$ , etc. as they occur in the theoretical formula; formulae obtained in this way are given in a forthcoming paper by Hamermesh and Schwinger in Phys. Rev. In the present paper,  $r$  has been replaced by  $\bar{r}$  wherever it occurs, with a consequent error of about  $\frac{1}{3}$  percent in the  $k'_{v',i}$ .

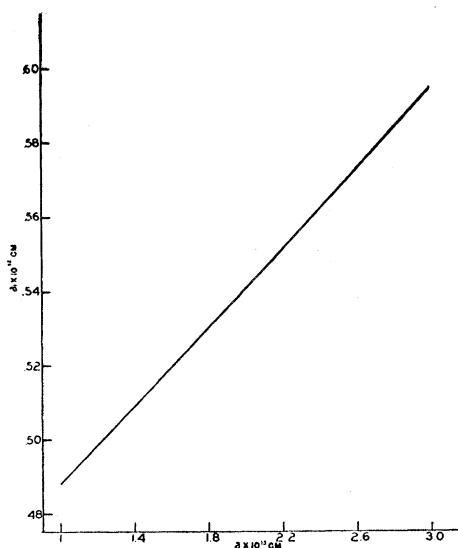


FIG. 4. Theoretical dependence of amplitude  $a_1$  on range  $a$ .

section  $\sigma_f$  since

$$\begin{aligned}\sigma_f &= \frac{3}{4}\sigma_1 + \frac{1}{4}\sigma_0 \\ &= \frac{3}{4}(4\pi a_1^2) + \frac{1}{4}(4\pi a_0^2) \\ &= \frac{\pi}{4}[(3a_1 + a_0)^2 + 3(a_1 - a_0)^2].\end{aligned}\quad (7)$$

In order to obtain an implied value for the force range, we also get from Eqs. (6) values for  $a_1$  and  $a_0$  themselves, and then use the theoretical relation between  $a_1$  and  $a$  as given by Breit and Kittel.<sup>13</sup> Of the four sets of solutions to Eqs. (6), the two with positive  $a_1$  are ruled out because the triplet state, being real, must have a negative  $a_1$ ,<sup>14</sup> and a third solution is ruled out because the implied value of the force range is much too large (8 or  $9 \times 10^{-13}$  cm). The remaining solution may be substituted into the theoretical relation, which, for slow neutrons (of energy much less than the deuteron binding energy), is simply<sup>13</sup>

$$\alpha^2 a_1^2 = 1 + \alpha a + 0.3447\alpha^2 a^2 + 0.0246\alpha^3 a^3 - 0.0117\alpha^4 a^4 + \dots \quad (8)$$

where  $\alpha = (M|E_0|/\hbar^2)^{1/2} = 2.29 \times 10^{12}$  cm<sup>-1</sup>,  $M$  = neutron mass, and  $E_0$  = binding energy (triplet

state). The relation (8) between amplitude and force range is shown in Fig. 4.

Given any two of the values  $\sigma'_{ortho}$ ,  $\sigma'_{para}$ ,  $\sigma_f$  and  $a$ , one may infer the remaining two by use of Eqs. (6), (7), and (8). Thus in Table II the data of this experiment are used to predict  $\sigma_f$  and  $a$ .  $\sigma_f$  compares well with the directly measured cross section  $21 \times 10^{-24}$  cm<sup>2</sup>.<sup>15</sup>  $a$  appears to differ from the value found for the  $p$ - $p$  force range which was found to have a most probable value  $2.8 \times 10^{-13}$  cm, and to fall definitely within the interval  $2.8 \times 10^{-13}$  cm  $\pm$  25 percent.<sup>16</sup>

One may prefer to consider the correlation in the opposite manner; in Table III and Figs. 5 and 6 the accepted numbers  $\sigma_f = 20.8$  barns and  $a = 2.8 \times 10^{-13}$  cm are used to predict  $\sigma'_{ortho}$  and  $\sigma'_{para}$ ; the same is done in Table IV and Figs. 5 and 6 for the values  $\sigma_f = 19.7$  barns and  $a = 1.54 \times 10^{-13}$  cm, which provide the best fit for our data. Tables II-IV and Figs. 5 and 6 together give fully the relations between theory and experiment for these two pairs of quantities.

#### 4. DISCUSSION

The implied value of the free proton cross section agrees within experimental error with direct measurements. However, the implied force range cannot be brought into coincidence with the  $p$ - $p$  measurements.

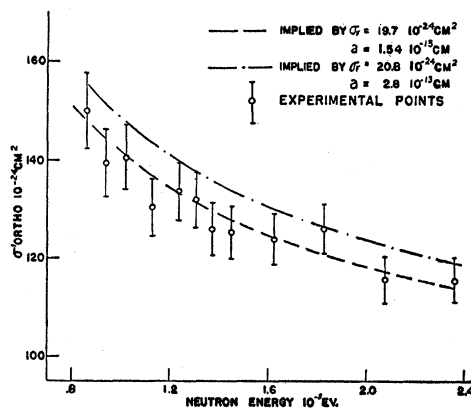


FIG. 5. Effective orthohydrogen cross section.

