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The Relative Probabilities and Absolute Cross Sections of the D – D Reactions

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Measurements with a gas target have been made of the relative probabilities and absolute cross sections of the two possible D – D reactions, by simultaneously counting the helium and hydrogen nuclei produced by the reactions. The counters were separated from the target by a thin silica window. The ratio of the cross section of the neutron-producing reaction to that of the proton-producing reaction was found to increase from 0.95 at 120 kev to 1.06 at 250 kev. The "neutron" cross section was 0.019 barn at 120 kev and 0.038 barn at 250 kev while the "proton" cross section increased from 0.020 barn at 120 kev to 0.049 barn at 300 kev. The results can be fitted to an expression of the form $\sigma E = \sum a_l P_l$ and the values of the coefficients a_l are given.

I. INTRODUCTION

When deuterium is bombarded with deuterons of any energy two nuclear reactions are possible:

$$D^2 + D^2 \rightarrow H^3 + H^1$$
$$\rightarrow He^3 + n.$$

Many investigations have been carried out on the first of these reactions but the other has been relatively neglected. This is mainly due to the circumstance that either the He particles or the neutrons must be counted, and the first is troublesome owing to the short range of the He nuclei, the second owing to scattering and to the necessity of calibrating the neutron counter. Theoretically, it has usually been considered that the two reactions would have the same probabilities.

The experimental results, however, indicate that the ratio of the cross section of the second reaction (the neutron-producing reaction), to the cross section of the first (the proton-producing one), hereafter called the branching ratio, does not in general equal unity, nor does it remain constant with energy. The experiments of Blair and his co-workers[1] in the range 1 to 3.5 Mev indicate a tendency for the branching ratio to fall with energy while Manley, Coon, and Graves and Graves, Graves, Coon and Manley[2,3] found that the branching ratio increased with energy in the energy range 100 to 300 kev. Pepper[4] concludes that the branching ratio is constant below 60 kev. Previous work in this laboratory[5] appeared to shown that the branching ratio increased with energy in the region 70 to 160 kev. Of these authors, only Blair and his associates used a gas target.

Again, the values for the absolute cross section of the proton reaction given by Sanders, Moffat, and Roaf[6] and by Bretscher, French, and Seidl[7] do not agree.

In view of these inconsistencies, it appeared important to redetermine the variation of the branching ratio with voltage using a gas target. This approximates to an ideally thin target in which there can be no dispute as to the energy of the incident particles when a reaction takes place. Moreover, the use of a gas target enables absolute cross sections to be determined without the necessity of knowing, for example, the rate of loss of energy of deuterons in heavy ice.

II. APPARATUS

The high voltage was provided by a Philip's type generator, capable of giving voltages up to 500 kev. An rf ion source was employed.[8] The beam was analyzed magnetically, to ensure that only D^+ ions of definite energy caused reactions. At first a 90° analyzer was used, but later this was replaced by a 10° analyzer, which had a smaller defocusing action on the beam. The beam passed into the target through the two canals of a differential pumping system. A diagram of the canal system and the target is given in Fig. 1.

In lining up the beam the following procedure was adopted. First a maximum current was obtained through the analyzer. The first canal section was then bolted to the analyzer, and an observation chamber placed at its far end. The beam was centered on the entrance port of the canal, as seen through a window. The canal was pivoted about an axis through the entance port with ball and socket joint, and could be rotated in any direction (±15°), by means of the threaded rods and flexible section contained in its construction. Its orientation was adjusted until a

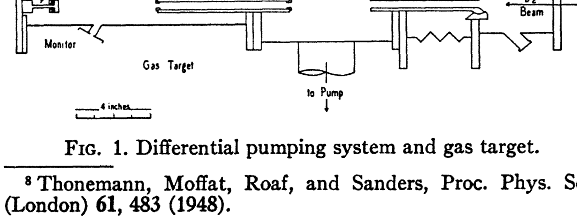

Fig. 1. Differential pumping system and gas target.

