

functions for the metal ions. These are not known, but on the assumption that certain of the coefficients are equal the expression for the spontaneous polarization simplifies with the result  $P^0=0.02(T_0-T)^{\frac{1}{2}}$  coulomb  $m^{-2}$ . Matthias and Remeika<sup>4</sup> found a maximum spontaneous polarization of about 23 microcoulombs  $cm^{-2}$ , with which this estimate is in satisfactory agreement, suggesting a critical temperature  $T_0$  in the neighborhood of 600°C or higher. It must be emphasized that we present here little more than an order-of-magnitude estimate, based on the equating of certain coefficients of Eqs. (1) and (2) which are not actually equal, but only of the same order. For the region above the upper Curie point, the same equations give a dielectric constant of the order of  $10^6/(T-T_0)$ . Experiments have not yet been made in the latter temperature region. Both the spontaneous polarization and the dielectric constant expressions have the expected form, of course, and the coefficients themselves are in the same range of values as have been found for other ferroelectric materials.

The author is greatly indebted to Professor J. C. Slater, who suggested this problem and offered much helpful advice during its study. He also wishes to thank his wife, Fronie French Schweinler, for performing

many of the numerical calculations. The financial support of an AEC Predoctoral Fellowship and of the ONR is gratefully acknowledged.

#### APPENDIX

##### The Polarizability of Oxygen in $Li_2O$

Lithium oxide,  $Li_2O$ , has a cubic space lattice with  $a_0=4.61\text{\AA}$ . The space group is  $O_h^8$ , and there are four  $Li_2O$  per unit cell. The structure is the fluorite ( $CaF_2$ ) structure. In this structure all ionic positions are determined from the symmetry alone; that is, there are no free crystallographic "parameters." When equations like Eq. (11) are written, with the dyadics  $D_{ij}$  appropriate to this structure, and the left-hand sides  $\epsilon_0 E_i$  are set equal to  $P_i/X_i$ , it is found that the Lorentz factor of  $\frac{1}{3}$  is strictly justifiable for the total polarization of both types of ions in the fluorite structure. Using the Maxwell relation  $K=n^2$  between the dielectric constant  $K$  and the index of refraction  $n$ , we find that  $8X_{Li}+4X_O=3(n^2-1)/(n^2+2)$  or  $(2\alpha_{Li}+\alpha_O)/\epsilon_0=3a_0^3(n^2-1)/4(n^2+2)=26.6\text{\AA}^3$ , where  $n=1.644$  (for visible light) was used in evaluating the constant. The polarizability of the two lithiums is very small, and so this is essentially the polarizability of the oxygen ion alone. Perhaps the best estimate of the lithium polarizability is that of Bragg,<sup>21</sup> who found from the index of refraction of the alkali halide crystals  $\alpha_{Li}/\epsilon_0=0.75\text{\AA}^3$ . This in conjunction with the above leads to the value  $\alpha_O/\epsilon_0=25.1\text{\AA}^3$ .

<sup>21</sup> W. L. Bragg, *The Crystalline State* (G. Bell and Sons, London, 1949), first edition, Vol. I, p. 182.

### Total Cross Sections for 14-Mev Neutrons\*†

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With deuterons from the electrostatic generator of the Department of Terrestrial Magnetism incident on a tritium-zirconium target, and with the neutrons detected by a terphenylxylene scintillator and photomultiplier, total cross sections of C, H, D, O, and N for  $14.10\pm 0.05$  Mev neutrons were measured. The effective neutron energy was evaluated from the kinematics of the  $T(d,n)He^4$  reaction, the dependence of the reaction's cross section on energy and angle, and the multiple scattering of the deuterons and their rate of energy loss in the target. A least squares analysis of the transmissions of six thicknesses each of carbon and polyethylene (40 to 85 percent transmissions) yielded, for total cross sections  $\sigma$  in barns:  $\sigma_C=1.279\pm 0.004$  and  $\sigma_H=0.689\pm 0.005$ . Transmissions of light and heavy water gave  $\sigma_D=0.803\pm 0.014$  and  $\sigma_O=1.64\pm 0.04$ . Transmission of melamine gave  $\sigma_N=1.7\pm 0.1$ .

#### I. INTRODUCTION

IF an analysis of neutron-proton interactions is not to be seriously complicated by either relativistic effects or the presence of nonzero angular momentum states, the neutron energy should not greatly exceed 15 Mev. At this energy, capture is negligible, so that the total cross section is purely a scattering cross section.

There have been a number of measurements of total cross sections for 14-Mev neutrons, for a number of

elements,<sup>1-5</sup> and while all of the earlier work shows a neutron-proton cross section of about 0.7 barn, the state of the technique was such as to leave considerable spreads in the values of both the cross section and the relevant energy. The advent of  $d,T$  neutrons has made it feasible to reduce these spreads markedly, as appears in the work of Meyer and Nyer<sup>6</sup> and in the work we

<sup>1</sup> Salant, Roberts, and Wang, *Phys. Rev.* **55**, 984 (1939).

<sup>2</sup> E. O. Salant and N. F. Ramsey, *Phys. Rev.* **57**, 1075 (1940).

<sup>3</sup> Ageno, Amaldi, Bocciarelli, and Trabacchi, *Phys. Rev.* **71**, 20 (1947).

<sup>4</sup> W. Sleanor, *Phys. Rev.* **72**, 207 (1947).

<sup>5</sup> A. H. Lasday, *Phys. Rev.* **81**, 139 (1951).

<sup>6</sup> D. I. Meyer and W. Nyer, Los Alamos Report No. 1279 (1951).

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† A brief account of this work was presented at the Chicago meeting of the American Physical Society, October, 1951.

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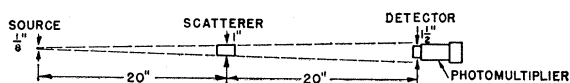


FIG. 1. Geometrical arrangement of the experiment. Deuteron beam is incident at source spot at an angle of  $89.27^\circ \pm 0.1^\circ$  with direction of observation.

report here, which was being carried on concurrently. These values are in essential agreement; the somewhat higher accuracy claimed for our values is the result of, perhaps, the choice of a more efficient detector. Whereas Meyer and Nyer, like the earliest workers in this region,<sup>1-3</sup> used  $\text{Cu}(n,2n)$  radioactivity, we used a thick hydrocarbon scintillation counter.

The following account describes the measurements of total cross sections of H, D, C, N, and O. In the first three elements a special effort to attain high accuracy was made. A theoretical analysis of the  $n-p$  value by one of us (G.A.S.) is made in the accompanying article.<sup>7</sup>

## II. METHOD AND APPARATUS

The cross section of a substance was determined from the measurement of the transmission of several different thicknesses. Figure 1 illustrates the geometrical arrangement in the horizontal plane.

The source of neutrons was the reaction  $\text{T}(d,n)\text{He}^4$ . A  $400 \pm 1$  kev deuteron beam was obtained from the small electrostatic accelerator of the Carnegie Institution's Department of Terrestrial Magnetism. The beam was collimated, through a diaphragm, and was incident at an angle of  $15^\circ$  from the vertical on a thick zirconium-tritium target. The direction of observation formed an angle of  $89.27^\circ (\pm 0.1^\circ)$  with the deuteron beam, the mean neutron energy then being  $14.10 \pm 0.05$  Mev (see Appendix I).

