Neutron-Proton Scattering Using Organic Crystal Scintillation Detectors*†‡

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The angular distribution of protons recoiling from neutrons of 13.7 Mev and 28.4 Mev has been investigated in the region of 0° to 90° in the center-of-mass system. Organic scintillation crystals of anthracene and stilbene were used as both sources and detectors of the recoil protons. The angular distribution observed at 13.7 Mev is consistent with spherically symmetric scattering, while the results at 28.4 Mev show an anisotropy with a favoring of scattering of the neutrons in the backward direction.

I. INTRODUCTION

THE experimental study of neutron-proton scattering has long been expected to be a fruitful source of information leading to knowledge of nuclear forces. Consequently, a very large amount of both experimental and theoretical work covering a wide variation in energy has been performed on the neutron-proton scattering problem. It is now generally accepted that the scattering is isotropic in the center-of-mass system (without preclusion of an asymmetry of approximately five percent) for energies below about 15 Mev.[1] At approximately 20 Mev,[2] there are indications of an asymmetry favoring scattering in the backward direction. This is definitely confirmed at 27 Mev[3] and becomes more pronounced at the higher energies of 40, 90,[4] and 260 Mev.[5] In the present experimental work, the scattering of fast neutrons by protons has been studied by the observation of recoil protons in an organic scintillation crystal which was used as both source and detector of the recoils.

II. EXPERIMENTAL APPARATUS AND PROCEDURE

The neutrons for the present experiments were obtained from the $D(d,n)He^3$ and the $T(d,n)He^4$ nuclear reactions. Because of the very low yield of high energy neutrons from the $T(d,n)He^4$ reaction, a technique of using an organic scintillation crystal as both a source and detector of recoil protons was employed. Since this technique was at the time unexploited, it was deemed desirable to check it at a lower energy, where the angular distribution is known to be isotropic.

A plan view of the physical arrangement of the experimental apparatus is shown in Fig. 1. Deuterons of 11.75 ± 0.12 Mev[6] accelerated by the University of Illinois cyclotron were collimated by a set of lead slits, passed through a thin gas target, and were stopped in a 0.030-inch lead foil approximately 13 feet from the target. A scintillation counter was accurately positioned in a light-tight copper box such that a line from the center of the gas target to the crystal made an angle of $11.69°\pm0.05°$ with the deuteron beam direction.

The gas target used by Laughlin and Kruger[1] was used to contain the deuterium for the source of neutrons from the $D(d,n)He^3$ reaction. A somewhat similar target, shown in Fig. 2, was filled to about 25 cm of Hg pressure with a tritium-hydrogen mixture of 60 percent tritium for the second neutron source. When not in use, the tritium was stored as uranium tritide in the small iron container. Filling the target was accomplished by heating the UT_3 to approximately 450°C, which produced decomposition and resulted in essentially complete evolution of the tritium gas into the target chamber.[7] The gas volume was emptied by opening the bellows valve between the target chamber and the uranium container. This permitted the tritium to react with the uranium forming the UT_3 and emptying the target volume to a pressure of approximately 10^{-4} mm of Hg.

Because of the finite size of the target, the angle of emission of the neutrons from the nuclear reactions varied over 7.6°, and this plus the degradation of the deuteron energy in passing through the gas in the target resulted in a finite energy spread in the neutrons which

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‡ A preliminary report of this work was given at the Denver meeting of the American Physical Society in July, 1952 [Phys. Rev. 88, 162 (1952)].

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[1] Kruger, Shoupp, and Stallman, Phys. Rev. 52, 678 (1937); W. F. Caplehorn and G. P. Rundle, Proc. Phys. Soc. (London) A64, 546 (1951); J. S. Laughlin and P. G. Kruger, Phys. Rev. 73, 197 (1948); H. H. Barshall and R. F. Taschek, Phys. Rev. 75, 1819 (1949).

[2] E. W. Baldwin, Phys. Rev. 83, 495 (1951).

[3] Brolley, Coon, and Fowler, Phys. Rev. 82, 190 (1951).

[4] Hadley, Kelly, Leith, Segrè, Wiegand, and York, Phys. Rev. 73, 1114 (1948); Brueckner, Hartsough, Hayward, and Powell, Phys. Rev. 75, 555 (1949).

[5] Kelly, Leith, Segrè, and Wiegand, Phys. Rev. 79, 96 (1950).