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[1] Blair, Freier, Lampi, Sleater, and Williams, Phys. Rev. **74**, 553 (1948).
[2] Manley, Coon, and Graves, Phys. Rev. **70**, 101A (1946).
[3] Graves, Graves, and Manley, Phys. Rev. **70**, 101A (1946).
[4] T. P. Pepper, Can. J. Research **27**, 143 (1949).
[5] McNeill, Thonemann, and Price, Nature **166**, 28 (1950).
[6] Sanders, Moffat, and Roaf, Phys. Rev. **77**, 754A (1950).
[7] Bretscher, French, and Seidl, Phys. Rev. **73**, 815 (1948).
[8] Thonemann, Moffat, Roaf, and Sanders, Proc. Phys. Soc. (London) **61**, 483 (1948).

maximum current passed through the canal, and the rods were then locked. The target, to which the second canal was rigidly attached, was then screwed to the T-piece joining the diffusion pump to the target system, and the T-piece attached to the pump through a bellows section. A lattice-work sleeve fitted over the second canal, to ensure that when the target section was screwed to the first canal section the two canals would be parallel, and that therefore a maximum beam would pass through the second canal into the target. This sleeve is not shown in the figure.

The diffusion pump used was an Edwards 403, which has a nominal baffled speed for air of 200 liters/sec. It was found possible to maintain a pressure of about 0.1 mm Hg in the target without the pressure in the accelerator tube rising appreciably. About 200 cc of deuterium at NTP were used per hour, and this was ejected to the air and wasted. At this target pressure the loss of energy of the incident deuteron beam is negligible. The deuterium flow into the target was controlled by a needle valve, and a McLeod gauge was used to measure the target pressure. A reading of the gauge was taken every few minutes during a run, and the average pressure at any particular time was obtained from a pressure-time graph. After each run the gas from the target was sampled. K. I. Mayne very kindly analyzed the samples in a mass spectrometer to determine the relative amounts of deuterium and other gases present.

The beam intensity was measured by means of a calorimeter,[9] similar to that used by Moffat, Roaf, and Sanders[10] and had a sensitivity of about 0.3 mv/watt.

The beam current measurement is dependent directly on the beam voltage measurement, as the calorimeter measures watts, rather than amperes. The voltage was measured by the current flow through a high resistance of approximately 200 megohms. The value of the resistor was determined by a condenser discharge method with an accuracy of 1 percent. During the long runs necessary with the gas target, it was found that the high voltage tended to fluctuate, owing to the variations in the current drawn from the set. The resolving power of the magnet plus the canal system was such that a variation of 1.5 percent in the beam voltage would stop the beam entering the target, and if the voltage altered by more than 0.5 percent it was noticed by a fall in the current and returned to its original value. The ripple on the set did not exceed 0.5 percent of the steady potential.

The gas target is shown in Fig. 1. The main body was made from a section of rectangular wave-guide tubing of cross section 4 in.×2 in. and length 12 in. Both ends had flanges soldered to them, in one of which was pushed the target canal, and through a hole in the other was inserted the beam measuring calorimeter. The beam passed close to the top of the wave guide, so that the solid angle subtended by the beam to the counters was as large as possible. The deuterium gas entered the target through a tube in the bottom of the wave guide, and through a similar tube a lead was taken to the McLeod gauge. On the top surface of the guide was hard-soldered a flange which served as a bed for the counter turn-table. Finally, provision was made for the attachment of a subsidiary counter to the bottom of the guide to act as a monitor for the beam. All vacuum seals throughout the target and the rest of the system were made with rubber gaskets, or, where this was impracticable, with wax.

[9] J. Sanders, J. Sci. Instr. **26**, 36 (1949).
[10] Moffat, Roaf, and Sanders (to be published).