The target area from which the neutrons originated was determined by substituting a zinc sulfide screen for the zirconium-tritium target, the spot struck by the deuteron beam being visible through a window in the target assembly. The diameter of the neutron source projected in the direction of observation was about 3 mm. The neutrons were monitored by counting the alpha-particles with a proportional counter viewing the whole target area; the counter voltage was supplied by dry batteries.

The neutrons were detected by means of a liquid scintillation counter consisting of a cylindrical glass cell 4 cm in diameter and 2.5 cm long, filled with a solution of terphenyl in xylene<sup>8</sup> (5 g/liter) and cemented to an RCA 5819 photomultiplier operated at 1000 volts. The characteristics of counters of this type for the detection of neutrons have been described previously.<sup>9</sup> Pulses from the photomultiplier passed through a cathode follower stage and then through a 60-foot cable to an Atomic Instrument Company Model

204-B amplifier and were counted with a Model 101-M scaler. A Sorenson voltage regulator was used to minimize effects of line fluctuations on amplifier gain and on discriminator bias voltage, i.e., the voltage which an input pulse to the scaler had to exceed in order to trigger the discriminator. The output voltage of the regulated power supply for the photomultiplier was monitored with an electrostatic voltmeter with detectable deviations of about 0.5 volt in 1000 volts. The discriminator bias voltage was also monitored to insure its constancy, to within 0.1 percent.

Measurements of the dependence of counting rate on the above voltages showed that variations in counting rate which could arise from unnoticeable changes in the above voltages would be less than 1 percent. The same conclusion held for the alpha-monitor. Its counting rate as a function of its amplifier discriminator setting was determined with the scintillation counter, in this case, serving as monitor.

The advantage of the detector for this experiment is its high efficiency for fast neutrons. It is not directional, however, so that it responds to neutrons scattered into it from the surroundings. Such neutrons are of lower energy than those in the direct beam and can be discriminated against to some extent by biasing, and, furthermore, this background is constant for a fixed arrangement and can be determined accurately. It was evaluated by placing midway between source and detector a 33-cm tapered copper cylinder having a calculated transmission of 0.04 percent for 14-Mev neutrons. At our operating bias and with a 100-cm source-to-detector-distance, the ratio of scattered to direct neutrons was about 5.5 percent. This ratio was also measured at a 75-cm source-to-detector-distance, the 100-cm ratio being greater than the 75-cm ratio by a factor equal, within the statistics of the measurement, to  $(100/75)^2$ , which would be expected if we are dealing with a point source of neutrons plus a background independent of position. This result also shows that no gamma-rays from the copper were affecting the scintillation counter.

The detector efficiency was about 5 percent as determined from the alpha-monitor counting rate, an efficiency considerably greater than realized with the detectors used in the previous measurements in this energy range.<sup>1-5</sup> Measurements could thus be made in a relatively short time, reducing the effect of long-time fluctuations. The high efficiency also makes possible a good geometrical arrangement so that corrections for the scattering of neutrons into the detector by the scatterer are small.

## III. MEASUREMENTS

### A. Carbon and Polyethylene

The transmission of six thicknesses of carbon and six thicknesses of polyethylene were measured to determine their respective total cross sections, the transmission of

<sup>7</sup> G. A. Snow, *Phys. Rev.* **87**, 21 (1952).

<sup>8</sup> Reynolds, Harrison, and Salvini, *Phys. Rev.* **78**, 488 (1950); H. Kallmann and M. Furst, *Phys. Rev.* **79**, 857 (1950).

<sup>9</sup> Falk, Poss, and Yuan, *Phys. Rev.* **83**, 176 (1951).

the scatterers ranging from 40 to 85 percent. The scatterers were in the shape of accurately machined truncated cones having median diameters of 2.54 cm and tapers of  $1.5^\circ$  to match the divergence of the neutron beam. In length, they ranged from 1.6 cm to 8.9 cm.

The carbon was in the form of grade AGHT graphite specified to be 99.96 percent pure. To determine if it had an appreciable water content, a sample was heated in vacuum to  $115^\circ\text{C}$  for 15 hours. Its loss of weight amounted to less than 0.015 percent. The polyethylene was chemically analyzed with a macro method, employing 1 to 1.5 g samples, by R. Paulson of the National Bureau of Standards. The hydrogen content of the sample was found to be  $14.365 \pm 0.010$  percent, which checks the theoretical (corresponding to  $\text{CH}_2$ ) hydrogen content of 14.372 percent. The carbon content agreed with the theoretical value to better than 0.1 percent, and the absence of traces of impurities was established with the same precision. This result was confirmed by an infrared spectroscopic analysis carried out by Mr. R. G. Fulton of the Bakelite Company. This analysis determined the number of terminal methylene groups (one extra hydrogen) to be about 1 per 1500 carbons and the number of internal unsaturated bonds (2 fewer hydrogens) to be about 1 per 4000 carbons. Furthermore, no trace of any impurities was found.

The area of the neutron source was mentioned in the previous section. The scatterers were supported by a thin aluminum tray mounted on a length of 4.3-mm diameter aluminum tubing of 0.8-mm wall thickness. The presence of the support did not produce any detectable effect on the measurements. Scatterers and detector were aligned with the aid of a cathetometer. In the alignment procedure, a hollow dummy scatterer of the same median diameter and taper as the regular scatterers was used. It was fitted with a set of cross-hairs at each end so that the scatterer support could be adjusted in angle as well as in height. After this was done, the detector was placed in position. The center of the face of the glass cell containing the terphenyl-xylene solution was aligned with the aid of a locating pin and markings on the detector assembly which could be viewed through the cathetometer telescope. The centers of the source, scatterers, and detector deviated by less than 0.5 mm from the same straight line. The detector diameter of 4 cm was thus well within the shadow cone of the scatterer, which was 5 cm in diameter at the detector position.

The 33-cm copper cylinder used to determine the background was fitted with locating pins so that it, too, could be accurately aligned. Of necessity, it was supported by a heavier piece of tubing than used for the scatterers. However, the long length of copper itself shielded the holder from both source and detector so that it could not scatter neutrons into the detector, and in actual fact, no effect of the heavy holder on counting rate could be detected.

A fixed number of neutron counts and the corre-

TABLE I. Observed and corrected transmissions for different carbon and polyethylene scatterers (thicknesses in units of number of carbon atoms/cm<sup>2</sup>). Errors of transmissions are stated in text.

Substance	$n_C x / 10^{24}$	Observed transmission	Corrected transmission
Carbon	0.1332	0.8409	0.8406
	0.1984	0.7753	0.7749
	0.2927	0.6909	0.6903
	0.3726	0.6190	0.6184
	0.4806	0.5396	0.5389
	0.7545	0.3820	0.3812
Polyethylene	0.06280	0.8520	0.8517
	0.09271	0.7871	0.7866
	0.1376	0.6945	0.6939
	0.1746	0.6354	0.6347
	0.2247	0.5528	0.5520
	0.3534	0.3929	0.3920

sponding number of alpha-monitor counts were recorded for each scatterer. Blanks, i.e., measurements with no scatterer in the beam, were made before and after each measurement with a scatterer, the relative transmission for each scatterer being determined by using the mean of neighboring blanks, to minimize slow fluctuations of several percent in the ratio of neutron to alpha-monitor counts, which were sometimes observed under fixed conditions. The exact cause of these fluctuations is not known, but they may have been the result of slight physical changes in the target, such as the deposition of a carbon layer by the action of the beam.