[6] This energy was determined by measuring the range of the deuterons in air. This was done by passing the beam through a monitor consisting of a double-end-window proportional counter, whose stopping power was known, and then into a thin zinc sulfide screen placed on the end of a movable 5819 photomultiplier tube. This second detector had a threshold detection energy as measured with a polonium alpha-source of less than 30 kev. This measurement was checked by determining the range in photographic emulsion of protons accelerated by the cyclotron. The energy of the protons was determined from the range-energy relation in emulsion given by J. Rotblatt, Nature 167, 550 (1951), and the energy of the deuterons was derived from this value of the proton energy.

[7] For a description of the properties of uranium tritide, see F. H. Spedding *et al.*, Nucleonics 4, No. 1, 4 (1949).

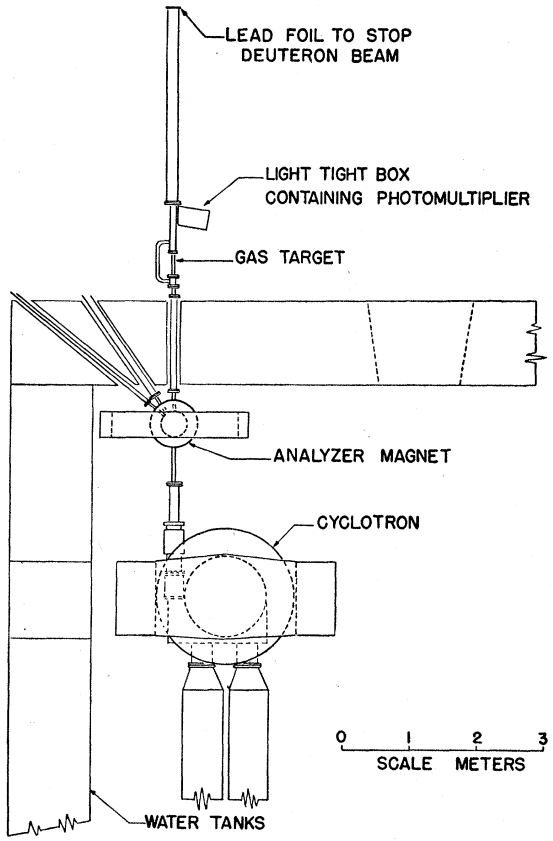

FIG. 1. Arrangement of experimental apparatus. The amplifier, discriminator, and associated electronic equipment were located in the cyclotron control room.

were to be scattered. The neutrons from the $T(d,n)He^4$ reaction had a mean energy of 28.4 Mev with a spread of 0.50 Mev, while those from the $D(d,n)He^3$ reaction were of mean energy 13.7 Mev with a spread of 0.80 Mev.

The hydrogenous radiator and detector consisted of an organic scintillation crystal of either anthracene or stilbene mounted in contact with the photosensitive surface of a selected RCA 5819 photomultiplier tube. The crystal was covered with a thin aluminum reflecting foil. The photomultiplier tube was magnetically shielded with four concentric cylindrical shields of annealed Permalloy, magnetically insulated from each other. The integrated output pulses from the photomultiplier were applied through a cathode follower to a Los Alamos type model 100 linear pulse amplifier,[8] through a pulse shaping circuit and into a twelve-channel pulse amplitude analyzer. The discriminator levels of the analyzing system were set by feeding pulses of known height into the system from an electronic pulse generator. Several checks on the stability of the electronic system showed the discriminator levels to be constant to within ± 0.05 volt over periods as long as 24 hours with the levels set at approximately 7 volts apart. This shift in the levels included the net effects of possible variations in the pulse generator, amplifier, pulse shaping circuit, and analyzer.

The voltage to the dynodes of the photomultiplier tube was supplied by a voltage dividing network of precision resistors connected across the output of a negative electronic power supply. This output voltage was monitored with an auxiliary circuit, and the voltage was maintained constant to within ± 0.1 volt by manual adjustment of the supply during the taking of data.

In order to obtain an absolute energy response of the scintillation detectors, pulse-height distributions from a well-collimated polonium alpha-particle source were taken in a standard geometry before and after each experimental run. The response of the counter to these alpha-particles had been normalized to the response of protons of known energy from the cyclotron. Thus, knowing the relative pulse height *vs* energy relation for the crystals used in addition to the above normalization factor, the response of the counters during any experimental run could immediately be converted to the energy of the protons originating the fluorescence.

