In view of the discussion above, the error quoted for the cross section is twice the standard deviation obtained for the six measurements, namely 13.8±2.2 millibarns.

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Angular Distributions and Yields of Neutrons from (*p*,*n*) Reactions

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Angular distributions of neutrons from (*p*,*n*) reactions on Mg, Al, Cu, Mo, Ag, Ta, Au, Th, and U were measured for 23-Mev protons incident on thick targets. All angular distributions show peaks in the forward direction; except for Mg and Al, the 0°/180° intensity ratios vary from 5 to 15 for ~13-Mev neutrons, and from 1.3 to 5 for ~8-Mev neutrons. The Mg and Al high-energy data show minima and secondary maxima. Yield determinations indicate that the number of neutrons per nuclear reaction increases with atomic number from about 0.25 for Mg and Al to about 2.0 for most heavy elements and more than 4 for Th and U (probably due to fission). Temperatures of the neutron energy distributions are estimated; for the heavy elements at the backward angles, they are about 0.85 Mev at 3-Mev neutron energy, and about 1.9 Mev at 10-Mev neutron energy. It is concluded that direct interactions are of considerable importance in these reactions.

It has generally been assumed that nuclear reactions induced by neutrons and protons in the 10- to 25-Mev energy region proceed by a compound nucleus interaction.[1,2] This assumption was originally based on theoretical predictions of very short mean free paths in nuclear matter for nucleons of this energy, but experimental supporting evidence was soon forthcoming. Measurements of energy spectra of emitted neutrons[3] and protons[4] showed them to be approximately Maxwellian. Excitation functions for (*n*,2*n*) and (α,2*n*) reactions confirmed this.[5] Excitation functions for proton-induced reactions on Cu^{63} and alpha-induced reactions on Ni^{60} were found[6] to be remarkably similar when plotted against the excitation energy of the compound nucleus, which is the same in the two cases. All of these facts were considered to be more or less complete confirmation of the compound nucleus hypothesis.

Recently, however, accumulated evidence indicates that mean free paths in nuclear matter are considerably longer than had previously been suspected.[7] It has been pointed out that the deviations from Maxwellian shape in the energy distributions of emitted particles are in the opposite direction from what is demanded by even very general considerations from compound nucleus theory.[8] It was found that (*n*,*p*) reaction cross sections in heavy elements are considerably larger than expected from compound nucleus interactions[9]; this was tentatively explained[10] by assuming that a small fraction of the reactions (~10 percent) take place by a direct interaction. Very convincing evidence for direct interactions was obtained by Eisberg and Igo[11] in their measurements of angular distributions and energy distributions from inelastic scattering of 32-Mev protons by various heavy elements.

In this paper these matters are further studied by measurement of angular distributions of neutrons from (*p*,*n*) reactions induced by ~20-Mev protons. In the process, data on neutron yields were automatically obtained, so that they are also presented.

EXPERIMENTAL

Angular distributions of neutrons from (*p*,*n*) reactions were measured by means of the internal, circulating, 23-Mev proton beam of the ORNL 86-inch cyclotron. The most feasible method of detecting neutrons in an experiment of this type is by activation detectors (sometimes called "threshold detectors"). This method has frequently been criticized as being potentially subject to large errors, but a rather extensive study indicated that, in these particular experiments at

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[1] N. Bohr, Nature **137**, 344 (1936).

[2] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

[3] P. C. Gugelot, Phys. Rev. **81**, 51 (1951); P. H. Stetson and C. Goodman, Phys. Rev. **82**, 69 (1951); B. G. Whitmore and G. E. Dennis, Phys. Rev. **84**, 296 (1951); E. R. Graves and L. Rosen, Phys. Rev. **89**, 343 (1953).

[4] P. C. Gugelot, Phys. Rev. **93**, 425 (1954).

[5] Brolley, Fowler, and Schlacks, Phys. Rev. **88**, 618 (1953); H. C. Martin and B. C. Diven, Phys. Rev. **86**, 565 (1952); D. J. Tendam and H. L. Bradt, Phys. Rev. **72**, 1118 (1947); Bleuler, Stebbins, and Tendam, Phys. Rev. **90**, 460 (1953).

[6] S. N. Ghoshal, Phys. Rev. **80**, 939 (1950).

[7] Feshbach, Porter, and Weisskopf, Phys. Rev. **90**, 166 (1953).

