With the quantities of material available at the time of these experiments, attempts to isolate the Tl^{209} activity were unsuccessful. Since then one of us[30] has been able to characterize the isotope as decaying with a 2.2-min. half-life by emission of a 1.8-Mev beta-particle. Gamma-rays if present constitute a maximum of one percent of the Geiger activity.

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Interaction of 12- to 13-Mev Neutrons with Deuterons*†

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By use of a CD_4-filled high pressure cloud chamber, the angular distribution of recoils from 12- to 13-Mev neutrons has been studied. The angular range 0° to 140° in the center-of-mass system was covered by these measurements. The distribution indicates a rather large asymmetry in the forward and backward directions with a minimum at about 60° in the center-of-mass system. The existence of a $D(n, 2n)p$ reaction was evident by the presence of tracks between 60° and 90° in the laboratory system which were too long to be accounted for by either deuteron or proton elastic recoils.

I. INTRODUCTION

Theoretical predictions which have been developed up to the present time concerning the angular distribution of neutrons elastically scattered by deuterons show a marked dependence on the type of nuclear forces assumed. Massey and Buckingham[1] have made extensive calculations concerning this problem below 11.5 Mev, and Verde[2] has recently published calculations concerning this problem for neutron energies up to 20 Mev. Since 1937 there have been several experimental determinations made of the angular distribution of $n-d$ elastic scattering.[3] Unfortunately, the discrepancies existing among these various experimental determinations of the angular distribution and the theoretical predictions are such that no definite conclusions can be drawn concerning the nature of the nuclear forces involved in $n-d$ elastic scattering. Rosenfeld[4] has presented an excellent summary of the experimental and theoretical work done on this problem up to 1948. The importance which this problem may have in the eventual understanding of the nature of nuclear forces makes it reasonable to accumulate more experimental evidence concerning $n-d$ elastic scattering.

II. APPARATUS

The high pressure cloud chamber used by Laughlin and Kruger[5] to measure $n-p$ scattering has been used to investigate $n-d$ scattering. A thin deuterium gas target bombarded by 10-Mev deuterons from the University of Illinois cyclotron served as a source of 12- to 13-Mev neutrons. The cloud chamber was filled with CD_4 and used D_2O and isopropyl alcohol as the vapor mixture. This served as a source of deuteron scattering centers as well as a detector of the deuteron recoils. Details of the cloud chamber, neutron source and collimator, and the resultant neutron spectrum may be found in reference 5.

The cloud chamber was surrounded by a temperature-controlled water-cooling system which was maintained at 21°C.

An Illex No. 3 Universal shutter was mounted directly in front of the camera lens to determine the exposure time. In case of shutter failure, caused by the excessive strains of solenoid operation, the shutter could be replaced without disturbing the camera lens position. The use of two General Electric A-H6 mercury arc lamps for chamber illumination, a coated $f/2.0$ Ektar 45 mm lens, and Linagraph Ortho 35 mm film gave satisfactory exposures at $f/3.5$ and 1/20 of a sec.

Neutrons were introduced into the cloud chamber only during expansion by placing a deuteron beam shutter made of sheet tungsten ahead of the thin deuterium gas target. At each expansion this beam

* This report is part of a thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the University of Illinois, 1950. A preliminary report of these data was given in Phys. Rev. **77**, 748 (1950).

† Assisted by a joint program of the ONR and the AEC.

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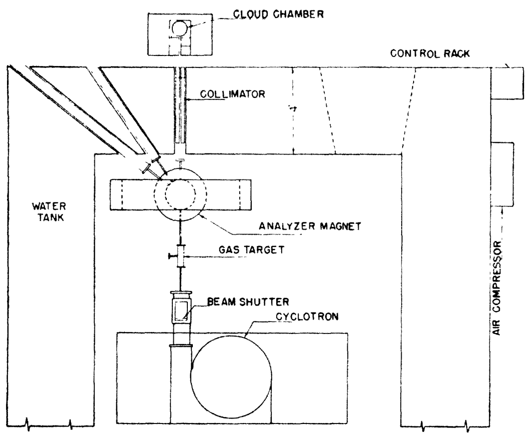

FIG. 1. Arrangement of experimental apparatus.

shutter was lowered out of the deuteron beam by means of an air-ram controlled from the cloud chamber cam system. Tungsten was used as a beam shutter since it has a low yield of gamma-rays and neutrons when bombarded by deuterons. The arrangement of the experimental apparatus is shown in Fig. 1.