Because of the large amount of background radiation produced by the cyclotron, the experimental data were obtained in a series of "runs," half of which were taken with the gas target filled, while the others were background runs taken with the gas target evacuated. During these runs, the integrated cyclotron beam to the main target was measured with a charge measuring circuit; the neutron flux inside the cyclotron shielding tanks was monitored with small gold foils using the well-known reaction

$$Au^{197}+n\rightarrow Au^{198}\rightarrow\beta^-+Hg^{198};$$

the gamma-intensity in the vicinity of the neutron source was measured with a Geiger counter in a standard geometry. These three measurements were all relative, and they were sufficient to determine the background

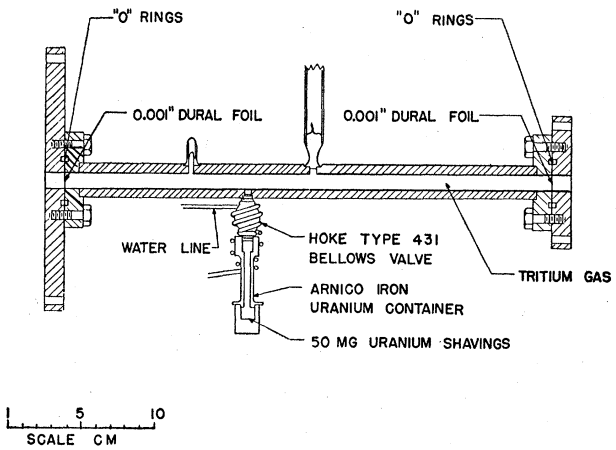

FIG. 2. Gas target used to contain tritium gas for source of high energy neutrons.

[8] W. C. Elmore and M. Sands, *Electronics* (McGraw-Hill Book Company, Inc., New York, 1949).

TABLE I. Angular distribution data at 13.7 Mev.

Center-of-mass recoil angle	Number of counts in recoil angular interval	Correction factor for finite size of crystal	Center-of-mass solid angle	Relative No. of recoils per steradian
15.7±2.1	5734	1.169	0.1456	0.61±0.16
36.1±3.7	6510	1.148	0.0990	1.00±0.021
45.0±2.8	6526	1.124	0.0990	0.98±0.027
52.8±2.4	6833	1.100	0.1077	0.92±0.024
60.2±2.0	7518	1.080	0.1063	1.01±0.035
67.0±1.8	7509	1.056	0.1049	1.00±0.036
73.4±1.6	8191	1.038	0.1047	1.07±0.040
79.7±1.4	7764	1.012	0.1080	0.96±0.055
86.3±1.2	9898	0.9965	0.1220	1.07±0.086

with a precision of ±5 percent plus the normal statistical variations.

III. ANALYSIS OF DATA

The data were analyzed in three groups which can be described as follows:

(1) the data for 13.7-Mev neutrons incident on anthracene;
(2) the data for 28.4-Mev neutrons incident on anthracene;
(3) the data for 28.4-Mev neutrons incident on stilbene.

The treatment included the determination of the actual number of counts in each pulse-height interval resulting from the incidence on the crystal of the nearly monoergic neutrons from the source. This was done by subtracting the proper background from the data taken with the target filled with gas. For the data at 28.4 Mev, the background counting rate in the pulse-height interval corresponding to protons recoiling at 47° in the laboratory system was approximately two-thirds of the total counting rate and decreased to about one-half and one-third of the total counting rate at recoil angles of 43° and 37°, respectively. At recoil angles of less than 33°, the background was quite small, being only about 3 to 4 percent of the total counting rate. For the data at 13.7 Mev, the background counting rate was about one-half of the total counting rate at proton recoil angles of 43° in the laboratory system and decreased to about 30 percent of the total counting rate at recoil angles of 36°. At recoil angles of less than 30°, the background rate varied between 5 and 20 percent of the total rate. The pulse-height intervals as set by the discriminator levels in the pulse analyzer were then converted to recoil proton energy intervals by making use of the pulse height *vs* energy relations for the crystals along with the energy normalization factor obtained with the polonium alpha-particles. The proton energies corresponding to the discriminator settings could immediately be converted to laboratory recoil angles by means of the following equation:[9]

$$\cos^2\theta = E_P/E_n,$$

where E_p = recoil proton energy; E_n = mean neutron energy. An angular distribution in center-of-mass coordinates was then obtained in the usual manner, with the small relativistic correction for the data at 28.4 Mev included for rigor.