[8] B. L. Cohen, Phys. Rev. **92**, 1245 (1953).

[9] E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).

[10] H. McManus and W. T. Sharp, Phys. Rev. **87**, 188 (1952).

[11] R. M. Eisberg and G. Igo, Phys. Rev. **93**, 1039 (1954).

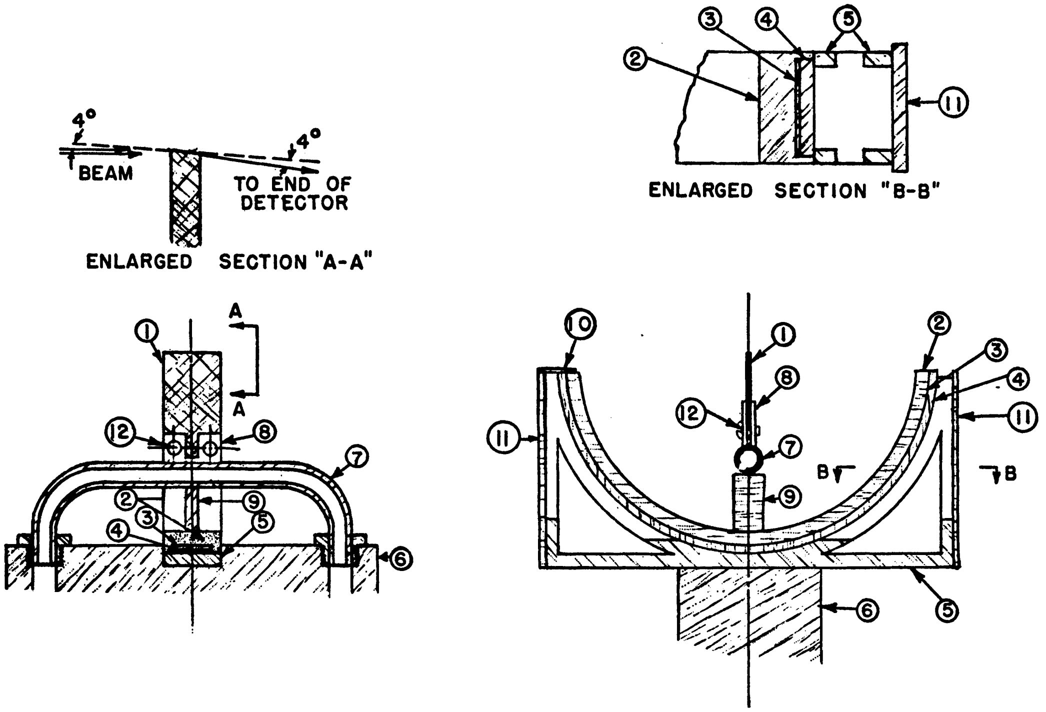

FIG. 1. Target assembly for measuring angular distributions of neutrons from (p,n) reactions: (1) target, (2) detector cover (carbon), (3) detector foils (Al and Ag), (4) detector backing (carbon), (5) assembly body (brass), (6) header for cooling water (brass), (7) water cooling tube (copper), (8) target holder (brass, soldered to cooling tube), (9) wedge (carbon) for holding the detector cover, foils, and backing against the assembly body, (10) detector positioner (carbon), (11) shields (carbon), (12) target clamping screws. Principle sections are through center of assembly. Distance from target tip to detectors is $3\frac{3}{8}$ in. Nearest material, not shown, (dees) is $4\frac{1}{4}$ in. from closest part of assembly.

least, errors which could materially affect the conclusions are highly improbable.

The neutrons were detected by silver and aluminum threshold detectors, the reactions being

$$\mathrm{Ag}^{107}(n,2n)\mathrm{Ag}^{106}\rightarrow\mathrm{Pd}^{106}+\beta^{+}\ (24\ \mathrm{min}),$$

$$\mathrm{Al}^{27}(n,p)\mathrm{Mg}^{27}\rightarrow\mathrm{Al}^{27}+\beta^{-}\ (9.6\ \mathrm{min}).$$

The silver reaction has an energetic threshold at 9.6 Mev, and is theoretically expected to rise quite rapidly to a maximum within a few Mev. Since the energy distributions of the neutrons are Maxwellian, the average energy of the neutrons it detects is about 13 Mev. The aluminum reaction has an energetic threshold at about 2.0 Mev, but theoretically the cross section should remain quite small until the energy available to the emitted proton approaches the Coulomb barrier, which is about 4.5 Mev. Since the neutron energy distributions are Maxwellian, the average energy of the neutrons it detects is about 8 Mev.

