

The Neutrons and Gamma-Rays from the Disintegration of C^{12} by Deuterons

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The excitation curves for the production of gamma-rays and neutrons resulting from the bombardment of carbon by deuterons have been determined over the deuteron energies 0.7 to 1.9 Mev. A careful study of the very narrow gamma-ray resonance occurring at 1.435 Mev has shown that the half-width of the resonance is 5.5 kev and the cross section is 0.62 barn at resonance. Other gamma-ray resonances were observed at 0.91, 1.16, 1.30, and 1.73 Mev with cross sections, respectively, of 0.29, 0.34, 0.39, and 0.97 barn. No resonance for the production of neutrons has been observed at, or near, 1.435 Mev. Resonances for the production of neutrons were observed at energies of 0.91, 1.16, 1.30, 1.62, and 1.76 Mev with the total cross section at resonance, respectively, of 0.12, 0.14, 0.19, 0.19 and 0.22 barn. The angular distributions of the neutrons have been studied; the distributions are quite complex and vary radically with changes in the bombarding energy. At 1.26 Mev there are about five times as many neutrons at 160° as at 0° to the direction of the deuteron beam.

INTRODUCTION

RESONANCE yields occurring in the deuteron bombardment of carbon were first observed in 1940 by Bennett and Bonner. In a series of experiments done at the Rice Institute¹⁻⁴ and at the University of Minnesota^{5,6} the excitation yields for the various disintegration products were determined. The nuclear reactions occurring in C^{12} and the energy released in each reaction are

- (1) ${}_6C^{12} + {}_1H^2 \rightarrow {}_7N^{14} \rightarrow {}_6C^{13} + H^1 + 2.7 \text{ Mev},^7$
- (2) ${}_6C^{12} + {}_1H^2 \rightarrow {}_7N^{14} \rightarrow {}_6C^{13} + H^1 - 0.5 \text{ Mev},^3$
- (3) ${}_6C^{12} + {}_1H^2 \rightarrow {}_7N^{14} \rightarrow {}_7N^{13} + n^1 - 0.281 \text{ Mev}.^8$

The gamma-rays emitted from the excited ${}^*C^{13}$ of reaction (2) have been determined to have an energy of 3.11 Mev,⁹ which is to be compared to a value of 3.2 expected from the measured values of the energy released in reactions (1) and (2). The energy of the gamma-ray involved in the very narrow resonance at 1.435 Mev was checked and found to be about 3 Mev,³ the same as for the other broader resonances.

One of the objects of the present experiment was to obtain the natural width of the sharp resonance at 1.435 Mev. In the previous experiments the observed width of this resonance was 10 kev, but since this was the width expected from the spread in energy of the beam of deuterons, the data showed only that this level was narrower than 10 kev. With

the improved resolution of our apparatus, the natural width could be obtained.

Another object of these experiments was to find if neutrons from reaction (3) showed a resonance at 1.435 Mev. The earlier experiments indicated no resonance, but a careful search for resonant effects is important since it is very difficult to understand theoretically why any level in the excited ${}^7N^{14}$ nucleus, no matter what its parity and angular momentum, cannot break up into N^{13} and a neutron.

A study of the angular distribution of the neutrons of reaction (3) has been prompted by the somewhat different form of excitation curves obtained by measurements of the positron activities of N^{13} (a measure of total neutron production over all angles) and curves obtained by direct detection of the neutrons emitted in a small solid angle from a carbon target. Similarly Bailey, Freier, and Williams⁶ have discovered a very marked difference between the excitation curves for the production of neutrons taken at 0° and 90° to the bombarding deuteron direction.

APPARATUS

The Rice Institute Van de Graaff generator was used to accelerate the deuterons. Regulation of the beam-analyzing electromagnet (supplied now by a bank of high capacity storage batteries instead of a generator as in previous descriptions¹⁰), in combination with a potential stabilizer used to modulate an electron beam to the central electrode, gave a deuteron beam with an estimated resolution of 0.01 percent, or less, of the operating voltage.¹⁰ A beam current integrating device, designed by B. E. Watt,¹¹ gave a count on a mechanical register for each 0.0416 microcoulomb of deuteron charge collected at the target.