The exposed film, after being processed, was projected through the same camera and mirror system as was used to photograph the interior of the cloud chamber. The stereoscopic projections were viewed on the screen of the measuring engine which is shown in Fig. 2. Tests were made on this projection and measuring system to study the effects of film shrinkage and the failure of the camera gate mechanism to relocate the film exactly. These tests showed that the reproducibility in a measurement of the recoil angle was $\pm 1°$ for the recoil range 0° to 70°, and the azimuthal angle measurements were reproducible to $\pm 2°$ for the azimuthal range of 0° to 360°.

The heavy methane gas used in the cloud chamber was synthesized by Tracerlab, Inc., Boston, Massachusetts. A sample of this gas was analyzed in a mass spectrograph by Dr. N. D. Coggeshall of the Gulf Research and Development Company. The following analysis was obtained:

CD_4 85.4 percent	N_2 4.6 percent
CD_3H 9.2 percent	O_2 0.8 percent.

III. EXPERIMENTAL PROCEDURE

These data were taken in two separate runs. During the high pressure run the cloud chamber was operated at a compressed pressure of 15.84 atmos., an expanded pressure of 13.84 atmos., and a 162 sec. cycle time. A compressed pressure of 6.33 atmos., an expanded pressure of 5.60 atmos., and a cycle time of 90 sec. were used during the low pressure run. The laboratory recoil angular range investigated was 0° to 60° in the high pressure run and 31° to 70° in the low pressure run. The chamber pressures used were such that the entire range of a deuteron recoiling from a 12- to 13-Mev neutron was contained within the chamber. This was necessary since the neutron spectrum[5] incident on the chamber showed a d, d peak of neutrons at 12- to 13-Mev superimposed on a continuous background. For this reason it was necessary to use a track length selection criterion in order to choose only those tracks which could have been deuterons elastically recoiling from 12- to 13-Mev neutrons.

Since the cycle time of the chamber was long, it was desirable to make the yield of recoils as large as possible by allowing neutrons to enter the chamber slightly before and throughout the expansion process. Thus the pressure differential in the chamber and the one Mev energy spread of the incident d, d neutrons made it necessary to accept a spread of recoil track lengths for each recoil angle. The maximum and minimum acceptable recoil ranges as a function of recoil angle are plotted for the two runs in Fig. 3. These were calculated from the usual range-energy curves and the energy-momentum considerations for the $n-d$ elastic scattering process. The stopping power of the chamber gas as a function of recoil range in air was used to convert the recoil range in air to range in the chamber.

The range limits shown in Fig. 3 assured that all recoils from 12- to 13-Mev neutrons would be accepted. However, recoils from 13- to 14-Mev neutrons formed while the chamber was compressed as well as any recoils from 11- to 12-Mev neutrons formed while the chamber was expanded also would have ranges falling within these limits. From the known time, relative to expansion, that neutrons were allowed to enter the chamber

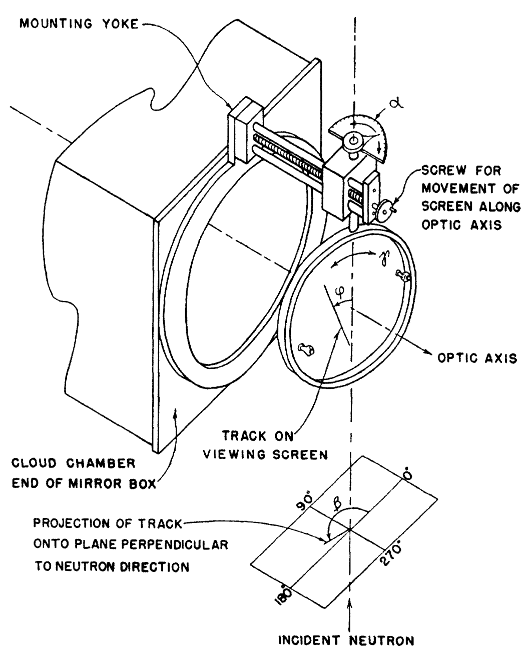

FIG. 2. Schematic diagram of the measuring engine. The angles φ and β are the laboratory recoil angle and the azimuthal angle, respectively.

and from the shape of the neutron spectrum it was calculated that 88 percent of the recoils were due to 12- to 13-Mev neutrons while the remaining 12 percent were due to 11- to 12-Mev and 13- to 14-Mev neutrons.