The target assembly is shown in Fig. 1. The explanation for its various features will become evident in the discussion to follow. In its simplest form it consists of a target of the element to be studied at the center of a semicircle, with an aluminum and silver foil lying along the circumference of the semicircle and covered by an absorber sufficiently thick to remove all charged particles. The principle difficulty was in the determination and elimination of background.

The most obvious way of being certain that the detected neutrons originate in the target is to stop the entire beam there. In the ORNL 86-inch cyclotron this cannot be done because the beam is 6 inches wide, while the largest practicable radius for the detector semicircle is $3\frac{1}{2}$ inches; this would give very poor angular resolutions and, in addition, there would be no way of measuring backgrounds. These problems were solved by inserting a carbon probe in the cyclotron 180 deg from the target assembly. Narrow targets can then be used, with the remainder of the beam being stopped by the probe. Since the probe is twenty times further from the detectors than the target, and since carbon gives very low neutron production,[12] the probe does not produce an appreciable background. Furthermore, stopping the major portion of the beam on the probe allows determinations of background. This is done by making consecutive runs with and without the target, but with the same current striking the probe. With the exception of the very small and very large angles, which will be discussed below, the background measured in this way was less than 10 percent. In fact, about 80 percent of

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[12] Carbon-12 has a (p,n) threshold near 20 Mev, so that the principle neutron production is from carbon-13 (a 1.1 percent isotope) and impurities.

all neutrons, as measured by an energy-insensitive neutron counter at a large distance from the cyclotron, originate in the target even though more than 80 percent of the current strikes the probe.

In the runs made with the target removed, a high activity is produced in the small angle detector foils; it falls off very rapidly with increasing angle. By using proton detectors it was found that this was due to stray beam striking the detector covers, thus producing neutrons very close to the detectors. The best way to eliminate this difficulty, it was found, was to make the targets thick, so as to act as a shield against this stray beam. Use of thick targets somewhat obscures the detailed results of the measurements by making the protons which produce the neutrons by (p,n) reactions nonmonoenergetic. Since only the general form rather than the detailed shape of the angular distributions is used in the theoretical interpretation, this is not a major difficulty. Moreover, the use of thick targets alleviates many other experimental problems.

There still remains the difficulty of measuring the background at small angles. One approach was to make bombardments with a carbon target. These runs gave small angle intensities about 10 percent of those with other target elements, but there were strong indications that a large fraction of that was due to neutrons from the carbon target.[12] Backgrounds at the forward angles were therefore assumed to be the same as those at 45 degrees, which were generally about 5 percent. Part of this background is undoubtedly due to the above-mentioned stray beam, but no large error is possible by this procedure.

In the backward direction there is considerable background in the silver-detector data, presumably due to stray beam striking the back of the detector holder. At 165 deg this is frequently larger than foreground. It drops off rapidly with decreasing angle, averaging about 40 percent at 150 deg. Since this correction is probably uncertain by 25 percent, it introduces an error of more than 10 percent at angles greater than 150 deg. In the figures, the data at these angles are therefore indicated only by a dashed line showing its general trend, although even this is uncertain. In the aluminum-detector data, this background is not nearly as large; even at 165 deg it was never larger than 20 percent.

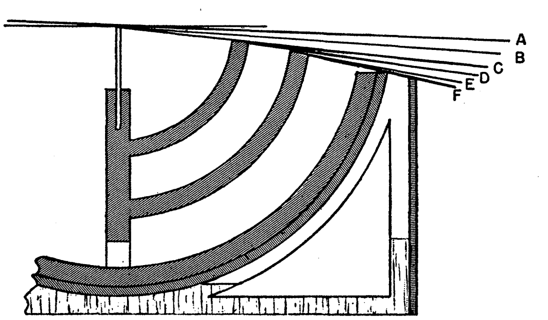

FIG. 2. Target modification to test effect of elastically scattered protons. Except for effects of transparencies and imperfections, beam particles scattered from the target cannot get below line B; if they could, they would not get below line C unless scattered from inner carbon piece; if they were scattered from it, they could not get below line D; etc. This should eliminate any maximum in the forward direction due to (p,n) reactions induced in the detector cover by elastically scattered protons.