<sup>15</sup> Henry B. Hanstein, Phys. Rev. 59, 489 (1941).

\*\* This value is in good agreement with that of  $20.8 \times 10^{-24}$  cm<sup>2</sup> which was based on measurements by members of the Manhattan Project.

<sup>16</sup> Breit, Thaxton, and Eisenbud, Phys. Rev. 55, 1018 (1939).

<sup>13</sup> Breit and Kittel, Phys. Rev. 56, 744 (1939).

<sup>14</sup> E. Fermi, Ricerca Scient. VIII-II, 13 (1936).



Because of the smallness of  $(3a_1+a_0)^2$  as compared to  $(a_1-a_0)^2$ ,  $\sigma'_{\text{ortho}}$  and  $\sigma_f$  are dependent almost entirely on each other alone (see Eqs. (6) and (7)). Hence it is the ortho-measurement which gains added reliability from the check obtained with the well-established value  $\sigma_f$ , and any significant error in the implied range must be due to the observed  $\sigma'_{\text{para}}$ . A comparison of the  $\sigma'_{\text{para}}$  columns in Tables II, III, and IV shows that the observed  $\sigma'_{\text{para}}$  would have to be about 60 percent too large if the deviation from the  $p$ - $p$  range were caused in this way. One systematic error which could produce this discrepancy would be the contamination of the para-gas with ortho-. A contamination of about one to two percent would be sufficient to account for this. If a determination with a second para sample had been made, this possibility would perhaps have been reduced. If the hydrogen capture cross section does not vary inversely with neutron velocity over such a wide range of velocities, then the para-hydrogen cross section can be considerably different from the values given, since at the energies used the capture and scattering cross sections are of comparable magnitude.

The errors described in the experimental section add to about  $\pm 5$  percent for both para- and ortho-. Since the energy dependence of the cross sections is in good agreement with theory, we

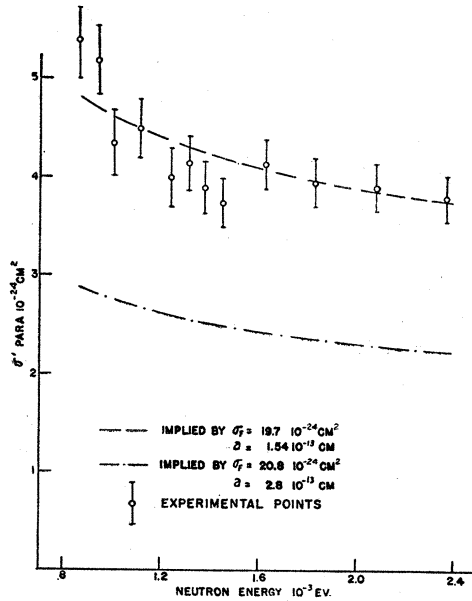


FIG. 6. Effective parahydrogen cross section.

TABLE IV. Values of  $\sigma'_{\text{ortho}}$  and  $\sigma'_{\text{para}}$  implied by  $\sigma_f = 19.7$  barns and  $a = 1.54 \times 10^{-12}$  cm.\*

Neutron energy, ev	Implied $\sigma'_{\text{ortho}}$ barns	Implied $\sigma'_{\text{para}}$ barns
0.0008673	148	4.82
0.001250	133	4.35
0.001463	127	4.18
0.001723	122	4.02
0.001830	121	3.97
0.002386	114	3.76

\* Implied values are:  $-a_1 = 0.516 \times 10^{-12}$  cm;  $a_0 = 2.33_2 \times 10^{-12}$  cm;  $(a_1 - a_0)^2 = 8.151$  barns;  $(3a_1 + a_0)^2 = 0.626$  barns;  $\sigma'_{\text{ortho}} = 8.151 \times (2k'_{1,1} + k'_{1,0}) + 0.626k'_{1,1}$ ;  $\sigma'_{\text{para}} = 0.626k'_{0,0}$ .

may obtain the limits of the implied range by giving a spread of  $\pm 5$  percent (which will be about three times the probable error) to the average values of  $(a_1 - a_0)^2$  and  $(3a_1 + a_0)^2$  listed in Table II. This permits a maximum square-well range of  $1.99 \times 10^{-13}$  cm and a minimum of  $1.12 \times 10^{-13}$  cm.