A total of 4009 photographs was taken during the high pressure run, and 3042 photographs were taken during the low pressure run. All of the photographs from the low pressure run and half of the photographs from the high pressure run were examined twice, and independent measurements on acceptable tracks made. Satisfactory agreement was obtained in the two independent measurements. For this reason the remaining photographs were examined only once. 3271 tracks from the high pressure run and 958 tracks from the low pressure runs were found to have acceptable range-angle correlation as defined by the curves in Fig. 3. These acceptable tracks were grouped into 10° recoil angular intervals.

The use of two correction factors was necessary in conversion of the observed tracks to an angular distribution. A length correction factor was necessary because: (1) the portion of the cloud chamber in which a recoil could originate and not strike the opposite chamber wall is a function of the recoil range, recoil angle, and the azimuthal angle; (2) it was difficult to detect a short range recoil near the beam entrance side of the chamber because of the presence of electron tracks produced by gamma-rays from the collimator. An azimuthal correction was necessary since a different fraction of the azimuthal interval was used in the observation of recoils in each 10° recoil interval. The fraction of the azimuthal interval used was determined by the following factors: (1) the light beam illuminating the chamber was $1\frac{1}{2}$ inches high, thus placing a geometrical limit on the azimuthal interval in which all recoils of a given recoil angle were fully illuminated; (2) it was determined experimentally that the observer failed to detect some of the very short range recoils if the azimuthal angle was near 90° or 270°.

These two corrections were determined experimentally. For each 10° recoil interval a track origin distribution was plotted showing the experimentally observed number of tracks originating per centimeter across the chamber along the path of the neutron flux. The usable length of the chamber for each 10° recoil interval was taken as that portion of the chamber in which the number of recoils originating per centimeter was constant within statistical limits. The experimental length correction factor was the reciprocal of the usable length of the chamber and for convenience was normalized so that the smallest value was unity.

The azimuthal correction factor was determined by making azimuthal distribution plots for each 10° recoil interval. Since the four azimuthal quadrants were symmetrical with respect to the light beam and to the point of observation, all of the tracks were folded into the first azimuthal quadrant. From these plots the fraction of the quadrant in which all tracks were fully illuminated and detected by the observer was determined, and the experimental azimuthal correction factor was the reciprocal of this fraction. In plotting the azimuthal distributions only those tracks originating in the usable length of the chamber for each 10° recoil interval were used.

These two correction factors allowed these data to be converted to an angular distribution. The corrected number of tracks per unit solid angle for each 10° recoil interval was determined by the product of the observed number of tracks in the azimuthal interval where all tracks were observed, the azimuthal correction factor, the length correction factor, and the reciprocal of the solid angle included in each recoil interval.

IV. EXPERIMENTAL RESULTS

Figure 4 shows some examples of the tracks photographed in the chamber. The length correction factors and the azimuthal correction factors were experimentally determined as outlined above and are given in Tables I and II.

It was determined experimentally that 93 percent of all recoils with tracks longer than 4 mm originated in the collimated region of the chamber. Presumably the remaining 7 percent of the tracks were due to uncollimated neutrons which were scattered into the chamber and produced recoils. It was observed that 7.2 percent of those recoils originating outside of the collimated region had a length such as to be an acceptable track