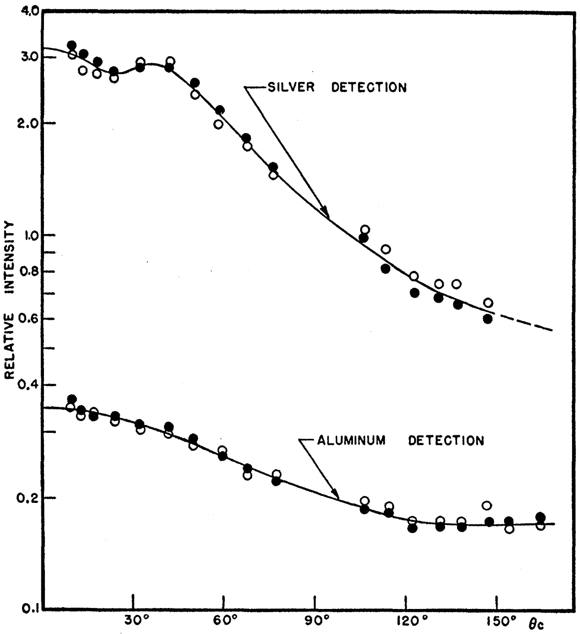

FIG. 3. Angular distribution of neutrons from Al(p,n). Energy of incident protons was 23 Mev, and target was thick. Each set of points represents an average of two or three runs. Silver and aluminum detect only neutrons above 9.5 Mev and 2.0 Mev, respectively; the average energy of the neutrons they detect is about 13 Mev and 8 Mev. Data are corrected for center-of-mass angle and solid angle, and background has been subtracted.

In addition to these backgrounds which are present when the target is removed, there are still two possible sources of background that are inherently due to the target itself:

(1) Protons, elastically scattered from the target, could produce neutrons in the detector covers. Since thick targets were used, it was possible to bevel the tops of the targets so that protons would require several scatterings in the target before they could reach the detector covers (see Sec. A–A, Fig. 1). The effect was then studied by constructing the target assembly shown in Fig. 2. Protons scattered from the target would have to undergo several scatterings in order to reach the detector cover, so that these effects should be greatly reduced. Aside from causing a uniform decrease in intensity relative to backward angles (due to absorption), it did not change the observed angular distributions; the effect of scattered protons was thus considered to be negligible.

(2) Neutrons originating in the target could be scattered into the detectors. Every effort was made to reduce this by keeping the amount of extraneous material to the bare minimum. The geometry was such

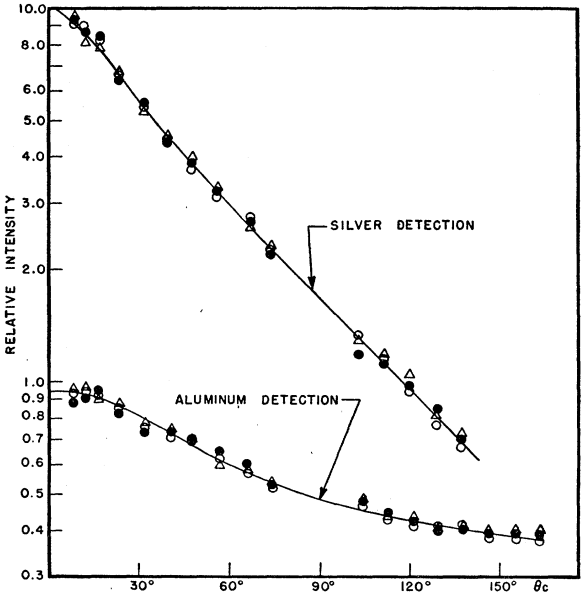

FIG. 4. Angular distribution of neutrons from Cu(p,n). See caption for Fig. 3. Ordinate scale is same as in Fig. 3.