The response curve for the anthracene counter was taken from known results[10–12] for energies up to 16.4 Mev, plus an extrapolation from this energy through a point obtained at 29.2 Mev by observation of the pulse heights resulting from the maximum energy protons recoiling from neutrons of 29.2 Mev. This continuation of the curve was found to agree with that predicted by Birks,[13] if the constants in his equation are calculated from the well-investigated lower energy region.

The presently available data for stilbene response[10,12] extend up to about 14.7 Mev. To extend this response curve to the energy needed for analysis of these data, the response of stilbene at 28.7 Mev was measured with neutrons of that energy. The response curve was also calculated for the energy region 5.5 to 30 Mev after the method of Birks.[13] These two curves were found to be in essential agreement, and the calculated curve was used in the analysis.

A correction to the number of counts in each recoil angular interval was required because of the finite size of the crystals. This is complicated by the fact that the recoil protons which originate at points in the crystal such that they pass out of the crystal produce pulses of smaller size than those recoils of the same energy completely stopped in the crystal. These smaller pulses are then subject to confusion with the pulses from recoil protons of less energy wholly stopped in the crystal. The

TABLE II. Angular distribution data at 28.4 Mev taken with anthracene crystal.

Center-of-mass recoil angle	Number of counts in recoil angular interval	Correction factor for finite size of crystal	Correction factor for (n,p) reaction protons	Center-of-mass solid angle	Relative No. of recoils per steradian
11.3±3.3	1776	2.146	1.00	0.0764	1.50±0.58
29.3±5 0	2462	1.919	1.00	0.1145	1.24±0.037
41.0±3.4	2521	1.720	1.00	0.1117	1.17±0.039
50.3±2.6	2778	1.517	1.00	0.1186	1.07±0.032
58.8±2.2	3140	1.366	1.00	0.1227	1.05±0.043
66.5±2.1	3415	1.237	1 00	0.1139	1.11±0.045
73.5±1.6	3873	1.136	1.00	0.1160	1.14±0.059
80.2±1.4	4058	1.049	0.959	0.1119	1.10±0.077
87.0±1.2	5343	0.9914	0.780	0.1242	1.00±0.083
94.2±0.9	6012	0.9345	0.714	0.1257	0.96±0.15

[9] Relativistic corrections will modify this equation slightly for the 28.4-Mev data, but the corrections to the values of θ are of the order of less than 0.5 percent.

[10] Taylor, Jentschke, Remley, Eby, and Kruger, Phys. Rev. **84**, 1034 (1951).

[11] Franzen, Peele, and Sherr, Phys. Rev. **79**, 742 (1950).

[12] W. G. Cross, private communication.

[13] J. B. Birks, Phys. Rev. **84**, 364 (1951); Proc. Phys. Soc. (London) **A64**, 874 (1951).

determination of these contributions was calculated[14] by first finding the fraction of the proton recoils in each recoil angular interval which were not completely stopped in the crystal. This was done by obtaining an expression for this quantity in terms of the recoil angle, the differential cross section for the neutron-proton scattering process, the length of the recoil in the crystal (which is a function of the recoil angle), and the dimensions of the crystal. This expression was then numerically integrated over the recoil angular intervals to give a correction factor for each angular interval. In the evaluation of the integrals, the differential cross section in the center-of-mass system was taken as a constant at 13.7 Mev, and at 28.4 Mev the relative cross section given at 27.2 Mev by Brolley *et al.*[3] was used. Since these latter two energies are comparable and, further, since the recoil angular intervals were of the order of only four degrees, so that the variation of the cross section over the interval was quite small, this approximation in the second case should certainly be valid.

Those recoils which were not completely stopped in the crystal then gave pulses which corresponded to protons recoiling at larger angles and thus were incorrectly counted in the recoil angular intervals which included these larger angles. To obtain the number of pulses incorrectly counted in each interval, the pulse height actually observed was determined as a function of the recoil angle and of the origin of the proton recoil in the crystal. Then from purely geometrical considerations, since the origin of the recoil is equally probable anywhere in the crystal, the desired correction factor was obtained as a function of the correction factor for the proton recoils which were not completely stopped in the crystal. These two factors were then combined into a single correction factor for each recoil angular interval and are presented in that manner in the experimental results.

Because of the carbon component of the crystals, the effects of possible nuclear reactions resulting from neutrons incident on carbon were considered in the analysis of the data. Possible reactions include $C^{12}(n,\alpha)$-

TABLE III. Angular distribution data at 28.4 Mev taken with stilbene crystal.