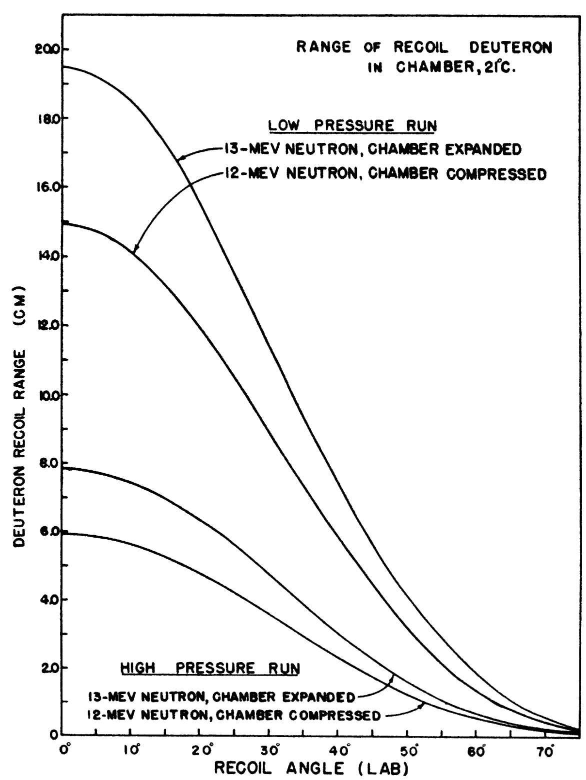

FIG. 3. Range of deuteron recoils in the chamber.

if the neutron direction was assumed parallel to the collimator axis. Assuming a uniform distribution of these uncollimated neutrons throughout the entire chamber, it was calculated that 0.5 percent of the acceptable tracks originating in the collimated region could have been due to random, uncollimated neutrons.

The experimental results are summarized in Tables I and II. The results of the low pressure run were fitted to the high pressure run in the three overlapping recoil intervals by the method of least squares. The errors assigned to the relative numbers of tracks per unit solid angle in each recoil interval are the probable errors. The calculation of these errors is considered in the following section. It is to be noted that only those usable tracks which fell in the azimuthal intervals where all tracks were detected were used in the final analysis of these data. Thus while a total of 4229 tracks were measured as having an acceptable range-angle relation, only 1534 of these were used.

Figure 5 shows the angular distribution of the observed tracks. A comparison also is made with some experimental results of Coon and Taschek,[3] the two curves being normalized at the 60° center-of-mass recoil angle. It is to be noted that these data have not been corrected for the presence of confusable recoil or disintegration protons. The recoil protons could arise from the presence of ordinary hydrogen in the scattering material, and a D(n, $2n$)p reaction would give rise to reaction protons. This background of confusable protons will be discussed further in a following section.

The data of Coon and Taschek shown in Fig. 5 also include a background of protons of the same order of magnitude, so that a direct comparison of data seems reasonable. Considering the difference in incident neutron energies the two curves are in fair agreement in the 0° to 140° center-of-mass recoil interval. A comparison of these data to the theoretical predictions[1,2] shows agreement only in the sense of a pronounced asymmetry with a minimum near the 60° center-of-mass recoil angle. No good fit of these experimental data with any of the theoretical curves could be made.

V. EXPERIMENTAL ERRORS

The errors assigned to the experimental points in Fig. 5 are probable errors based on the probable errors in the following factors:

(1) Probable error in the number of usable tracks in each recoil interval which were in that fraction of the azimuthal quadrant where all tracks were detected.
(2) Probable error in the length correction factor.
(3) Probable error introduced by confusable recoils from uncollimated neutrons.

The azimuthal correction factor is a pure numerical ratio and the solid angle included is a trigonometric calculation. Hence these factors introduce no errors.