Nine independent runs were made on each polyethylene scatterer and six on each carbon scatterer. About 20,000 neutron counts were accumulated in each run, the number of monitor counts being greater by factors of about 2 to 5, depending on the scatterer. The probable error of any one value of transmission based on the number of detector and monitor counts entering into its calculation is in the range of 0.2 percent to 0.3 percent. The probable error as determined from the deviations of the independent runs from the average for any one value of transmission is in the range of 0.2 percent to 0.4 percent. Background measurements with the copper absorber were interspersed throughout each run. A day-to-day correction for the cosmic-ray background of the detector was made, never amounting to more than 0.3 percent.

The relative transmission,  $T$ , as a function of absorber thickness is given ideally by the relation

$$T = e^{-\sigma n x},$$

where  $\sigma$  is the total cross section and  $n x$  is the absorber thickness in atoms/cm<sup>2</sup>. Because of the finite dimensions of the detector and scatterers, neutrons can be scattered into the detector from the scatterer, resulting in a transmission higher than that given by the above expression. Correction for this effect, shown in Appendix II, is largest for the thickest scatterer for which, in our geometrical arrangement, it amounted to only 0.2

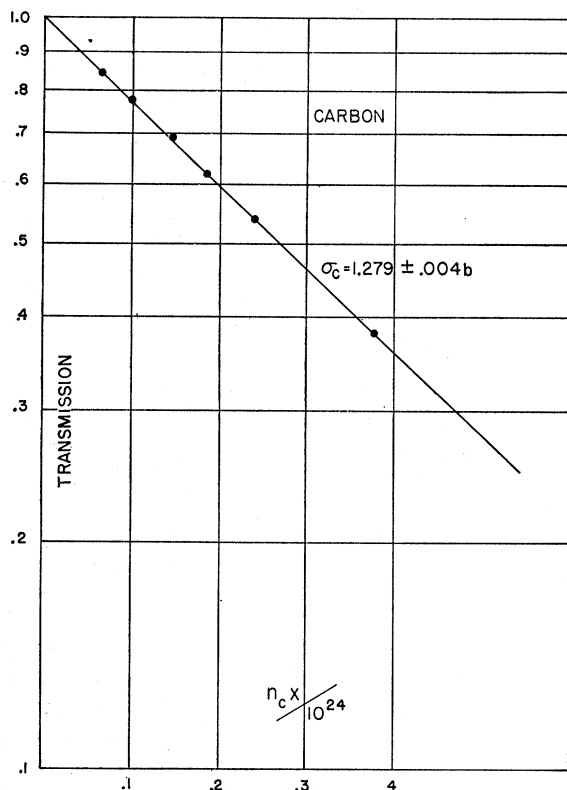
TABLE II. Summary of carbon and hydrogen cross section measurements in the 14-Mev neutron energy range.

Observers	Neutron energy Mev	Neutron cross section in barns	
		C	H
Salant and Ramsey <sup>a</sup>	14	1.27±0.04	0.70±0.06
	15	1.36±0.05	0.66±0.07
Agno <i>et al.</i> <sup>b</sup>	14	1.23±0.015	0.694±0.019 <sup>c</sup>
Sleator <sup>c,d</sup>	14	1.30	0.69
Lasday <sup>e</sup>	13.9	1.24±0.06	0.77±0.04
Meyer and Nyer <sup>f</sup>	14.2	1.29±0.02	0.675±0.02
Present paper	14.10±0.05	1.279±0.004	0.689±0.005

<sup>a</sup> See reference 2.<sup>b</sup> See reference 3.<sup>c</sup> See reference 4.<sup>d</sup> Sleator made measurements at mean neutron energies ranging from 6.5 Mev to 21.1 Mev. The values at 14 Mev are taken from his plots of cross section vs energy which also include data obtained by others in previous measurements.<sup>e</sup> See reference 5.<sup>f</sup> See reference 6.<sup>g</sup> If we use our value of  $\sigma_C$ , the value of  $\sigma_H$  from the data of Agno *et al.* is  $0.673 \pm 0.020$  b.

percent. Transmissions of cylinders of greater width than the truncated cones were also measured. With diameters ranging from 3.8 to 5.1 cm, there was only random departure, within the 1 percent accuracy of these measurements, from the narrow-cone results, consistent with the calculations of the Appendix.

Table I gives the observed and corrected transmission for polyethylene and carbon. In Figs. 2 and 3 the logarithms of the transmissions are plotted against absorber thickness in atoms of carbon/cm<sup>2</sup> for both

FIG. 2. Carbon transmission vs scatterer thickness in units of number of carbon atoms/cm<sup>2</sup>.

substances. (In addition, transmissions were measured with a 75-cm source-to-detector-distance; these differed from the measurements at 100 cm distance by less than the probable error of about 0.5 percent.) For a carbon or polyethylene scatterer of length  $x$ , the number of carbon atoms/cm<sup>2</sup> is

$$(N/A)(M/V)x,$$

where  $N$  is Avogadro's number,  $A$  the atomic weight of carbon or the molecular weight of polyethylene,  $M$  the mass, and  $V$  the volume of the scatterer.  $V$  was computed from the measured dimensions to within 0.15 percent, and as a check by weighing some of the scatterers in air and in water. The two methods agreed to less than 0.3 percent when the weights were reduced to weights in vacuum. It should be pointed out that since the length appears as a factor in the volume, it, together with any error in its measurement, cancels out of the above expression for the number of atoms/cm<sup>2</sup> when the volume is calculated from the dimensions of the scatterer.

The slopes of the straight lines through the sets of points in Figs. 2 and 3 are  $-\sigma_C$  and  $-(\sigma_C + 2\sigma_H)$  for the carbon and polyethylene, respectively. The lines and the probable errors in their slopes were determined by the method of least squares,<sup>10</sup> in which errors were attributed solely to the transmission values. This procedure was justified, since these errors were considered to outweigh the errors in  $nx$ .

The results for the total cross sections at  $14.10 \pm 0.05$  Mev are

$$\sigma_C = 1.279 \pm 0.004 \text{ b}, \quad \sigma_C + 2\sigma_H = 2.657 \pm 0.009 \text{ b},$$

from which

$$\sigma_H = 0.689 \pm 0.005 \text{ b}.$$

For comparison, results in this energy range are presented in Table II.