The results indicate that the neutron spin must be  $\frac{1}{2}$  rather than  $\frac{3}{2}$ . The scattering theory was generalized to arbitrary neutron spin by Schwinger;<sup>17</sup> for neutron spin  $\frac{3}{2}$  his formulae reduce to

$$\sigma'_{\text{para}} = \frac{1}{4}k'_{0,0}(5a_2 + 3a_1)^2$$

and

$$\sigma'_{\text{ortho}} = \frac{1}{4}k'_{1,1}[(5a_2 + 3a_1)^2 + 10(a_2 - a_1)^2] + 20k'_{1,0}(a_2 - a_1)^2 \quad (9)$$

while

$$\sigma_f = \frac{\pi}{16}[(5a_2 + 3a_1)^2 + 15(a_2 - a_1)^2] \quad (10)$$

where  $a_2$  and  $a_1$  are the amplitudes of the hypothetical quintuplet and triplet scattered waves respectively. Our results for 20°K neutrons, substituted into (8), imply a free proton scattering cross section of only 7 barns, so that a neutron spin of  $\frac{3}{2}$  is incompatible with these data.<sup>18</sup>

## 5. CONCLUSION

The observed values of the ortho- and para-hydrogen scattering cross sections generally confirm the Schwinger-Teller theory; they are consistent with a free proton cross section of about

<sup>17</sup> J. Schwinger, Phys. Rev. 52, 1250 (1937).

<sup>18</sup> Similar measurements in reference (5) indicated that the neutron spin is  $\frac{1}{2}$ ; however at that time  $\sigma_1$  was believed to be 14 barns, which affected the treatment of the correlation.

21 barns, and for an assumed square-well  $n$ - $p$  interaction they imply a force range of  $1.54 \times 10^{-13}$  cm, experimental error allowing this value to lie between  $1.12$  and  $1.99 \times 10^{-13}$  cm.

#### 6. ADDITIONAL ACKNOWLEDGMENTS

We are very grateful to Professor Edward Teller for guidance in problems of the scattering

theory, and to Professor Robert Wilson, Mr. Anton Grubman, and Mr. Frederic de Hoffman for cooperation in the course of the experiment and in its evaluation.

This paper is based on work performed under Contract No. W-7405-eng-36 with the Manhattan Project at the Los Alamos Scientific Laboratory of the University of California.

## Search for Positron-Electron Branching in Certain Beta Emitting Isotopes

W. C. BARBER

*Department of Physics, University of California, Berkeley, California*

(Received August 13, 1947)

The  $\beta$ -rays emitted by the isotopes Ga<sup>70</sup>, As<sup>76</sup>, Br<sup>80</sup>, Br<sup>82</sup>, Rb<sup>86</sup>, Rh<sup>104</sup>, Ag<sup>108</sup>, In<sup>116</sup>, Sb<sup>122</sup>, Sb<sup>124</sup>, I<sup>128</sup>, Re<sup>186</sup>, and Au<sup>198</sup> have been examined for positrons by the "trochoidal" method of Thibaud. Br<sup>80</sup> is the only one of the above isotopes which was found to emit positrons as well as electrons. In studying Rb, a new activity of about 40-day half-life was found.

### 1. INTRODUCTION

WHEN a radioactive isotope of charge  $Z$  is isobaric with stable isotopes of charge  $Z-1$  and  $Z+1$  there exists the possibility of decay by positron emission or orbital electron capture as well as by electron emission. There are many isotopes in this situation, yet only a few of them are known to emit both positrons and electrons. The present experiments were undertaken with the idea that a sensitive detection method might show this branching in cases where it had hitherto been undetected. The trochoid provides an efficient means of making a complete separation of positrons and electrons, hence it was selected for the experiments.

### 2. EXPERIMENTAL APPARATUS

The trochoid was first used by Thibaud<sup>1</sup> to verify the existence of positrons and to study some of their properties. The instrument as used in the present experiments is simply a brass vacuum chamber shaped like a 270-degree section of a doughnut. A source is placed at one end of the chamber, and a Geiger-Müller counter is mounted at the other. The chamber fits between the poles of a circular-pole electromagnet in such a way that electrons from the source describe trochoidal paths around to the detector. When the magnet

current is reversed only positrons can reach the detector. A diagram of the apparatus with a possible electron trajectory is shown in Fig. 1.

To make the source readily changeable a vacuum tight "well" with a 0.001-in. copper window was mounted in the source position. Absorption of the  $\beta$ -rays was studied by putting aluminum absorbers in the well between the source and the window. In experiments where it was desirable to have a minimum of absorbing material between source and detector the well was not used, and the source was mounted directly inside the vacuum chamber.

The counter had a 0.001 inch aluminum window which was waxed onto a cylindrical brass shell. The brass shell had three slots on one side through which the  $\beta$ -particles entered. Plateaus of about 100 volts were obtained when the counter was filled with 3 cm of argon and 2 cm of alcohol.

The apparatus was tested by measuring the relative number of positrons and electrons emitted by Cu<sup>64</sup>. It was found, as is well known,<sup>2</sup> that the observed ratio of electrons to positrons depends on the source thickness and on the counter-window thickness. With a source 0.0002 in. thick and a 0.001-in. aluminum counter

<sup>1</sup> J. Thibaud, Phys. Rev. **45**, 781 (1934).

<sup>2</sup> K. Sinma and F. Yamasaki, Scientific Papers of the Institute of Physical and Chemical Research, Tokyo, **35**, 16 (1938).