Other sources of error such as small temperature and

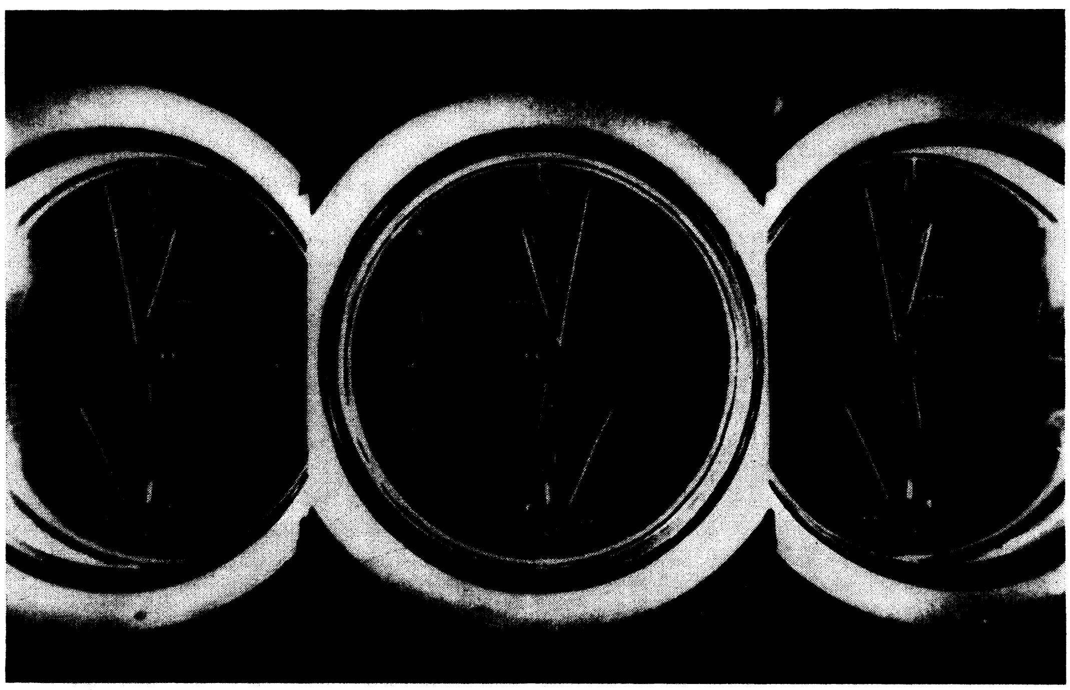

FIG. 4. Photograph taken during the high pressure run with cloud chamber filled to 15.84 atmos. with CD_4. The tracks due to recoils are easily discernible, accompanied by some electron tracks produced by the gamma-rays incident on the chamber wall from the collimator. The neutron collimation is evident by the restricted region across the chamber in which recoils originated. The direction of the incident neutrons is from the bottom to the top of the picture along the diameter of the chamber.

TABLE I. Results of high pressure run.

Lab recoil angle CM recoil angle CM scattering angle	0°–10° 0°–20° 180°–160°	11°–20° 20°–40° 160°–140°	21°–30° 40°–60° 140°–120°	31°–40° 60°–80° 120°–100°	41°–50° 80°–100° 100°–80°	51°–60° 100°–120° 80°–60°
No. of acceptable tracks	399	499	260	382	779	952
No. of acceptable tracks in usable part of chamber	297	405	206	314	596	704
Usable length of chamber	6.0 cm	7.0 cm	8.0 cm	9.0 cm	9.0 cm	9.0 cm
Length correction factor	1.50	1.29	1.12	1.00	1.00	1.00
No. of usable tracks in azimuthal interval where all tracks are detected	209	170	96	145	281	441
Azimuthal correction factor	1.50	3.00	3.00	3.00	3.00	3.00
CM solid angle	0.379	1.09	1.67	2.05	2.18	2.05
Corrected no. of tracks per unit CM solid angle	1240	604	190	213	387	645
Relative no. of tracks per unit CM solid angle	6.53±0.32	3.18±0.17	1.00±0.07	1.12±0.06	2.03±0.08	3.39±0.11

pressure variations in the chamber and small uncertainties in the accepted values of the atomic stopping power of the elements in the chamber gas all lead to small uncertainties in the incident neutron energy. These uncertainties are negligible compared to the spread of one Mev in the accepted neutron energies. The calculated probable errors are set forth in Table I and II.

VI. PROTON BACKGROUND

There were three possible sources of protons in this experiment which could have produced protons having track lengths confusable with those of acceptable deuteron recoils. These processes were $n-p$ elastic scattering, (n, p) reactions, and the $D(n, 2n)p$ reaction.

Because of the presence of the elements N, O, and C in the chamber gas, it was necessary to investigate the possibilities of an (n, p) reaction on these elements. The $C(n, p)B$ reaction has a Q-value of -11.2 Mev and hence protons from this reaction are of too low an energy to be confusable. The atomic ratio of O and N atoms to D atoms in the chamber gas was 0.42 and 2.5 percent, respectively. These small atomic ratios combined with the relatively small cross sections[6] for (n, p) reactions on N and O leads to a negligible number of confusable protons from this process.