## B. Deuterium

The total cross section of deuterium was determined by measuring the transmission of a hollow aluminum cell alternately filled with light and heavy water. The cell was 7.3 cm long, 3.2 cm in diameter, had end faces of 0.8-mm thickness, and gave a transmission of 48 percent when filled with light water. Two series of measurements were made on separate occasions, one with a source-to-detector-distance of 100 cm and the other with a 75-cm distance. They differed by less than the probable error associated with each. If we denote by  $R$  the ratio of the transmissions of heavy water to light water, we have

$$R = \exp[-b2n_O(\sigma_D - \sigma_H)x],$$

where  $n_O$  is the number of oxygen atoms/cm<sup>3</sup>, assumed to be the same for light and heavy water (an assumption valid to better than 0.1 percent) and  $b$  is the concen-

<sup>10</sup> See, for example, W. N. Bond, *Probability and Random Errors* (Edward Arnold and Company, London, 1935).

tration of heavy water in our heavy water sample;  $(\sigma_D - \sigma_H)$  is the difference in total cross sections for deuterium and hydrogen and  $x$  is the length of the sample. For our particular cell,  $n_O x = 0.2390 \times 10^{24}/\text{cm}^2$  and our measured value of  $R$  is  $0.950 \pm 0.004$ , the probable error in  $R$  being determined from the spread of the individual values about the average. The uncertainty in  $R$  produces an uncertainty of almost 10 percent in  $(\sigma_D - \sigma_H)$ . The concentration of heavy water in our sample was determined from density measurements. It decreased from 96.4 percent before the transmissions were measured to 91.2 percent afterwards. This dilution might have resulted from an exchange between the sample and the water vapor in the air while it was being poured back and forth, or during the measurements, as the cell was not tightly enclosed, or from an incomplete flushing of the cell with heavy water prior to filling. The uncertainty in density, however, affects  $(\sigma_D - \sigma_H)$  by an amount small compared to that caused by the probable error in  $R$ .  $(\sigma_D - \sigma_H)$  was computed using the mean of the above mentioned extreme concentrations and adding to its uncertainty the  $\pm 2.6$  percent arising from the extreme values. Our result is

$$\sigma_D - \sigma_H = 0.114 \pm 0.013 \text{ b.}$$

Taking the value of  $\sigma_H$  from the previous section, we obtain for the total cross sections of deuterium for  $14.10 \pm 0.05$  Mev neutrons

$$\sigma_D = 0.803 \pm 0.014 \text{ b.}$$

Ageno *et al.*<sup>3</sup> measured the deuterium cross section in this energy range by determining the transmission of a heavy paraffin sample. Their value is  $0.864 \pm 0.028$  b; if we use our value of  $\sigma_C$ , their value becomes  $0.841 \pm 0.028$  b. The result of Meyer and Nyer<sup>6</sup> is  $0.81 \pm 0.03$  b.

### C. Oxygen and Nitrogen

The total cross sections of oxygen and nitrogen were determined with a lower accuracy than was attained for carbon and hydrogen, as fewer thicknesses of scatterers were used and fewer counts were accumulated.

The oxygen cross section was obtained from the transmissions of three hollow aluminum cells filled with water. One of the cells has been described in connection with the deuterium measurements. The other two were similar but shorter. No scattering-in correction was applied to these data as it is small compared to the uncertainty in the transmissions. In Fig. 4, the logarithms of the transmissions are plotted against absorber thickness in number of oxygen atoms/cm<sup>2</sup>. In measuring the relative transmissions, it was more convenient to make blank measurements with no cell rather than using an empty cell. A correction was therefore made for the slight attenuation (2 percent) caused by the aluminum-end faces, using for the aluminum cross section the value obtained by Ageno *et al.*<sup>3</sup> Use could then be made of the origin in fitting a line through the points.

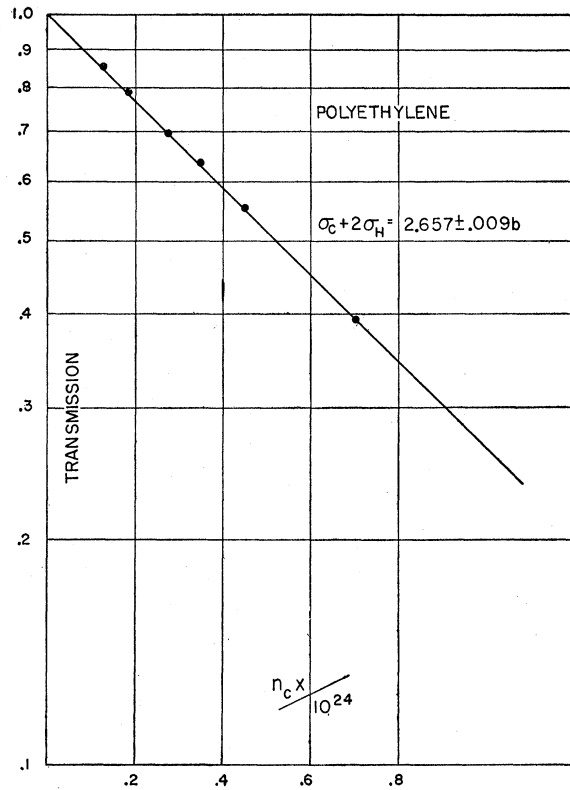


FIG. 3. Polyethylene transmission vs scatterer thickness in units of number of carbon atoms/cm<sup>2</sup>.

The slope of the best fitting straight line on a least squares basis gives directly

$$\sigma_O + 2\sigma_H = 3.02 \pm 0.04 \text{ b.}$$

Using our value of  $\sigma_H$  we find for the total cross section of oxygen at  $14.10 \pm 0.05$  Mev

$$\sigma_O = 1.64 \pm 0.04 \text{ b.}$$

Meyer and Nyer<sup>6</sup> report values of  $1.62 \pm 0.04$  b and  $1.60 \pm 0.04$  b for this cross section from measurements

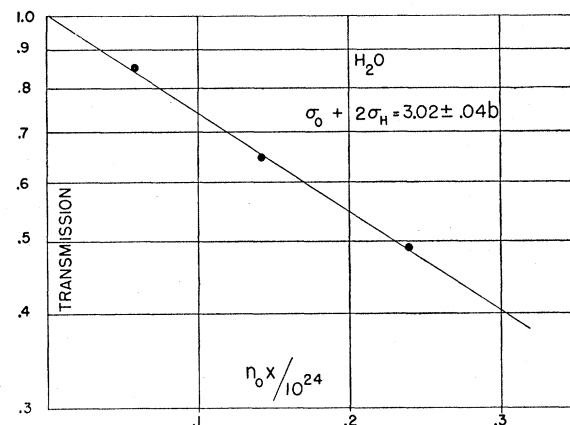


FIG. 4. Transmission of water vs thickness in units of number of oxygen atoms/cm<sup>2</sup>.

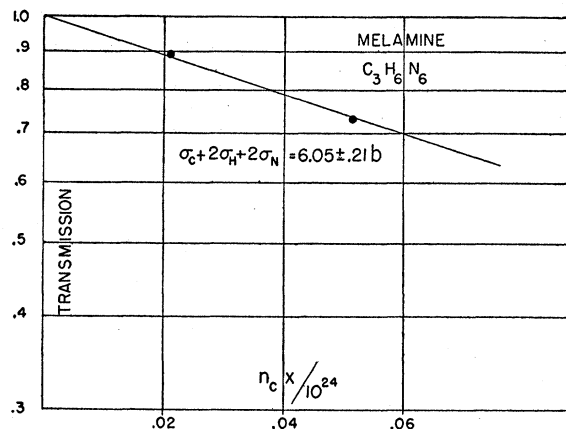


FIG. 5. Transmission of melamine vs thickness in units of number carbon atoms/cm<sup>2</sup>.

on water and formic acid, respectively. If we use our values of  $\sigma_H$  and  $\sigma_{poly}$  in their data, both of their values become  $1.59 \pm 0.04$  b.