III. EXPERIMENTAL PROCEDURE

The three charged particles produced by the D−D reaction have widely different rates of loss of energy, and it is thus possible to differentiate among them using a single proportional counter. As the proton pulses, in the counters used, were very small compared with those produced by the He^3 nuclei (about $\frac{1}{10}$ as large), it was found better to count the protons in a second chamber, separated from the first by an Al foil. The counting of the protons was done simultaneously with the counting of the heavier particles in the first chamber.

The twin counters used were screwed on to a threaded tube mounted on the counter turn-table, and thus could be rotated about the beam. For simplicity the two counters were made as a single unit, and filled to the same pressure. They were both of the side-on type, the counter wires, of 100μ tungsten, being terminated in small glass beads. The first counter, in which He^3 and H^3 nuclei were counted, was of width 1 cm, whereas the path length of the protons in the second counter was 5 cm, to compensate for their smaller rate of loss of energy. As described in a previous letter,[5] a thin Si window was used between the target and the first counter, this window allowing all of the charged reaction particles to pass through it. The partition between the twin counters, which was removable, had an insert for receiving an Al foil, which was held taut and in place by a spring washer. This foil was of sufficient thickness to stop the He^3 and H^3 nuclei entering the second chamber. A number of holes around the outside edge of the partition allowed free passage for the counter gas. The counters could be filled to any desired pressure with a 10:1 argon-alcohol mixture. It was found that 12 cm of this mixture gave optimum conditions for resolution between the different particles. A copper plate, sufficiently thick to stop all of the charged particles, could be moved across the entrance to the collimating system by means of a rod passing through a Wilson seal, to enable backgrounds to be measured. In previous experiments, when an end-on counter was used to count the protons, difficulties had been experienced, owing to the fact that the number of tritons counted in the first chamber was not equal to the number of protons in the second counter. This was probably due to the end-on counter not being equally efficient at different distances from the wire, but to check that this was not the case with the present arrangement, the counters and collimating system were tested with alpha particles before being used on the D−D reaction. For this experiment the Al foil was removed from the partition.

Conventional electronic circuits were employed, with a head and main amplifier, cathode-ray oscillograph monitor, and pulse-height discriminator and scaler, or pulse-height selector and scaler, connected to each counter. With the counting rates used, there was negligible error caused by the finite resolving time of the counters or the electronic equipment.

The optimum counter conditions were first found using an Al target; i.e., a target in which deuterium gas from the beam is occluded. This conserved deuterium, and also enabled the best conditions of counter pressure, wire potential, etc., to be found rapidly, owing to the large yield obtained from this type of target.

Owing to the time lag in the current measurement caused by the use of a calorimeter, a GEC G-M4 counter was used to monitor the beam. This was attached to the underside of the wave guide. If the count registered by the monitor during a run was very high or very low, the run was rejected, as it indicated that a large fluctuation in the beam current had taken place. The calorimeter had a half-period of 1 minute, and during the experiment the millivoltmeter attached to the thermocouples was read every minute. Alternate "direct" and background runs were taken, to avoid any errors due to fluctuations in the background. The background in the counters was almost entirely caused by neutrons which were due to the D−D reaction taking place at solid parts of the target; e.g., the calorimeter and canals, where deuterium was occluded.

TABLE I. Values of the branching ratio of the D−D reactions.

Voltage	Branching ratio σ_n/σ_p	Statistical error
120 kev	0.95	0.04
140	1.00	0.07
160	1.00	0.04
250	1.06	0.04

Counting rates of about one per second were obtained during the direct runs, and normally about 4000 counts were taken. The background was of the same order as the hydrogen count, and about a third of the actual count in the counting of the He^3 particles (the difference is caused by the fact that the He^3 pulses were much bigger than the H pulses). This causes the statistical errors to be much larger than the figure 4000 would lead one to expect. It was not found possible to reduce the background appreciably by the introduction of paraffin wax, owing to the small distance between the counters and the target.