Center-of-mass recoil angle	Number of counts in recoil angular interval	Correction factor for finite size of crystal	Correction factor for (n,p) reaction protons	Center-of-mass solid angle	Relative No. of recoils per steradian
11.3±3.8	1079	1.640	1.00	0.0764	1.17±0.46
29.3±5.3	1400	1.561	1.00	0.1145	0.96±0.028
41.0±3.5	1621	1.452	1.00	0.1117	1.06±0.030
50.3±2.6	1806	1.343	1.00	0.1186	1.03±0.052
58.8±2.4	1972	1.234	1.00	0.1227	1.00±0.042
66.5±2.0	2147	1.164	1.00	0.1139	1.11±0.052
73.5±1.7	2286	1.085	1.00	0.1160	1.08±0.064
80.2±1.5	2609	1.043	0.976	0.1119	1.20±0.12
87.0±1.3	3398	1.014	0.808	0.1242	1.13±0.14

[14] M. E. Remley, Ph.D. thesis, University of Illinois, Urbana, Illinois, 1952 (unpublished).

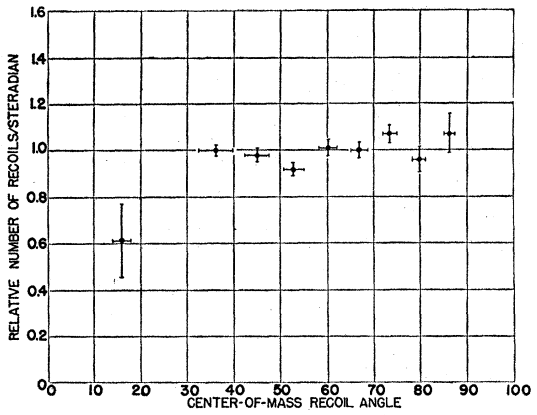

FIG. 3. Experimental angular distribution of recoil protons from 13.7-Mev neutrons; data taken with an anthracene crystal.

Be^9, $C^{12}(n,3\alpha)n$, and $C^{12}(n,p)B^{12}$. Because of the large threshold energies required for these reactions and, further, because of the smaller fluorescence efficiency of the crystals for alpha-particles, none of the reaction products from these gave pulses which could be confused with the recoil protons in the angular region investigated at 13.7 Mev. The first two reactions also gave no pulses subject to confusion with the higher energy neutrons, but reaction protons from the $C^{12}(n,p)B^{12}$ reaction can be produced with energies from 16.0 Mev to 9.4 Mev (assuming no excited states in the residual B^{12} nucleus). These protons produced pulses which could be confused with the recoil protons. It had been hoped that if the cross section for this reaction was appreciable, a correction could be made by the taking of data with two crystals with different carbon to hydrogen ratios. Unfortunately, the data were not accurate enough in the appropriate angular intervals to permit a meaningful correction of this type to be determined.

Available information from which a feasible correction might be determined is a measurement of the total cross section of the $C^{12}(p,n)N^{12}$ reaction. A value of 0.02 barn ±100 percent for this cross section has been obtained at 32 Mev by the linear accelerator group at the University of California.[15] From a theoretical standpoint this cross section should be approximately the same as that for the $C^{12}(n,p)B^{12}$ reaction. Therefore, a correction was calculated on the assumptions that the total cross section for the (n,p) reaction was 0.02 barn, that the protons were emitted isotropically in the laboratory system, and further that there were no excited states produced in the residual B^{12} nucleus.

IV. RESULTS AND CONCLUSIONS

The results are summarized in Tables I, II, and III and are shown in Figs. 3 and 4 as a function of the proton recoil angle in the center-of-mass system. The uncertainties shown are the probable errors for each

[15] L. Alvarez and H. Tyren, private communications.

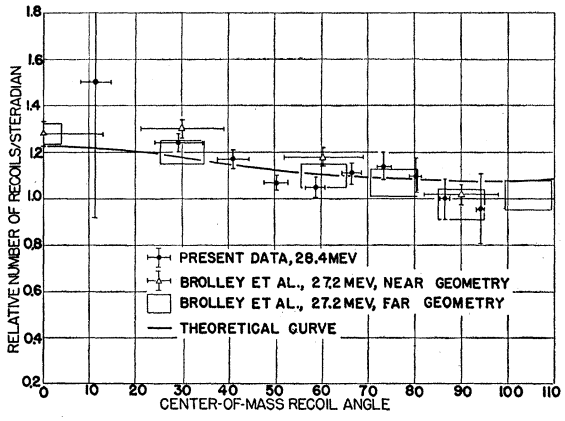

FIG. 4. Experimental angular distribution of recoil protons from 28.4-Mev neutrons; data taken with anthracene crystal. The theoretical curve is one calculated by Christian (see reference 3) for scattering at 27.2 Mev with a Yukawa potential of range 1.35×10^{-13} cm plus a tensor force.

point as determined by combining the probable errors in the various quantities required to obtain the angular distribution by the least square method.