The total cross section of nitrogen was determined from measurements on the compound melamine,  $C_3H_6N_6$ . The melamine was purified by Dr. A. Bothner-By of this laboratory. It was heated before use to remove possible traces of water. Spectroscopic examination for metallic impurities, by M. Slavin of BNL, showed none. R. C. Anderson and Y. Delabarre of BNL analyzed the sample for carbon and hydrogen by means of a manometric method<sup>11</sup> for determining  $CO_2$  and  $H_2O$  found in combustion. The hydrogen to carbon ratio was 1.98, differing from the expected value of 2 by an amount comparable to the error of the determination. The amounts of carbon and hydrogen added up to the value expected for a pure compound. The percentage of nitrogen was determined, by J. A. Fancher and L. M. Pollack of BNL, by the Kjeldahl method, which yielded a value of 65.2 percent as compared with the expected 66.6 percent. The nature of the method is such as to give an underestimate, and taking into account the above mentioned checks, it was considered that errors resulting from assuming the sample to be pure melamine were less than those associated with the transmission measurements.

The transmissions of the two shorter aluminum cells were measured when they were filled with melamine in the form of a powder. The effective density of the melamine was obtained from the dimensions of the cells and the weights of melamine they contained and differed by 0.4 percent in the two cells. Transmissions were determined with a 75-cm source-to-detector distance and with a detector of terphenyl crystal 4 cm long and 4 cm in diameter. Because of its extra length, it had a somewhat higher efficiency for neutrons than did the cell used in our other measurements, but its

<sup>11</sup> Anderson, Delabarre, and Bothner-By. Submitted to Anal. Chem.; a modification of the method of J. J. Naughton and M. M. Frodyma, Anal. Chem. **22**, 711 (1950).

integral bias curve was steeper than that of the more transparent liquid.

Figure 5 shows the logarithms of the two transmissions plotted against absorber thickness in number of nitrogen atoms/cm<sup>2</sup>. Again, no correction was made for scattering into the detector. The attenuation of the aluminum end faces was taken into account as with the oxygen determination. Since the carbon and hydrogen in melamine are in the same ratio as in polyethylene, our result for the slope of the best fitting line can be expressed as

$$\sigma_{poly} + 2\sigma_N = 6.05 \pm 0.20 \text{ b.}$$

Using our value of  $\sigma_{poly}$  we find, for the total cross section of nitrogen at  $14.10 \pm 0.05$  Mev,

$$\sigma_N = 1.7 \pm 0.1 \text{ b.}$$

Meyer and Nyer<sup>6</sup> obtained the value  $1.39 \pm 0.05$  b from a measurement with liquid nitrogen. The disagreement might be the result in part of uncertainty in the determination of the density of liquid nitrogen.

We are very happy to acknowledge our indebtedness to Dr. M. A. Tuve for putting the facilities of the Department of Terrestrial Magnetism at our disposal and to Dr. N. P. Heydenburg and Mr. C. Little of that department for their willing and invaluable help with the electrostatic generator. We take pleasure, too, in thanking the Los Alamos Scientific Laboratory for the use of the tritium source, Mr. R. G. Fulton of the Bakelite Company for the infrared analysis, Mr. R. Paulson of the National Bureau of Standards for the very accurate chemical analysis of our polyethylene sample, and our colleagues of the Brookhaven National Laboratory Chemistry and Medical Departments, Dr. R. C. Anderson, Dr. A. A. Bothner-By, Mr. M. Slavin, Miss Y. J. Delabarre, Dr. J. A. Fancher, and Mrs. L. M. Pollack for purification and analyses of the melamine. One of us (G.A.S.) is thankful to Dr. H. S. Snyder and Dr. D. A. Kleinman of Brookhaven National Laboratory for some helpful discussions.

#### APPENDIX 1

##### The Energy Spectrum of the Neutrons

The large  $Q$  of the reaction  $T(d,n)He^4$  makes this reaction an excellent source of nearly monochromatic neutrons of 14-Mev energy. (See the discussion of Hanson, Taschek, and Williams.<sup>12</sup>) We wish to determine the energy spectrum of the neutrons emerging at an angle of  $89.27^\circ \pm 0.1^\circ$  with respect to a 400-kev deuteron beam impinging on a thick zirconium target loaded with tritium. The factors which enter this determination are (1) the kinematics of the  $T(d,n)He^4$  reaction, (2) the cross section of the reaction as a function of energy and angle, and (3) the multiple scattering angular distribution of the deuterons and their rate of energy loss in the zirconium target.

<sup>12</sup> Hanson, Taschek, and Williams, Revs. Modern Phys. **21**, 635 (1949).

(1) *The kinematics of the T(d,n)He<sup>4</sup> reaction.*—Let  $E_d$  = deuteron kinetic energy in the laboratory system,  $E_n$  = neutron kinetic energy in the laboratory system,  $M_i$  = mass of particle  $i$ , and  $\theta$  = angle between the neutron and the deuteron. Nonrelativistically we then have

$$E_n = \frac{M_\alpha}{M_n + M_\alpha} Q + \left[ \frac{2M_n M_d}{(M_\alpha + M_n)^2} \cos^2 \theta + \frac{M_\alpha - M_d}{M_\alpha + M_n} \right] E_d + \frac{2(M_n M_d E_d)^{\frac{1}{2}}}{M_\alpha + M_n} \cos \theta \left\{ \left[ \frac{M_n M_d}{(M_\alpha + M_n)^2} \cos^2 \theta + \frac{M_\alpha - M_d}{M_\alpha + M_n} \right] E_d + \frac{M_\alpha}{M_n + M_\alpha} Q \right\}^{\frac{1}{2}}. \quad (\text{A1})$$

From the table of masses of light nuclei from nuclear disintegration energies given by Li *et al.*,<sup>13</sup> we obtain

$$Q = M_{\text{H}^3} + M_{\text{H}^2} - M_n - M_{\text{He}^4} = 17.58 \pm 0.02 \text{ Mev}. \quad (2)$$

In Fig. 6,  $E_n$ , given by Eq. (1), is plotted as a function of  $\theta$  for values of  $E_d$  ranging from 50 keV to 400 keV (Fig. 6 is plotted using an earlier value for  $Q$ , i.e.  $Q = 17.56$  Mev). The neutron relativistic kinetic energy is simply obtained by a perturbation calculation on Eq. (1). At  $\theta = 90^\circ$ , one obtains

$$E_n^{\text{(rel)}} - E_n^{\text{(nonrel)}} = -0.00142 E_n^{\text{(nonrel)}} \simeq -0.02 \text{ Mev}.$$

It turns out that the relativistic kinetic energy of the neutrons for  $Q = 17.58$  Mev is equal to the nonrelativistic kinetic range for  $Q = 17.56$  Mev. The two corrections cancel each other out.