To ensure that all particles were being counted, both pulse height discriminator bias curves and pulse height selector (kick-sorter) bias curves were taken. The equality of the triton and proton counts was checked. To guard against the possibility that the counter system was not symmetrical about the beam, the counters were turned through 180° and the equality, within the statistical error, of the proton counts in the two positions checked.

IV. RESULTS

For comparison, all data were transformed from the observation angle of 90° in the laboratory system to an angle of 90° in the center of gravity system, assuming an angular asymmetry of the form

$$\sigma_\phi=\sigma_{90}(1+A\cos^2\phi). \tag{1}$$

To make the comparison, all counts were multiplied by the expression

$$g(\phi)/(1+A\cos^2\phi).$$

The factor $g(\phi)$ is due to the change in solid angle in turning from one system to another, and is equal to $[\sin^2\theta/\sin^2\phi]\cos(\phi-\theta)$, where θ is the laboratory angle, 90° in this case, and ϕ is the center of mass angle.[11] The values of A were taken from the work of Manning, Huntoon, Myers, and Young.[12]

All readings were normalized to unit current, time, and pressure in the target. In dividing any particular reading by the current, account had to be taken of the fact that the calibration of the calorimeter altered with different target pressures. As the background was virtually independent of the gas pressure, the actual normalizing formula was

$$\left\{\frac{\text{count}}{\text{current}\times\text{time}}-\frac{\text{background}}{\text{current}\times\text{time}}\right\}\Big/\text{pressure}.$$

By a comparison of the He and H counts for the same number of reactions, i.e., with unit beam current, unitary target pressure and in unit time, the ratio of the cross sections, the branching ratio, is directly obtained. The results are given in Table I. The errors indicated are statistical. In addition, there are measurement errors which are considered to have a maximum value of 2.7 percent. These enter from the reading of the pressure ($\frac{1}{2}$ percent), and the thermocouple emf ($\frac{1}{2}$ percent) (both readings are averaged over a run), and from the fact that inaccuracies in the reading of, or variations in, the beam voltage will introduce an error directly into the determination of the current from the calorimeter readings (1.5 percent) and also indirectly into the branching ratio values as the branching ratio is a function of the high voltage (0.2 percent). In the graph of the results, Fig. 2, the statistical errors are indicated. For comparison, the values of the branching ratio found previously using a solid target are given.

[11] Haxby, Allen, and Williams, Phys. Rev. **55**, 140 (1939).
[12] Manning, Huntoon, Myers, and Young, Phys. Rev. **61**, 371 (1941).

From the readings obtained in the branching ratio experiments it is possible to obtain the absolute cross sections of the two reactions. This entails the calibration of the calorimeter and the McLeod gauge and the calculation of the solid angle subtended by the collimating system. The calibration of the calorimeter was carried out using a Metrosil resistor as a heating unit, the wattage dissipated in the heater being measured by a potentiometer method. As with gas in the target gaseous conduction of heat competes with the metallic conduction down the thin walled tube, the calibration depends on the gas target pressure, and this variation was also determined. The McLeod gauge was calibrated by the normal technique of expanding small volumes of gas at pressures of 10 cm or so, which can be measured accurately by an Hg manometer and vernier microscope, into large volumes.

The collimating system consisted of two circular apertures, A and B, of diameters $2\frac{1}{2}$ and 4 mm separated by a distance of 2 cm. The direct calculation of the solid angle with such a system is difficult, and therefore an indirect method was employed. It was assumed for a first approximation that the hole nearer the beam was a slit. With such a system the calculation is straightforward.[13] The expression for the solid angle thus obtained was multiplied by a factor G, to correct for the use of a circular aperture. G was determined graphically, with an estimated error of less than 2 percent. Apart from all possible errors introduced in the measurement of the solid angle, there is a possibility that there may be a central core to the beam, which is not necessarily in the geometric center of the target area. Although optical observation did not indicate the presence of such a core, the possibility may not be excluded when the maximum errors are being considered. With the canal diameters and the distance of the beam from the counters used, this would introduce a maximum error of 5 percent, on top of the purely measurement errors (including that in G), of another 5 percent.