¹ W. E. Bennett and T. W. Bonner, *Phys. Rev.* **58**, 183 (1940); Bonner, Hudspeth, and Bennett, *Phys. Rev.* **58**, 185 (1940).

² Rogers, Hudspeth, Bonner, and Bennett, *Phys. Rev.* **58**, 186 (1940).

³ Bennett, Bonner, Hudspeth, Richards, and Watt, *Phys. Rev.* **59**, 781 (1941).

⁴ W. E. Bennett and H. T. Richards, *Phys. Rev.* **71**, 565 (1947).

⁵ Bailey, Phillips, and Williams, *Phys. Rev.* **62**, 80 (1940).

⁶ Bailey, Freier, and Williams, *Phys. Rev.* **73**, 274 (1948).

⁷ W. E. Stephens, *Rev. Mod. Phys.* **19**, 19 (1947).

⁸ Bonner, Evans, and Hill, *Phys. Rev.* **75**, 1398 (1949).

⁹ Dougherty, Hornyak, Lauritsen, and Rasmussen, *Phys. Rev.* **74**, 712 (1948).

¹⁰ Bennett, Bonner, Mandeville, and Watt, *Phys. Rev.* **70**, 882 (1946).

¹¹ B. E. Watt, *Rev. Sci. Inst.* **17**, 334 (1946).

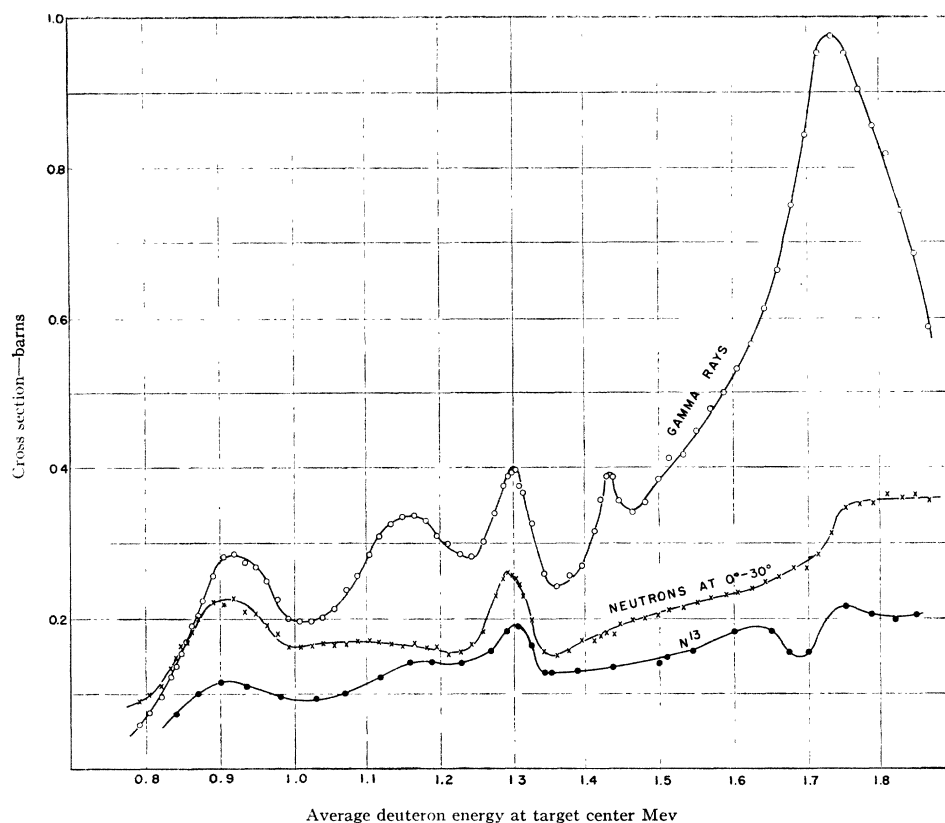


FIG. 1. Experimentally determined cross sections for the production of gamma-rays and N^{13} as a function of the energy of the bombarding deuterons. The yield of neutrons at $0^\circ \pm 30^\circ$ is not in terms of absolute cross section, but shows only relative shape of this yield function.