It is evident from Fig. 6 that there is an extremely small circle of confusion in neutron energy at  $\theta \simeq 98^\circ$ , i.e.  $E_n = 13.98 \pm 0.015$  Mev. This can be fully utilized only if a thin target is used. With a thick target, the

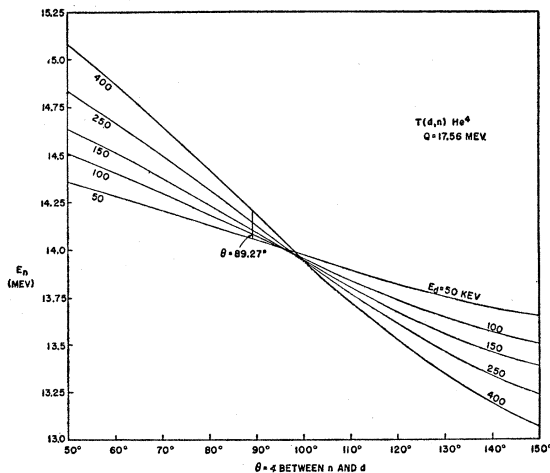


FIG. 6. Neutron energy vs angle between neutron and deuteron for different deuteron energies,  $E_d$ , for the reaction T(d,n)He<sup>4</sup>.

<sup>13</sup> Li, Whaling, Fowler, and Lauritsen, Phys. Rev. **83**, 512 (1951).

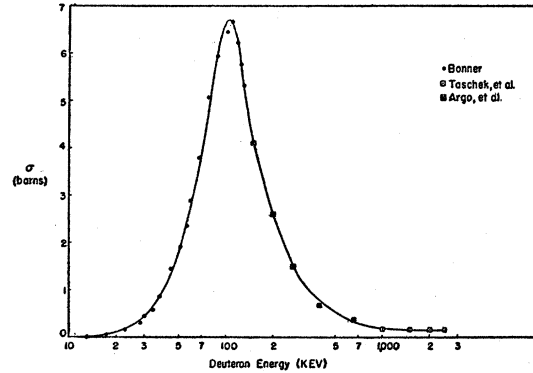


FIG. 7. Total cross section of the T(d,n)He<sup>4</sup> reaction vs deuteron energy.

excellent energy resolution is smeared out somewhat by the multiple scattering of the deuterons in the target. This scattering smears out the distribution of angles between the direction of motion of the deuteron just as it is captured in the target and the fixed direction of observation of the neutron that is emitted.

(2) *The cross section of T(d,n)He<sup>4</sup> as a function of energy and angle.*—Bonner<sup>14</sup> has measured the cross section as a function of deuteron energy from 15 keV to 133 keV with very thin targets by detecting the alpha-particles at an angle of  $90^\circ$  with respect to the incident deuterons. He finds  $\sigma$  to be a rapidly increasing function of deuteron energy up to a maximum of 6.65 barns at 110 keV. The rapid rise is the result primarily of the rise in the Coulomb barrier probability. For this same range of deuteron energies, Allan and Poole<sup>15</sup> have shown that the angular distribution of the alpha-particles (and hence of the neutrons) is symmetric in the center-of-mass system. Taschek *et al.*<sup>16</sup> have measured the cross section as a function of angle for 1-Mev to 2.5-Mev deuterons impinging on gaseous tritium. They find the total cross section to be about 0.15 barn, and a distinct asymmetry in the angular distribution in the center-of-mass system that essentially increases with energy. Recently, Argo and co-workers<sup>17</sup> have measured the total cross section in the region from 53 keV to 1000 keV, assigning an error of  $\pm 10$  percent in the region from 125-keV to 800-keV-deuteron energy. They also measured the angular distribution of the cross section at various energies. Within a rather large experimental error, they conclude that the angular distribution is isotropic in the center-of-mass system up to a deuteron energy of 267 keV and becomes progressively less isotropic in the center-of-mass system until, at an energy of 800 keV, the yield is isotropic in the laboratory system. In Fig. 7 the total cross section for the T(d,n)-

<sup>14</sup> T. W. Bonner, AEC Unclassified Report No. 939 (1950).

<sup>15</sup> D. L. Allan and M. J. Poole, Proc. Roy. Soc. (London) **A204**, 500 (1951).

<sup>16</sup> Taschek, Hemmendinger, and Jarvis, Phys. Rev. **75**, 1464 (1949), and Atomic Energy Commission Declassified Report No. 2250 (1948).

<sup>17</sup> H. V. Argo, private communication.

He<sup>4</sup> reaction is plotted as a function of incident deuteron energy. The results of Argo<sup>17</sup> and Taschek<sup>16</sup> in the energy region  $E_d \geq 150$  kev have been smoothly joined to Bonner's results for  $E_d \leq 133$  kev. It is clear from this figure that deuteron energies much less than 50 kev do not make any significant contribution to the total cross section.

The differential cross section  $\sigma(\theta)$  in the laboratory system is obtained for a given deuteron energy by a simple solid angle transformation from the center-of-mass coordinate system. The differential cross section  $\bar{\sigma}$  in the center-of-mass system is experimentally constant up to 267 kev<sup>17</sup> and is assumed constant up to 400 kev. The ratio  $\sigma(\theta)/\bar{\sigma}$  was obtained as a function of  $\theta$  for several different deuteron energies.

(3) *The rate of energy loss and the multiple angular distribution of the deuterons in the Zr target.*—Warshaw<sup>18</sup> has measured the stopping power of low energy protons in various materials including silver ( $Z=47$ ) and copper ( $Z=29$ ). Since the experimental stopping power curves for these two elements are close together, a very good interpolation can be used for zirconium ( $Z=40$ ). The stopping power  $S$  in units of kev/atom/cm<sup>2</sup> is obtained as a function of  $E_d$  using the relationship  $S_{\text{deuteron}}(E) = S_{\text{proton}}(E/2)$ . An extrapolation of the experimental points was necessary below  $E_d = 100$  kev. The relative number of neutrons that arise from the capture of deuterons in a differential energy range  $\Delta E_d$  of different energies  $E_d$  is proportional to the cross section  $\sigma(E_d)$  times the length of path traversed by the deuteron in losing energy  $\Delta E_d$ . This length is proportional to  $\Delta E_d/S(E_d)$  so that the relative number of neutrons from energies between  $E_d$  and  $E_d + \Delta E_d$  is proportional to  $\sigma(E_d)/S(E_d)$ . The function  $\sigma(E_d)/S(E_d)$  compared to  $\sigma(E_d)$  is relatively larger on the low energy side of the peak and smaller on the high energy side.  $S(E_d)$  is a slowly varying function of deuteron energy in the region under consideration.

To evaluate the multiple scattering angular distribution of the deuterons in the target, we use the projected angle distribution functions calculated by Snyder and Scott<sup>19</sup> in the small angle approximation ( $\sin\theta \simeq \theta$ ,  $\cos\theta \simeq 1$ ). Energy loss will be included by a method essentially the same as that described by

TABLE III. Approximate scattering distribution function parameters for incident deuterons of energy  $E_d = 400$  kev.

$E$	$z$	$\eta_0$	$b$	$\eta_1$
50	32.3	0.031	2.66	0.613
100	25.8	0.020	9.33	0.327
150	21.0	0.013	34.6	0.170
250	12.9	0.011	72.5	0.117

<sup>18</sup> S. D. Warshaw, Phys. Rev. **76**, 1759 (1949).

<sup>19</sup> H. S. Snyder and W. T. Scott, Phys. Rev. **76**, 220 (1949); **78**, 223 (1950).