The readings taken will give the differential cross sections at 90° to the beam. The total cross section can be obtained by multiplying the differential cross section by $4\pi(1+A/3)$, assuming an angular asymmetry of the form of Eq. (1). As before, the values of A at different voltages are taken from the work of Manning *et al.* In Table II the differential and total cross sections of the two possible reactions are given between 120 and 300 kev, with the assumed value of A recorded. The errors indicated are statistical. In addition, errors in calibration and measurement, amounting to a maximum possible value of 20 percent, must be considered. This figure is made up of a 10 percent geometric error, $3\frac{1}{2}$ percent from the calibration and reading of the McLeod, $4\frac{1}{2}$ percent from the current measurement (2 percent from the wattage calibration, 2 percent from beam voltage uncertainty and $\frac{1}{2}$ percent reading error), $\frac{1}{2}$ percent from the estimation of the deuterium content of the target gas, and $1\frac{1}{2}$ percent from the high voltage. This comes from the variation of cross section with voltage. In the graphs of the results, Figs. 3 and 4, 20 percent errors are indicated.

[13] Herb, Kerst, Parkinson, and Plain, Phys. Rev. **55**, 998 (1939).

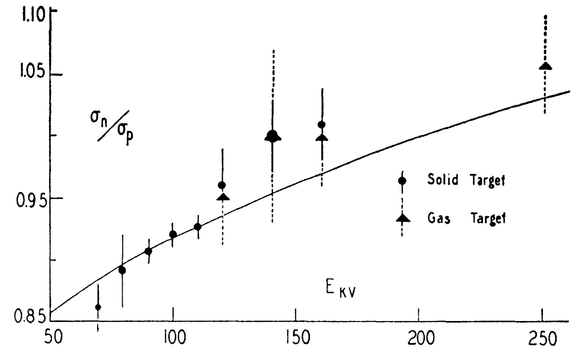

FIG. 2. The ratio of the cross sections of the $D(d,n)He^3$ and the $D(d,p)H^3$ reactions (branching ratio) as a function of voltage. The solid target results are taken from reference 5.

V. DISCUSSION

Following Konopinski and Teller,[14] an attempt was made to fit the results to an expression of the form $\sigma E = \sum_l a_l P_l$ where σ is the total cross section at energy E, P_l is the interpenetrability of two deuteron waves with relative angular momentum l, and a is a constant, equal to $\pi(2l+1)\cdot|\alpha_l|^2\cdot\hbar^2/2m$ where $|\alpha_l|$ is the intrinsic probability of a particular reaction taking place. In the voltage range considered only three terms of the expansion need be considered, owing to the rapid decrease of P_l with E. The values of P_l are given in a paper by Hunter and Richards,[15] and have been independently computed by Spiers.[16] In fitting the proton cross-section curve, consideration has been given to the values obtained by Blair, *et al.*, in the Mev range,[1] and by Sanders, Moffat, and Roaf in the 0- to 50-kev region,[6] as well as to the present results. In fitting the $D(d,n)He^3$ cross-section curve, consideration was

TABLE II. Differential and total cross sections of the D−D reactions at various voltages.

Voltage (kev)		Diff. σ (barns)	Total σ (barns)	(A)	Stat. error (percent)
120	σ_p	0.0013	0.020	(0.55)	3
	σ_n	0.0012	0.019		3
140	σ_p	0.0017	0.026	(0.63)	5
	σ_n	0.0017	0.026		5
160	σ_p	0.0016	0.026	(0.70)	3
	σ_n	0.0016	0.026		3
250	σ_p	0.0021	0.036	(1.00)	3
	σ_n	0.0022	0.038		3
300	σ_p	0.0023	0.049	(1.18)	6

[14] E. J. Konopinski and E. Teller, Phys. Rev. **73**, 822 (1948).
[15] G. T. Hunter and H. T. Richards, Phys. Rev. **76**, 1445 (1949).
[16] J. A. Spiers, private communication.