The presence of CD_3H and isopropyl alcohol in the chamber made possible confusable protons from $n-p$ elastic scattering. The atomic H/D ratio was determined to be 3 percent from the chamber gas analysis. Energy considerations showed that 8.0 to 8.6 Mev neutrons would produce confusable proton recoils. If it is assumed that the experimental angular distribution is approximately that for $n-d$ elastic scattering, a rough calculation of the number of confusable protons can be made relative to $n-d$ scattering. The relation

[6] *The Science and Engineering of Nuclear Power* (Addison-Wesley Press, Inc., Cambridge, Massachusetts, 1947), Appendix C.

TABLE II. Results of low pressure run.

Lab. recoil angle CM recoil angle CM scattering angle	31°–40° 60°–80° 120°–100°	41°–50° 80°–100° 100°–80°	51°–60° 100°–120° 80°–60°	61°–70° 120°–140° 60°–40°
No. of acceptable tracks	33	150	436	339
No. of acceptable tracks in usable part of chamber	17	92	360	264
Usable length of chamber	3.0 cm	6.0 cm	10.0 cm	10.0 cm
Length correction factor	3.33	1.67	1.00	1.00
No. of usable tracks in azimuthal interval where all tracks are detected	7	26	77	82
Azimuthal correction factor	6.00	6.00	6.00	6.00
CM solid angle	2.05	2.18	2.05	1.67
Corrected no. of tracks per unit CM solid angle	68	120	226	295
Relative no. of tracks per unit CM solid angle fitted to high pressure run	1.06±0.27	1.88±0.25	3.54±0.27	4.62±0.35

used is

$$\frac{Y_{n-p}}{Y_{n-d}}=KN\frac{\int_{\varphi 1}^{\varphi 2} 2\pi d\sigma_{n-p}(\varphi)\sin\varphi d\varphi}{\int_{\varphi 1}^{\varphi 2} 2\pi d\sigma_{n-d}(\varphi)\sin\varphi d\varphi},$$

where K is the ratio of numbers of 8.0- to 8.6-Mev neutrons to the numbers of 12- to 13-Mev neutrons, N is the H/D atomic ratio, the Y's are the relative yields, and where the $n-p$ scattering is assumed to be isotropic in the center of mass system. K was determined from the observed spectrum[5] of neutrons. The above integrals were evaluated numerically after the total area under the integrand curves had been normalized to the following total cross sections,[4]

$$\sigma_{n-p}=0.85 \text{ barn at } 8.3 \text{ Mev},$$
$$\sigma_{n-d}=0.78 \text{ barn at } 12.5 \text{ Mev}.$$

The $n-d$ angular distribution was extended by the extrapolation of Coon and Taschek[3] in the 140°–180° center-of-mass recoil interval. The results of these calculations are given in Table III.

An inelastic $n-d$ scattering process is energetically possible for incident neutron energies above 3.3 Mev. This $D(n, 2n)p$ process presumably will have a continuous spectrum of proton energies and thus represents a possible source of confusable protons in any $n-d$ elastic scattering experiment. If monoenergetic neutrons were used one probably could isolate the peak of $n-d$ recoils from the continuous proton spectrum by a suitable recoil length or energy analysis. No such discrimination was possible in this experiment because of the continuous background spectrum of incident neutron energies.

However in the 60°–90° laboratory recoil interval, the track lengths of the elastic recoils were very short, with an upper limit in length being imposed by $n-p$ scattering. In this angular interval, 44 tracks were observed in the low pressure run which were too long to be attributed to $n-p$ scattering. If it is assumed that $n-d$ elastic scattering accounts for most of the acceptable recoils observed in the low pressure run in the 31°–70° lab recoil interval, a rough value of the upper limit of the $(n, 2n)$ cross section can be calculated. The relation used for this calculation is

$$\frac{Y_{n,2n}}{Y_{n-d}}=\frac{2\pi\int_{\varphi_1}^{\varphi_2} d\sigma_{n,2n}(\varphi)\sin\varphi d\varphi}{2\pi\int_{31^\circ}^{70^\circ} d\sigma_{n-d}(\varphi)\sin\varphi d\varphi}$$

where the Y's are the observed yields corrected for azimuth and usable length of the chamber, and where