Molière.<sup>20</sup> For the single scattering cross section we take

$$\sigma(\theta) = \frac{4z^2 Z^2 e^4}{M^2 v^4} \frac{1}{\theta^2 + \eta_0^2}, \quad (\text{A2})$$

where  $M$ ,  $v$ ,  $ze$  are the mass, velocity, and charge of the scattered particle,  $Z$  is the atomic number of the scattering particle, and, from Molière,<sup>20</sup>

$$\eta_0 = \theta_0 (1.13 + 3.76\alpha^2)^{\frac{1}{2}}, \quad (\text{A3})$$

where

$$\alpha = zZe^2/\hbar v, \quad \theta_0 = \hbar/Mva, \quad \text{and } a = 0.466 \times 10^{-8}/Z^{\frac{1}{2}} \text{ cm.}$$

This form for  $\sigma(\theta)$  is valid for kinetic energies of the incident particle greater than  $100 zZ^{4/3} \text{ ev} = 14$  kev.  $W(\eta|z)$ , calculated by Snyder and Scott, is the angular distribution function for the projected angle  $\eta$  between the initial particle direction and the particle direction at the end of the track for various values of  $z$ . ( $z =$  distance traversed by the particle in units of the mean free path,  $\lambda$ , for single scattering.) In order to include energy loss, we transform to the distribution function  $U(\delta|z)$ , where  $\delta = \eta'/\sqrt{z}$ ,  $\eta' = \eta/\eta_0$  and  $U(\delta|z) = z^{\frac{1}{2}} W(\eta'|z)$ .  $U(\delta|z)$  is a slowly varying function of  $\delta$  and  $z$ . For a particle of incident energy  $E_0$  and final energy  $E$ ,  $U(\delta|z)$  is obtained by using the following average values for  $z$  and  $\eta_0$ :

$$z = L(E) \langle (1/\lambda) \rangle_{Av}, \quad \eta_0 = [ \langle \eta_0^2/\lambda \rangle_{Av} / \langle (1/\lambda) \rangle_{Av} ]^{\frac{1}{2}}, \quad (\text{A4})$$

where

$$L(E) = \int_{E_0}^E \frac{dl}{dE} dE = \int_E^{E_0} \frac{dE}{NS(E)}$$

= distance in cm traversed by the particle.

$$\left\langle \frac{1}{\lambda} \right\rangle_{Av} = \int_{E_0}^E \frac{1}{\lambda} \frac{dl}{dE} dE / L(E),$$

$$\left\langle \frac{\eta_0^2}{\lambda} \right\rangle_{Av} = \int_{E_0}^E \frac{\eta_0^2}{\lambda} \frac{dl}{dE} dE / L(E).$$

The pertinent values of  $z$  are less than 100, so that an extrapolation of the tabulated values of the slowly varying function  $U(\delta|z)$  was necessary. For our purpose the final angular distribution function  $W(\eta|z)$  can be approximated by a straight line of the form

$$W(\eta|z) = b^{\frac{1}{2}} - b\eta. \quad (\text{A5})$$

It then follows that

$$\int_0^{\infty} W(\eta|z) d\eta = 0.5. \quad (\text{A6})$$

The values of  $z$ ,  $\eta_0$ ,  $b$  and  $\eta_1$  [the maximum angle, i.e.  $W(\eta_1) = 0$ ] are listed in Table III for an incident

<sup>20</sup> G. Molière, Z. Naturforsch. **3a**, 78 (1948).



deuteron energy  $E_d=400$  kev and final energies  $E=50, 100, 150, 250$  kev.

The straight line approximation to  $W(\eta|z)$  has the effect of cutting off the tail of the distribution and increasing the half-width. From the values of  $\eta_1$  in Table III, we see that  $W(\eta|z)$  is large only for values of  $\eta \ll 30^\circ$ , so that the small angle approximation is justified.

To evaluate the neutron energy distribution, let  $\theta$ =angle between the direction of a deuteron in the target and the fixed direction of the emergent neutron beam;  $\theta_0=89.27^\circ$ =incident deuteron direction relative to the neutron beam, and  $N(E_n)dE_n$ =relative number of neutrons of energy between  $E_n$  and  $E_n+dE_n$ . Then

$$N(E_n)dE_n \approx \int \int_{(E_n \text{ constant})} d\theta dE_d \frac{\sigma(E_d)}{S(E_d)} \times \frac{\sigma(E_d, \theta)}{\bar{\sigma}(E_d)} \times W(\theta - \theta_0 | z(E_d)). \quad (\text{A7})$$

Each deuteron energy  $E_d$  is weighted by the factor  $\sigma(E_d)/S(E_d) \cdot \sigma(E_d, \theta)/\bar{\sigma}(E_d)$  describes the variation of cross section with laboratory angle, where  $\bar{\sigma}(E_d) = \sigma(E_d)/4\pi$ .  $W[\theta - \theta_0 | z(E_d)]$  describes the angular distribution of the deuterons of energy  $E_d$  about their incident direction. Consistent with the small angle approximation, scattering in the direction normal to the plane of the incident deuteron and the emergent neutron is neglected. This scattering has only a second-order effect on the energy of the emergent neutron. It is assumed that there is a uniform distribution of tritium in the target. The integral of Eq. (7) is evaluated numerically after changing the variables of integration from  $d\theta dE_d$  to  $dE_n dE_d$ . For this purpose we note that in the region about  $\theta = \pi/2$ , Eq. (1) can be approximated as

$$E_n = A(E_d) + BE_d^3 \cos \theta, \quad (\text{A8})$$

$$A(E_d) = 14.03 + 0.397E_d, \quad B = 2.131.$$

Using the small angle approximation  $\cos \theta \approx -(\theta - \frac{1}{2}\pi)$ , we obtain

$$N(E_n) \approx \int_0^{E_0} dE_d \frac{1}{2BE_d^3} \frac{\sigma(E_d)}{S(E_d)} \frac{\sigma(E_d, \theta)}{\bar{\sigma}(E_d)} \times W(\theta - \theta_0 | z(E_d)) \quad (\text{A9})$$

( $E_0 = 400$  kev).

The final result for  $N(E_n)$  vs  $E_n$  is plotted in Fig. 8. Approximately 95 percent of the neutrons have energies between 13.90 and 14.25 Mev. From this distribution we obtain the reciprocal of the mean value of  $1/E_n$ :  $\bar{E}_n = [\langle E_n^{-1} \rangle_{Av}]^{-1} = 14.10$  Mev. (Since, in this energy region, the  $n-p$  cross section is approximately proportional to  $E_n^{-1}$ ,  $E_n^{-1}$  must be averaged and its reciprocal taken in order to obtain the approximately correct mean neutron energy  $\bar{E}_n$  to be used for the theoretical cross section calculation. The ordinary mean neutron energy  $\bar{E}_n$  is identical to  $\bar{E}_n$  in this case, i.e.,  $\bar{E}_n = 14.10$  Mev.)