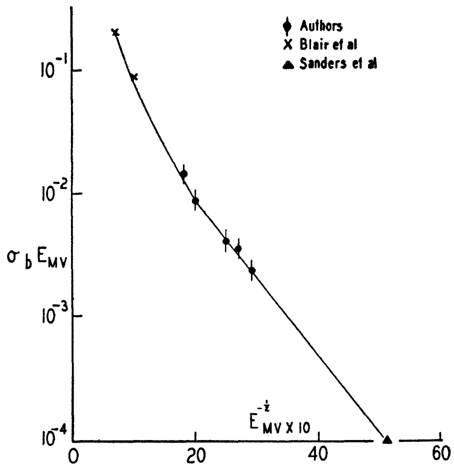

FIG. 3. A Gamow plot, $\log\sigma \cdot E$ *vs* $E^{-\frac{1}{2}}$, of the cross section of the reaction $D(d, p)H^3$.

taken of the results of Blair, *et al.*, Hunter and Richards, and of the present work. All these authors used gas targets. The best fit was obtained with the coefficients listed below. a_0, a_1, a_2, have been used to denote the coefficients a_l with l equal to 0, 1, and 2 in the $D(d, p)H^3$ reaction, and b_0, b_1, b_2, the corresponding coefficients in the neutron reaction.

$$a_0 = 0.006 \qquad b_0 = 0.0045$$
$$a_1 = 0.12 \qquad b_1 = 0.15$$
$$a_2 = 0.26 \qquad b_2 = 0.26.$$

The theoretical curve based on these coefficients is included in the graphs of the results.

The branching ratio (BR) will be given at any voltage by the expression

$$BR = (b_0P_0 + b_1P_1 + b_2P_2)/(a_0P_0 + a_1P_1 + a_2P_2)$$

where the P_l correspond to that voltage. Using the above values of the coefficients, the values of the branching ratio at various voltages have been calculated, and the theoretical line based on these figures is

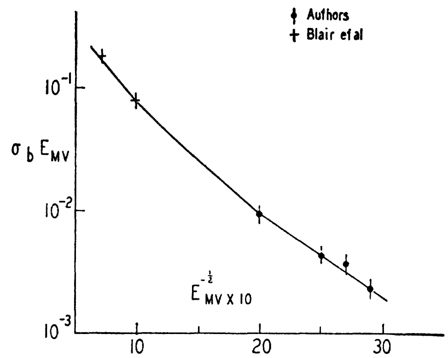

FIG. 4. A Gamow plot, $\log\sigma \cdot E$ *vs* $E^{-\frac{1}{2}}$, of the cross section of the reaction $D(d, n)He^3$.

included in Fig. 3. It is of particular interest to note that there is very close agreement between the values of the branching ratio obtained using an occluded target and those using the gas target. This may be due to a thin layer of deuterium absorbed on the surface of the copper block acting as the main target.

It will be seen from the graphs, that there is good agreement of the present work with the results obtained by other workers using a gas target. There is, however, considerable disagreement with the results of Bretscher, French, and Seidl, who used a heavy ice target. An incorrect value for the rate of loss of energy of deuterons in heavy ice could account for the discrepancy.

We wish to express our thanks to Professor Lord Cherwell for extending to us the facilities of the laboratory, to Mr. C. H. Collie for his continued interest and advice, and to all the members of the HT group for their generous support. Mr. J. A. Spiers is to be thanked for many valuable discussions. One of us (K. G. McN.), is indebted to the Harmsworth Trust of Merton College, Oxford, and to the Department of Scientific and Industrial Research for financial support at different times, and the other of us (G. M. K.), wishes to thank the Physics Department of the National Research Council of Canada for similar assistance.