Some of the carbon targets were prepared by heating small volumes of ceresin wax in a vacuum and collecting a thin film of the wax on a polished silver disk. The target thickness in $\mu\text{g}/\text{cm}^2$ was determined by weighing the wax deposit on a microbalance. These targets did not collect carbon or deuterium in quantities sufficient to be detected as an increase in gamma-rays or neutrons. However, ceresin wax targets when deposited on very thin silver foils and exposed to the deuteron beam in a vacuum were found to become thinner. More satisfactory targets of pure carbon were made by cracking benzene in an atmosphere of helium onto the surface of silver foils.¹²

The counter used to determine the angular distribution of the neutrons was a cylindrical proportional counter of 4.5-cm inner diameter and a sensitive length of 4.5-cm, with a one mil tungsten wire. The counter was filled with one atmosphere of hydrogen and two percent methane, and operated at 1920 volts with an Atomic Instrument Company type 204-B Linear Amplifier.

DETERMINATION OF THE EXCITATION CURVES

The Gamma-Rays

In order to study the gamma-rays that accompany the short range proton group of reaction (2), a thin-walled, argon-filled Geiger counter was placed at about 8 cm from the center of the target and at about 90° to the direction of the deuteron beam. Large lead blocks were used to shield the counter from stray gamma-rays originating from carbon contamination along the beam path. In addition, about an inch of lead was placed around the counter to absorb scattered x-rays coming from the high voltage electrode of the Van de Graaff generator and annihilation radiation coming from the N^{13} . The counting rate for this arrangement of the Geiger counter became too great above 1.5 Mev. To keep the counting rate of the background radiation a small percentage of the total counting rate, a much smaller Geiger counter was used in preference to moving the original counter farther from the target. Background determinations were usually made by allowing the entire beam to fall just on the low energy side of the analyzing slit system, thus blocking the beam from the target, but giving essentially the same amount of radiation from carbon

¹² G. C. Phillips and J. Richardson (to be published).

contamination elsewhere along the beam path. Under these conditions this background counting rate was about 10 percent of the total rate for a very thin target that was only $3.0 \mu\text{g}/\text{cm}^2$ of ceresin. For the thicker targets, which were ordinarily used, the background was correspondingly less.

To determine accurately the cross section for the production of gamma-rays, a thick, pure graphite target was bombarded with deuterons of 1.30-Mev energy. Since the "thin" target relative yield of gamma-rays as a function of energy was known, the cross section for gamma-rays could be calculated from the observed thick target yield by integrating the thin target curve from 0 to 1.30 Mev. A small counter with a copper cathode was placed 17 cm from the target and surrounded by 1.4 cm of lead. A counter efficiency of 1.65 percent was assumed for the 3.1-Mev gamma-rays.¹³ The transmission of the lead under identical geometrical conditions was determined experimentally by means of 2.62-Mev gamma-rays from mesothorium and found to be 48 percent. These data gave a cross section at 1.30 Mev of 0.39 barn. The gamma-ray data of Fig. 1 are all adjusted to this cross section. It is to be noted that in Fig. 1 the narrow resonance at 1.435 Mev does not have its true cross section indicated since the target was much thicker than the true resonance width.

The 1.435-Mev gamma-ray resonance has been observed in the present experiment with ceresin targets varying in thickness from 3 to $80 \mu\text{g}/\text{cm}^2$, which correspond to the range of 1 to 27 kev for deuterons of 1.4 Mev. Just previous to one of these observations with a 6-kev target, the beam analyzing magnet was calibrated using the narrow gamma-ray resonances found in the bombardment of fluorine by protons for reference energies.¹⁴ The resonances at 0.669 and 0.8735 Mev were studied using the molecular beam, and so calibrations were obtained at 1.338 and 1.742 Mev. With this calibration the peak of the narrow resonance, which had previously been designated as the 1.43-Mev resonance,¹⁵ was found to occur at 1.435 Mev.

The shape of the 1.435-Mev resonance was determined with good resolution by using a 1-kev thick ceresin target and by changing the bombarding energy in 1-kev steps (see Fig. 2). The energy width measured midway between the dotted line of Fig. 2 and the resonance peak was found to be 5.5 kev. This is thought to be nearly the natural width since the energy spread in the beam was less than 1.4 kev and the target was very thin. The asym-

metry observed on the sides of this resonance has been observed in all of the twelve separate examinations of the resonance made during this experiment. By using a 1-kev target, the ratio of yields at the peaks of the 1.30-Mev and the 1.435-Mev were carefully determined to be 1.58 so that the cross section at resonance peak for the 1.435-resonance is 0.62 barn.