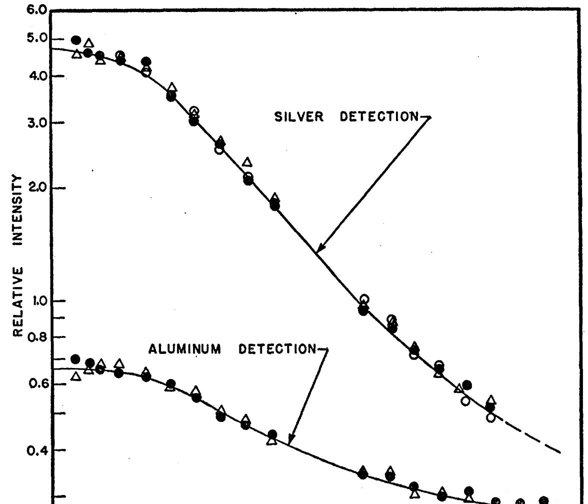

FIG. 5. Angular distribution of neutrons from Ag(p,n). See captions for Figs. 3–4.

that all detectors were closer to the target than to any part of the cyclotron dees. Calculations indicate that under these conditions effects due to scattered neutrons are quite small. To test this, thick pieces of carbon were added to the back and each side of the detector semicircle at the backward angles. Neither the intensities relative to those at the forward angles, nor the shape of the angular distributions was noticeably changed; thus this effect can be assumed to be negligible. As an added precaution, with a few trivial exceptions, everything in the experimental setup was maintained symmetric about 90 deg; this assures that scattered neutrons cannot have an important effect.

The expected errors in these measurements due to well-known effects, such as counting statistics, beta counting errors, and decay corrections, are about 6 percent at each point; this could have very little effect on the general shape of the angular distributions. Inaccuracies in target and detector alignment should not produce errors greater than about 4 percent in the asymmetry about 90 deg. Angles are known to within about 2 degrees.

RESULTS

Angular distributions were measured for Mg, Al, Cu, Mo, Ag, Ta, Au, Th, and U. Some typical data are shown in Figs. 3–6, and the lines through the data for all elements are shown in Figs. 7 and 8. Backgrounds have been subtracted, and corrections have been applied for center-of-mass motion. If it is assumed that the neutrons are emitted with a Maxwellian energy distribution characterized by temperature T, this can be corrected for quite accurately even though there is considerable uncertainty in the excitation functions for the detecting reactions. For example, these corrections are the same whether the excitation functions rise sharply to a plateau, rise gradually and monotonically, or consist of one or more resonances, either very wide or very narrow, providing these effects occur at about the same energy; and even the dependence on energy is relatively slight. These corrections are shown by the dashed lines in Figs. 7 and 8, as calculated for $T=2.5$ Mev for Mg and

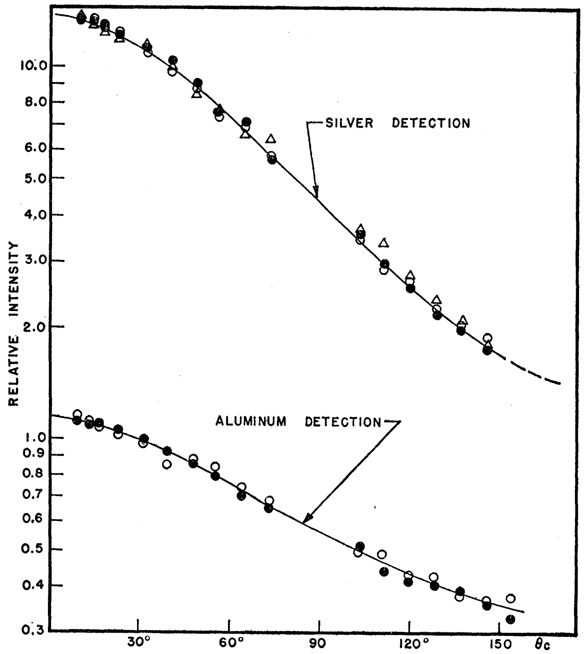

FIG. 6. Angular distribution of neutrons from Ta(p,n). See captions for Figs. 3–4.

Al, and for $T=2.0$ Mev for the other elements. There is some evidence for these values, as will be mentioned below, but the correction for other values of T can easily be estimated by assuming the percentage corrections to be inversely proportional to T.

The data in Figs. 3–6 are normalized to the same current striking the 180-deg probe; this is roughly equivalent to normalizing them to the same current striking the targets. The relative average cross sections for neutron production in the various target elements can thus be easily computed by correcting for the range of protons in the various materials. This is shown in Fig. 9 for neutrons detected by aluminum and by silver, and for all neutrons as detected by an energy-insensitive counter.