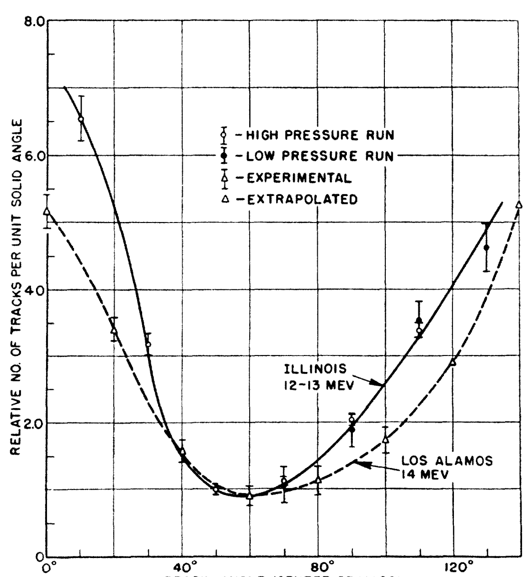

FIG. 5. Experimental angular distribution.

the integrated $n-d$ cross section was taken as 0.37 barn. Thus it was calculated that the integrated cross section for the disintegration process over the lab range 75° to 90° and yielding protons of energies between 1.0 and 3.4 Mev is about 0.014 barn. Similarly, the calculated value of the integrated cross section for the $(n, 2n)$ process over the laboratory range 60° to 90° and yielding protons of energies between 3.0 and 4.8 Mev is about 0.008 barn. These values represent upper limits on the integrated cross sections since, undoubtedly, a larger neutron energy interval, and hence a greater number of neutrons, was responsible for the production of disintegration protons than for the production of acceptable deuteron recoils. It is to be noted that the statistical errors to be attached to these values are of the order of 25 percent. It is impossible to make any calculation of the total cross section for the $D(n, 2n)p$ process from these data.

Chew[7] recently has made some theoretical calculations concerning the $D(n, 2n)p$ process. His ideas were developed for neutron energies above 90 Mev and are likely to be very inaccurate when applied to this experiment. These calculations are based primarily on phase space considerations, with the order of magnitude of the intensity derived from the known $p-p$ and $n-p$ cross sections in the 12.5-Mev range. Chew's calculations, when applied for 12.5-Mev neutrons, predict a total cross section for the $D(n, 2n)p$ reaction of about 0.24 barn with roughly 50 percent of the protons having energies between 0 and 2.0 Mev and 50 percent having energies between 2.0 and 10 Mev, with the high energy protons occurring in the forward directions. His calculations also predict cross sections of 0.013 barn and 0.007 barn corresponding to the experimental cross sections of 0.014 barn and 0.008 barn, respectively, which were mentioned above. It is felt that the agreement between these theoretical and experimental results is fortuitous in view of the very limited experimental data and the questionable application of Chew's calculations to 12.5-Mev neutrons.

A calculation of the ratio of confusable disintegration protons to acceptable deuteron recoils was made for each 10° recoil interval. For this calculation the $D(n, 2n)p$ cross section at 12.5 Mev was determined from Chew's predictions, and the experimental angular distribution curve, normalized to a total cross section of 0.78 barn, was used to determine the $n-d$ cross section for each 10° recoil interval. This method neglects the possibility that a larger number of neutrons may be responsible for disintegration protons than for elastic $n-d$ scattering. The results of these calculations are given in Table IV.

It is not believed by the authors that the results shown in Tables III and IV can be used to make a correction on the observed angular distribution in order

[7] G. F. Chew, University of California (private communications).

TABLE III. Confusable elastically scattered protons.

Lab recoil interval	Ratio of confusable proton recoils to acceptable deuteron recoils: High pressure run (percent)	Low pressure run (percent)
0°–10°	1.0	
11°–20°	1.8	
21°–30°	3.8	
31°–40°	4.5	5.2
41°–50°	2.9	3.4
51°–60°	1.9	2.2
61°–70°		1.3

TABLE IV. Confusable protons from the D(n, $2n$)p reaction.