The angular distribution of the gamma-rays was investigated at 1.73 Mev with the small Geiger counter. To do this it was necessary to rotate the counter about the target at a distance greater than had been used for the excitation curves. The background count was ~ 10 percent of the true counts. No asymmetry was found greater than 10 percent, the background counting rate.

The N^{13} Positrons

The total yield of neutrons integrated over all angles can be obtained by observing the positron activity of the residual N^{13} nucleus of reaction (3). To determine the relative number of positrons as a function of energy a quantity of N^{13} was prepared by deuteron bombardment for a half-life (600 seconds) of a thin ceresin target on a thick silver disk placed at 30° to the beam. Immediately after this bombardment the positron activity was observed through an aperture of 0.5-cm diameter at a distance of 6.7 cm from the target and at right angles to the beam. A thin-walled Geiger counter

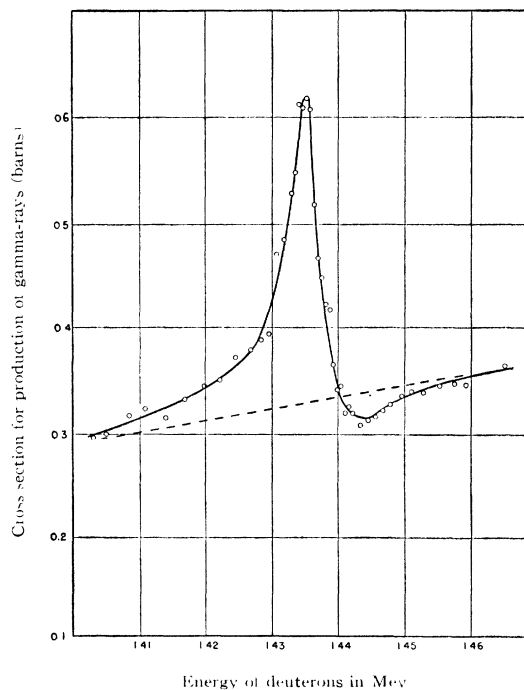


FIG. 2. Experimentally determined cross section in barns for the production of gamma-rays from a thin (1-kev) ceresin target at the narrow 1.435-Mev resonance.

¹³ Bradt, Gugelot, Huber, Medicus, Preiswerk, and Scherrer, *Helv. Phys. Acta* **19**, 77 (1946).

¹⁴ Bernet, Herb, and Parkinson, *Phys. Rev.* **54**, 398 (1938); R. G. Herb (1948) private communication.

¹⁵ Harris, Bonner, Evans, and Phillips, *Phys. Rev.* **73**, 649 (1948).

TABLE I. Data pertaining to the neutron and gamma-ray resonances.

Position of resonance Mev	Width at half-height keV (gamma-ray and neutron resonances)	Cross section at resonance peak in barns		Estimated relative intensity	
		Gamma-ray	Neutron	Gamma-ray	Neutrons
0.91	200	0.29	0.12	2.5	1.0
1.16	200	0.34	0.14	2.5	1.0
1.30	80	0.39	0.19	2.5	1.4
1.435	5.5	0.62		2.5	<0.1
1.62	200		0.19		1.0
1.73	200	0.97		9.0	
1.76	200		0.22		1.0

and a scale of 64 units were used to detect and record the number of positrons passing through the aperture. These measurements could not be used to determine the cross section because of backscattering of the positrons by the thick target backing, but the measurements gave relative yields. The cross section was determined by using a pure carbon target of $23.5 \mu\text{g}/\text{cm}^2$ on a thin silver foil ($7.75 \text{mg}/\text{cm}^2$). The target was placed normal to the deuteron beam and was bombarded for two half-lives (20 minutes). It was then quickly removed from the vacuum system and placed over the axis of a bubble-window Geiger counter, of window thickness $3 \text{mg}/\text{cm}^2$. The only absorbing material in the path of the positrons (end-point energy 1.24 Mev)¹⁶ was from the few centimeters of air and the counter window, and so nearly 100 percent of the positrons reached the sensitive region of the counter. The backscattering was negligible because of the thin target backing. Five independent measurements were taken at 1.30 Mev, with different target-counter separations; cross sections calculated from these measurements agreed within 10 percent.