By making use of the theoretical total reaction cross sections given by Blatt and Weisskopf,[2] the data for all neutrons from Fig. 9 can be corrected for the relative number of nuclear reactions to be expected, to give the relative number of neutrons per nuclear reaction; this is illustrated in Fig. 10. The excitation functions for the principle neutron-producing reactions in copper have been measured by activation experiments,[6,13] so that the absolute number of neutrons per reaction can be closely estimated for that element. This was used to normalize the data in Fig. 10. A point on the curve at $Z=83$ can be obtained from Kelly's excitation functions on bismuth.[14]

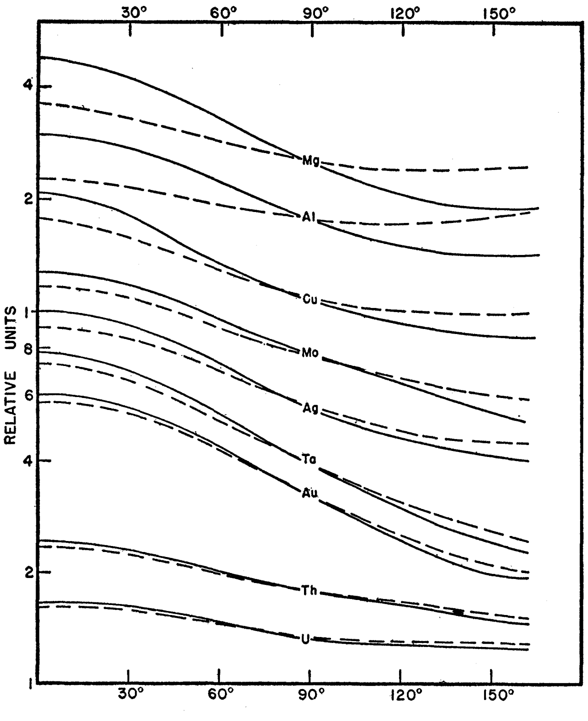

FIG. 7. Composite of aluminum detection data. Dashed lines show corrections for variation of energy with angle due to center-of-mass effects. They were calculated by assuming temperatures of 2.5 Mev for Mg and Al, and 2.0 Mev for the other elements. Corrections are approximately inversely proportional to the temperature.

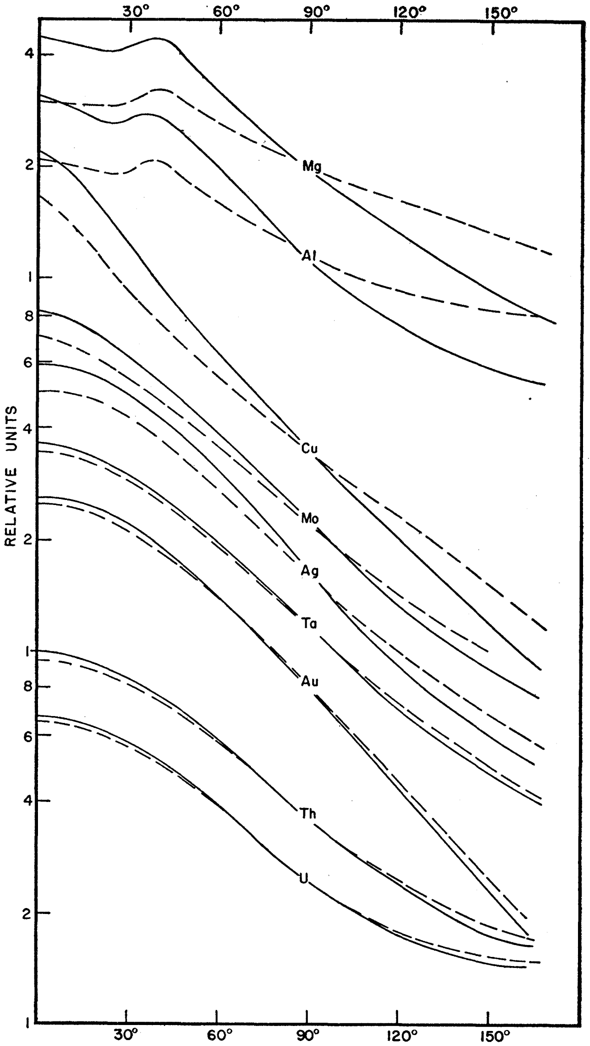

FIG. 8. Composite of silver detection data. See caption for Fig. 7.