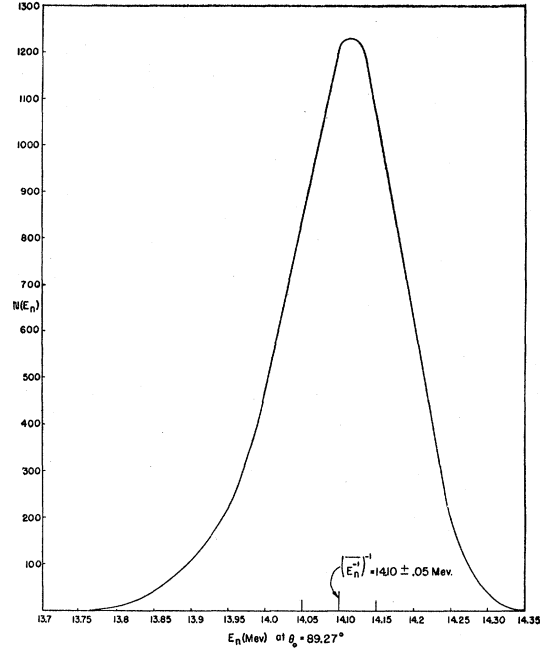


FIG. 8. Number of neutrons per unit energy range (arbitrary scale units) vs neutron energy for angle of observation of  $89.27^\circ$  with respect to incident deuteron beam.

The half-width at half-maximum of  $N(E_n)$  is 0.090 Mev. We assume that  $\bar{E}_n$  obtained from this calculation has an uncertainty of half that much, that is,  $\pm 0.045$  Mev. Besides this uncertainty, the  $Q$  of the  $T(d,n)He^4$  reaction is uncertain to  $\pm 0.02$  Mev. Finally the finite angular resolution of the neutron beam, i.e.,  $\pm 1^\circ$ , contributes a further spread in energy of  $\pm 0.015$  Mev. An approximate estimate of the effect on the mean neutron energy of the neutrons that are scattered, elastically and inelastically, back from the target holder and its enclosing chamber into the detector, shows that this effect shifts the mean neutron energy by an amount very much less than 0.01 Mev. Combining these uncertainties we obtain for the mean neutron energy

$$\bar{E}_n \equiv [\langle (E_n^{-1}) \rangle_{Av}]^{-1} = 14.10 \pm 0.05 \text{ Mev}, \quad (\text{A10})$$

using 17.56 Mev as the  $Q$  value. Adopting the better value  $Q=17.58$  Mev, and at the same time, subtracting the relativistic correction of 0.020 Mev, leaves the value of  $\bar{E}_n$  unchanged. Hence we obtain for the mean relativistic kinetic energy of the neutrons

$$(\bar{E}_n)_{rel} = 14.10 \pm 0.05 \text{ Mev}. \quad (\text{A11})$$

## APPENDIX II

### Scattering-in Correction

The observed transmission  $T$  of neutrons through a scatterer can be written as

$$T = T_0 + T_1 + T_2 + \dots = T_0(1 + T_1/T_0 + T_2/T_0 + \dots),$$

where  $T_0$  is the true transmission, i.e., the number of neutrons reaching the detector without being scattered in passing through the scatterer,  $T_1$  is the number of singly scattered neutrons reaching the detector, and  $T_2$  is the number of doubly scattered ones reaching the detector.

In our geometrical arrangement, we have source diameter, 0.3 cm; scatterer diameter, 2.5 cm; maximum scatterer length, 8.9 cm; source to scatterer and scatterer to detector distance, 50 cm. Under these conditions, the dimensions of the scatterers are sufficiently small compared to their distance from the source and detector so that the cosines of the scattering angles involved may be taken as unity. Following a procedure similar to that of Amaldi *et al.*<sup>21</sup> we then obtain

$$T_1/T_0 = (4A/d^2)n\sigma(0)L,$$

where  $A$  is the area of the scatterer,  $d$  is one-half the source-detector distance,  $n$  is the number of scattering nuclei/cm<sup>3</sup>,  $\sigma(0)$  is the differential cross section evaluated at zero degrees, and  $L$  is the length of the scatterer.

In the case of hydrogen,  $\sigma(0)$  was determined by considering the scattering to be isotropic in the center-of-mass system. This gives

$$\sigma_H(0) = \sigma_H/\pi,$$

where  $\sigma_H$  is the total cross section. In the case of a larger nucleus like carbon, small angle scattering has been treated as a diffraction effect by Bethe and Placzek,<sup>22</sup> who obtain for the differential cross section at small angles

$$\sigma(\theta) = R^2 \left[ \frac{J_1(R\theta/\lambda)}{\theta} \right]^2,$$

where  $R$  is the nuclear radius,  $\lambda$  is the neutron wave number over  $2\pi$ , and  $J_1$  is a Bessel function of the first kind. For a 14-Mev neutron,  $\lambda = 1.21 \times 10^{-13}$  cm.  $R$  is  $3.8 \times 10^{-13}$  cm for carbon.<sup>23</sup>  $\theta \leq 0.09$  for our arrangement. With these values  $J_1(R\theta/\lambda) \approx \frac{1}{2}R\theta/\lambda$ .

Putting in numerical values, we have for carbon

$$(T_1/T_0)_{\text{carbon}} = 2.36 \times 10^{-4}L,$$

<sup>21</sup> Amaldi, Bocciarelli, Cacciapuoti, and Trabacchi, *Nuovo cimento* **3**, 203 (1946).

<sup>22</sup> H. A. Bethe and G. Placzek, *Phys. Rev.* **57**, 1075 (1940).

<sup>23</sup> H. Feshbach and V. Weisskopf, *Phys. Rev.* **76**, 1550 (1949).

and for polyethylene

$$(T_1/T_0)_{\text{poly}} = 2.45 \times 10^{-4}L,$$

$L$  being the length of the scatterer in cm.

As we show below, the correction for double scattering is small enough with our geometrical conditions so that it can be neglected. The corrected transmissions in Table I were obtained from the observed transmissions by applying the above single scattering corrections which ranged from 0.04 percent for the shortest scatterer to 0.2 percent for the longest one.

A simple consideration of double scattering for our geometrical conditions leads to the following overestimate:

$$T_2/T_0 \leq \{4[n\sigma(0)]^2/d^2\}I,$$

where

$$I = \int \int_V dv_1 dv_2 / r_{12}^2.$$

The integral is over the scatterer volume, and  $r_{12}$  is the distance between two volume elements  $dv_1$  and  $dv_2$ .  $I$  has been approximately evaluated for a scatterer of square cross section having  $b/L \ll 1$ , where  $b$  is its width and  $L$  its length. This condition will hold sufficiently well for the longest scatterers for which the correction is largest. Making use of the identity

$$I = - \int \int_S \ln r_{12} d\mathbf{S}_1 d\mathbf{S}_2,$$

where the integral on the right side is over the rectangular surfaces of the scatterer, one obtains

$$I \simeq b^3 L [4 \tan^{-1}(L/b) - (2b/L) \ln(L/b) - c],$$

where  $c \simeq 1$ .

Using the previously given expression for  $T_1/T_0$ , we then have

$$T_2/T_1 \leq n\sigma(0)b [4 \tan^{-1}(L/b) - (2b/L) \ln(L/b) - c].$$

Putting in numerical values for the longest scatterer, we obtain

$$(T_2/T_1)_{\text{max}} \leq 0.21$$

or

$$(T_2/T_0)_{\text{max}} \leq 0.04 \text{ percent.}$$

Since the probable error associated with each of the transmission values is of the order of 0.3 percent, the double scattering correction was neglected.