If dN/dt is the number of N^{13} atoms decaying at the end of the bombardment of 20 minutes and n is the number of N^{13} formed per second during the bombardment, then

$$n = -4/3(dN/dt).$$

The experimental determination of dN/dt was obtained by plotting the logarithms of the number of Geiger counts obtained in 100-sec. intervals versus the time, for ten or so intervals after the cessation of bombardment. The times at which the points were plotted were 53 sec. from the beginning of each interval; i.e., the time at which the average number of positron counts over an interval would be expected. A line with a slope corresponding to a 10-minute half-life was drawn through the experimental points and extended back to the time bom-

bardment ceased, thus obtaining dN/dt at $t = 1200$ sec.

The thickness of the pure carbon target on the silver foil was obtained by comparing the ratios of the gamma-rays from this target and the thick target previously mentioned. These data were taken at 1.30 Mev with several geometrical arrangements of the counter about the targets, and gave the same ratio of yield to within 10 percent.

These measurements gave a cross section of 0.192 barn at 1.30 Mev for the production of N^{13} . The cross section for N^{13} production as a function of deuteron energy is shown in Fig. 1.

Table I gives the collected data pertaining to the neutron and gamma-ray resonances which were observed.

The Neutrons

The yields of neutrons at $0^\circ (\pm 30^\circ)$ as shown in Fig. 1 were obtained using a proportional counter 6 cm long and 1.6 cm in diameter filled with ethane at a pressure of 43 cm of Hg. The counter had a 4-mil wire and was operated in the proportional region at 2200 volts. These data are relative yields and not absolute cross sections.

The protons recoiling from the neutrons entering the counter were counted if their energy exceeded 300 keV, a value set by means of a discriminator attached to the output of a 204-B linear amplifier and before the input of an Atomic Instrument Company scale of 64. Gamma-rays from reaction (2) were not observable at this bias. A discussion of this type of counter has been given by Bethe and Barschall,¹⁷ who show that the counter sensitivity (assuming a proton track length small in relation to the counter diameter) is relatively insensitive to neutron energy.

Stray fast neutrons were partially shielded from the ethane counter by means of paraffin blocks interspersed among the lead blocks used to shield the gamma-ray counter. The counter was placed to subtend the largest solid angle possible at the target and was surrounded by about 5 cm of paraffin except on the side adjacent to the target. The paraffin shielding was effective in reducing the background counting rate.

The region in the vicinity of the 1.435-Mev gamma-ray resonance has been investigated for neutron resonances by means of the hydrogen-filled counter at laboratory angles of 45° , 90° , and 160° , and by means of the ethane counter at 0° . These data are shown in Fig. 3. There is no evidence for a sharp neutron resonance in this energy region.

¹⁶ K. Siegbahn and H. Slatis, Arkiv. f. Mat. Astr. O. Fys. **32A**, No. 9 (1945).

¹⁷ H. H. Barschall and H. A. Bethe, Rev. Sci. Instr. **18**, 147 (1947).

The Neutron Angular Distributions

Neutron angular distributions were taken at nine deuteron energies. The hydrogen-filled proportional counter with a linear amplifier, discriminator, and scaler was used to count the fast neutrons. This counter was non-directional and the bias setting corresponded to 0.15-Mev recoil protons as was determined by 3 bias *versus* counting rate curves for neutrons of different known energies. The counter was insensitive to the gamma-rays when set at its standard bias. By lowering the bias the over-all gain of the system was checked frequently with a radium source in a standard position. The counter was rotated about the target at a distance of 10 centimeters and it subtended a laboratory angle of $\pm 12^\circ$ at the target. The data was taken in 15° steps in the laboratory system. The carbon target was $135 \mu \text{g/cm}^2$ of ceresin wax on a thick silver disk. Data was taken by going several times through the angles and averaging. These averages are believed to be correct to within 10 percent. The background count was obtained by letting the beam fall on the defining slits only. The background was less than 10 percent.

The laboratory angles θ were converted into center of gravity angles φ by the relation

$$\sin(\varphi - \theta) = \left(\frac{E_{\text{deuteron}}}{78E_{\text{deuteron}} - 25.5} \right)^{\frac{1}{2}} \sin \theta$$

and the counting rates at each c.g. angle φ were corrected because of the variation of the solid angle subtended by the counter in the center of gravity coordinates.