Lab recoil interval	Ratio of confusable reaction protons to acceptable deuteron recoils (percent)
0°–10°	1.7
11°–20°	4.2
21°–30°	11
31°–40°	15
41°–50°	9.5
51°–60°	7.5
61°–70°	5.0

to obtain the true $n-d$ elastic scattering angular distribution. This is because of the unreliability of the estimated corrections for the $n-p$ and D(n, $2n$)p processes. The main errors in these are due to the questionable method of calculating the D(n, $2n$)p cross section and the fact that the corrections were derived on the assumption that the observed angular distribution was essentially that of $n-d$ elastic scattering. The purpose of including in this report the results shown in Tables III and IV is that they may indicate, approximately, the extent to which confusable protons are present in this experiment.

The angular distribution shown in Fig. 5 cannot be interpreted as the angular distribution for $n-d$ elastic scattering since as much as 20 percent of the tracks observed in some recoil intervals may have been confusable protons. It does seem reasonable to draw the conclusion that the general features of the distribution shown in Fig. 5 are those of $n-d$ elastic scattering. It is highly desirable to have more and better information concerning the D(n, $2n$)p reaction.

The authors are grateful to Dr. N. D. Coggeshall for his analysis of the CD_4 gas. They also wish to express their thanks to the members of the cyclotron staff for their help in carrying out this experiment.

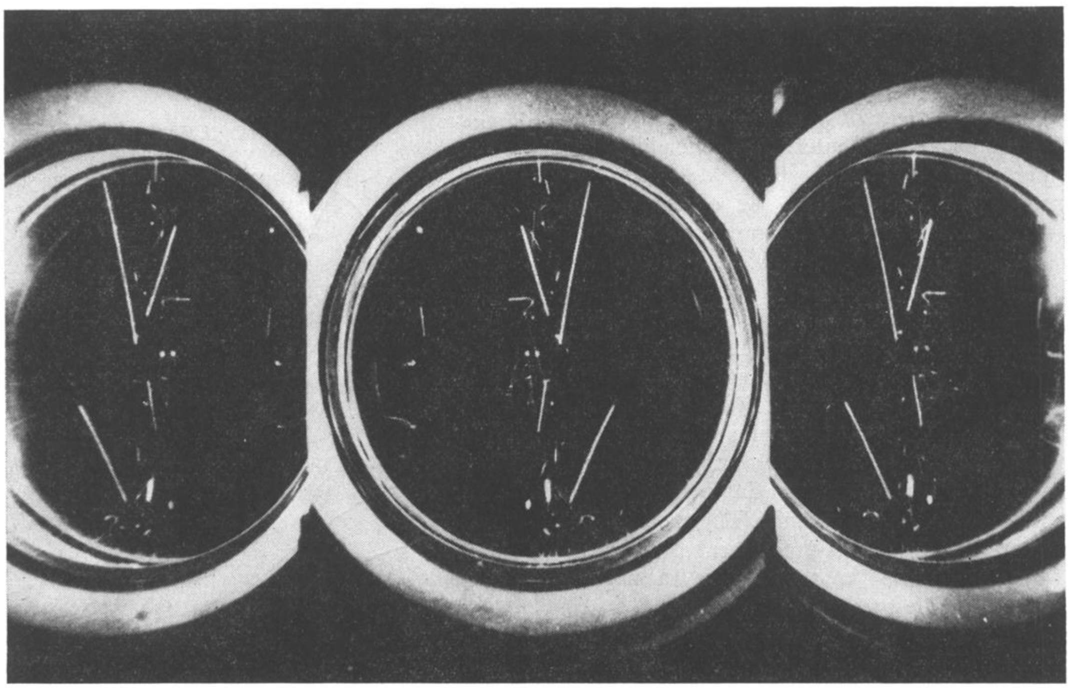

FIG. 4. Photograph taken during the high pressure run with cloud chamber filled to 15.84 atmos. with CD_4. The tracks due to recoils are easily discernible, accompanied by some electron tracks produced by the gamma-rays incident on the chamber wall from the collimator. The neutron collimation is evident by the restricted region across the chamber in which recoils originated. The direction of the incident neutrons is from the bottom to the top of the picture along the diameter of the chamber.