The major sources of uncertainty in each point other than the statistics of counting include that resulting from the background from the cyclotron, the uncertainty in the pulse height *vs* energy curves of the scintillation crystals, and uncertainties in the calculations required by the finite size of the crystal detectors. The uncertainties in the recoil angles and an additional uncertainty in relative cross section at each angle are a result of the experimental errors in determining the energy normalization factor required to convert the response of the scintillation counter to recoil proton energy. The results in the first recoil angular interval are extremely sensitive to this latter factor, and the large uncertainties in this particular interval are primarily owing to the variation in the energy normalization factor.

A study of the results at 13.7 Mev shows that with the exception of the point at 15.7° the distribution is consistent with spherically symmetric scattering. This point is open to serious question because of the extreme dependence on the exact value of the energy normalization factor. The probable error in this normalization factor was 2.7 percent. An analysis of the data was made with a normalization factor different from the experimentally measured factor by 3.0 percent, and the results then gave a completely isotropic angular distribution. Thus, it seems reasonable to conclude that the technique used in this experiment is applicable in the recoil angular region from 0° to 90° in the center-of-mass system, keeping in mind that there will be considerable uncertainty in the results for the first recoil angular interval unless the absolute sensitivity of the apparatus is known with a precision of the order of five times better than that obtained here.

Cross,[16] at Chalk River Laboratory, has also used the technique described here to obtain the angular distribution of neutron-proton scattering at 14 Mev. His results, for which he quotes an uncertainty of ±5 percent, also show spherical symmetry.

The results from the data taken at 28.4 Mev with the anthracene crystal are shown in Fig. 4 along with those of Brolley *et al.*[3] at 27.2 Mev. A study of these shows that the data presented here can be consistent with those at 27.2 Mev. Because of the relatively small difference in energy, the distributions should be expected not to differ radically.

In view of the assumptions under which the corrections for the (n,p) reaction were applied, there must be some doubt placed on the quantitative conclusions that can be drawn from the results of this investigation in the regions in which the reaction protons are confusable with the recoil protons. However, it seems reasonable to conclude that the scattering of the neutrons is anisotropic in the center-of-mass system with a favoring of scattering in the backward direction. The ratio of the differential cross section for backward scattering to that for scattering at 90° can be stated only with a large uncertainty, not only because of the confusable reaction protons, but also because of the inherent large uncertainty in the point at 11.3°. There is an indication, however, that the ratio is greater than that of 1.15 predicted by the calculation of Christian[3] for the scattering at 27.2 Mev using a Yukawa potential plus the inclusion of a tensor force. Also, it appears that the ratio obtained here is certainly greater than the 1.06 reported by Baldwin[2] at 18 to 21 Mev. More definite conclusions must await further knowledge of the (n,p) reaction.

In general, it can be stated that the technique employed here for the study of the scattering does combine the advantages of the continuous sensitivity of the nuclear emulsion and the capacity of simultaneous detection over all solid angles of the cloud chamber. These make it appropriate for use with a low yield. The inherent disadvantages include the fact that the recoil angles must be obtained from an indirect measurement; the pulse analysis must be converted to proton energy intervals (from which the recoil angular intervals are inferred) by means of an additional determination. In addition the large background of neutrons and gamma-rays from the cyclotron results in a high rate of background counting which tends to overshadow the counts from the protons recoiling at larger angles (angles greater than about 40° in this particular experiment) from the monoenergetic neutrons produced in the source. Also, the carbon constituent of the crystals gives rise to nuclear reactions which prevent the obtaining of valid data for the large recoil angles.

The authors would like to express their appreciation to the members of the cyclotron staff for valuable assistance in performing this work. The aid of G. R. Briggs, who designed the pulse generator and discriminator, is most gratefully acknowledged.

[16] W. G. Cross, Phys. Rev **87**, 223 (1952).