The counting rates at each deuteron energy and at each angle were also corrected for the efficiency of the counter. The efficiency of the counter is given by

$$F = \sigma_0 \mathfrak{N} D P E_n^{-1}$$

where $\sigma_0 = 4.16 \times 10^{-24} \text{ cm}^2$ is the neutron-proton cross section at 1 Mev, E_n is the neutron energy in Mev, \mathfrak{N} is the number of hydrogen atoms/cm³, D is the effective depth of hydrogen gas that the neutrons see, and P is the probability that if a recoil occurs inside the counter it will be counted as a pulse above the bias. P is not only a function of bias energy (the size of the "cone" of recoiling protons that will release the bias energy) but also of the ratio of proton track length L to counter radius R . P and F have been calculated for the counter.¹⁸

The efficiency of the hydrogen counter may also be experimentally determined. By integration of the observed angular distribution curves and comparison of this total yield into 4π -radians with the

yield of N^{13} as determined from the positron measurements an efficiency of the counter for the average neutron energy in that distribution is obtained. These experimental determinations agreed with the theoretical within about 15 percent. The corrections in yield were made using the theoretical efficiency. The angular distributions are shown in Fig. 4 and are believed to be correct to within 10 percent. A subsequent paper will analyze the neutron angular distributions.

CONCLUSIONS

In general, reactions (2) and (3) are competing reactions, and at the three lower resonances there are about equal probabilities of breaking up with the emission of neutrons or short range protons. However, certain marked differences in the resonance phenomenon of reaction (2) and (3) can be noted. There are resonances for neutron emission at 1.62 and 1.76 Mev that do not correspond to the large gamma-ray resonance at 1.73 Mev. It is interesting that the 1.73-Mev gamma-ray resonance occurs at the point of maximum slope of the neutron cross section suggesting that the neutrons show something akin to dispersion about this point. Although the resonance at 1.62 Mev was not observed at 0° , similar curves when counting neutrons at 90° have been obtained by Williams *et al.*⁶ for this region.

At the 1.435-Mev resonance there is no indication of a neutron resonance within the limits of the experiment. If a neutron resonance exists it must be relatively weak and $\sigma_\gamma/\sigma_{\text{neut.}} > 25$. The shape of the 1.435 resonance for gamma-rays is markedly asymmetrical; this resonance has been observed many times with different targets and different experimental arrangements and so it is certain that the asymmetry is real. It is interesting that a

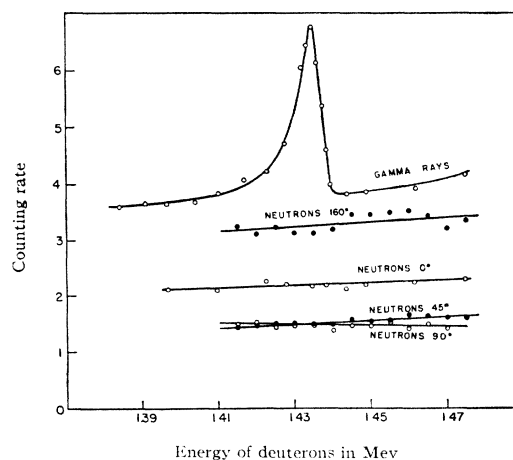


FIG. 3. The yields of gamma-rays and neutrons at 0° , 45° , 90° , and 160° at the narrow 1.435-Mev resonance. The 45° , 90° , and 160° neutron counting rates are correct ratios while the 0° counting rate is arbitrary.

¹⁸ J. C. Harris and G. C. Phillips (to be published).

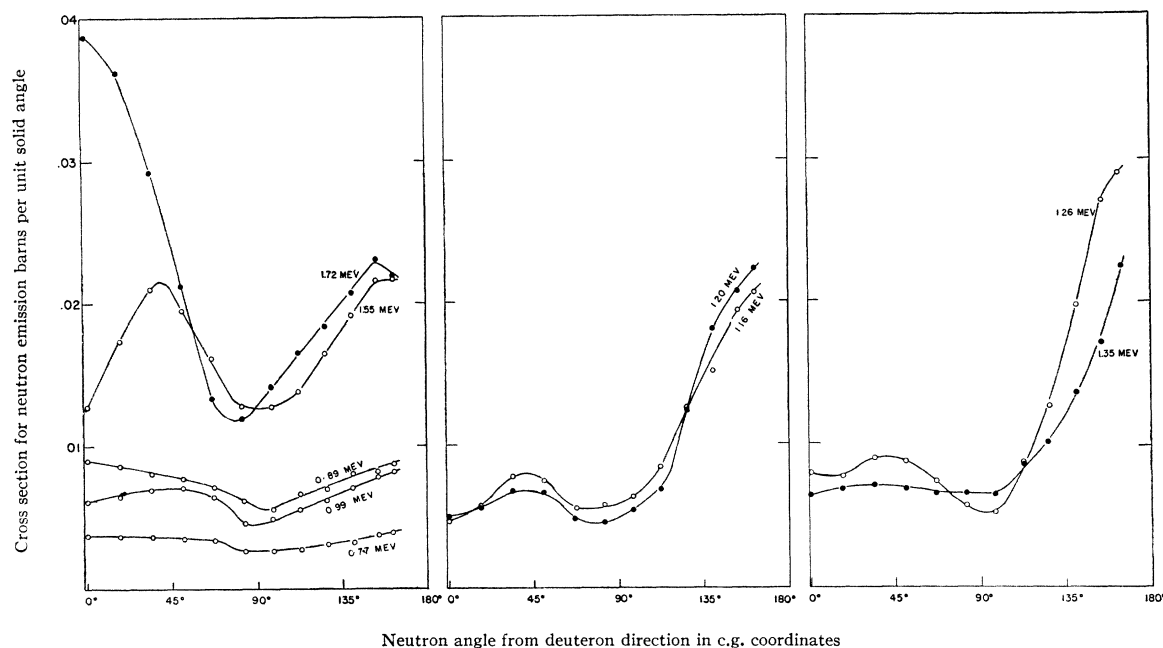


FIG. 4. The angular distributions of the neutrons in center of gravity coordinates for the deuteron bombarding energies indicated.

superposition of resonance and dispersion formulas will produce this shape. Similar shapes have been observed in the resonances associated with the bombardment of fluorine by protons.

The ratio of the cross sections for reactions (2) and (3) are several orders of magnitude different from what would be expected if the deuteron is captured by the C^{12} nucleus and the excited $^*N^{14}$ nucleus subsequently breaks up with the emission of either a short-range proton or a neutron. At the 0.91-Mev resonance the ratio of the number of short-range protons to neutrons is $\sigma_p/\sigma_n \sim 2.5$. At this bombarding energy the short-range protons have only an energy of 230 kev in the center of mass coordinates; the penetrability through the Coulomb barrier for $l=0$ is only approximately 10^{-4} . The effect of the Coulomb barrier should make the emission of neutrons far more likely than the emission of short-range protons; and indeed the cross section for reaction (2) should be much smaller than is observed.

In the case of the narrow resonance at 1.435 Mev the penetrability of the short-range protons is approximately $1/30$, while the observed ratio $\sigma_p/\sigma_n > 25$, and so the discrepancy in the expected number of neutrons and short range protons is > 750 .

Since the discrepancies are so large in the cross sections, it seems possible that the assumed mechanism of disintegration is wrong. There is the possibility that the entire deuteron does not enter the C^{12} nucleus in the case of low energy proton emission. The difficulty in proposing an Oppenheimer-Phillips reaction to interpret the results³ is that sharp resonances like the 1.435 Mev should not be observed. The lifetime of the nucleus would be expected to be of the order of the transit time of the proton across the nucleus, or about $5 \times 10^{-13} / 10^9 = 5 \times 10^{-22}$ sec. From the relation $\Delta E \cdot \Delta t \sim \hbar$, a width of 1.3 Mev is obtained, and this is completely incompatible with the sharp resonance that is observed. In view of this difficulty in the width of the level it seems that some other mechanism must be sought.

Critchfield has suggested an explanation in terms of a special model of the excited $^*N^{14}$ nucleus with a large rotational angular momentum. If the $^*C^{13}$ has high rotational angular momentum the probability of transition from $^*N^{14}$ to $^*C^{13}$ may be much greater than to a N^{13} nucleus without this high angular momentum.

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