Figures 11 and 12 show the relative fraction of all neutrons that are detected by aluminum and silver, respectively, at 0, 90, and 150 deg for the various elements. These were obtained by dividing the detector activities by the neutron counter readings.

DISCUSSION AND CONCLUSIONS

The principle conclusion from this work is that, contrary to what is expected from compound nucleus theory and in agreement with expectations from direct interactions,[15] the angular distributions are not nearly sym-

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[13] Cohen, Newman, Charpie, and Handley, Phys. Rev. **94**, 620 (1954).

[14] E. Kelly, University of California Radiation Laboratory Report UCRL-1044 (unpublished).

[15] The term "direct interaction" is used here to mean not only interactions consisting of a single, direct collision between the incident and emerging particles, but any interaction in which

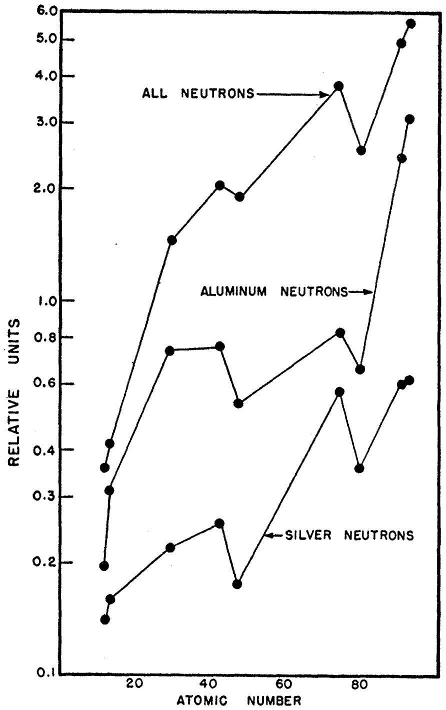

FIG. 9. Cross section for production of neutrons *vs* atomic number. This data was obtained from yields by correcting for stopping cross sections of the materials.

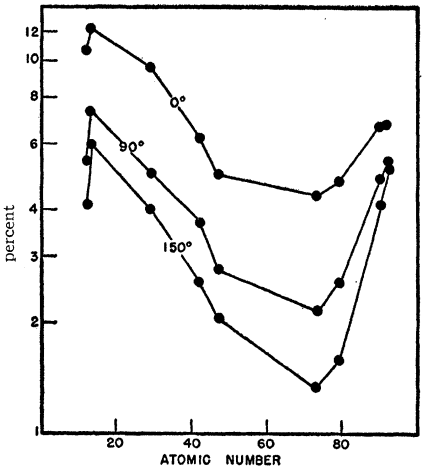

FIG. 11. Percent of all neutrons that are detected by aluminum. Ordinate was calibrated as described in final paragraph of this paper neglecting anisotropy of "all neutrons."

metric about 90 deg, but rather are quite strongly peaked in the forward direction. The large magnitude of the effect is especially interesting. For the high-energy neutrons, the 0-deg intensity is in many cases higher than the 180-deg intensity by an order of magnitude. This implies that almost none of these neutrons are produced in compound nucleus reactions.[16] The minima at about 25 deg and the subsequent maxima at about 40 deg in the magnesium and aluminum data suggest that some sort of resonant process analogous to that in deuteron stripping is in effect. Such a process

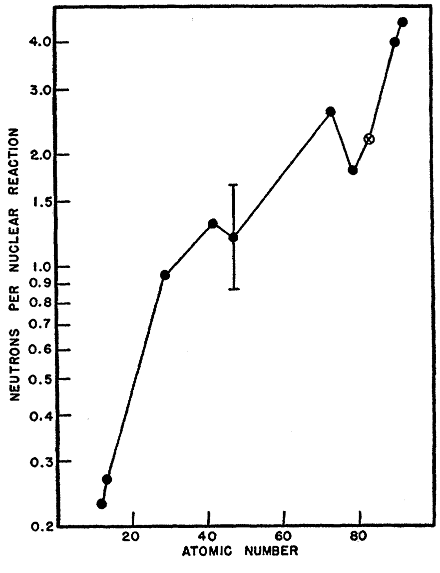

FIG. 10. Neutrons per nuclear reaction. Cross is data from reference 14.

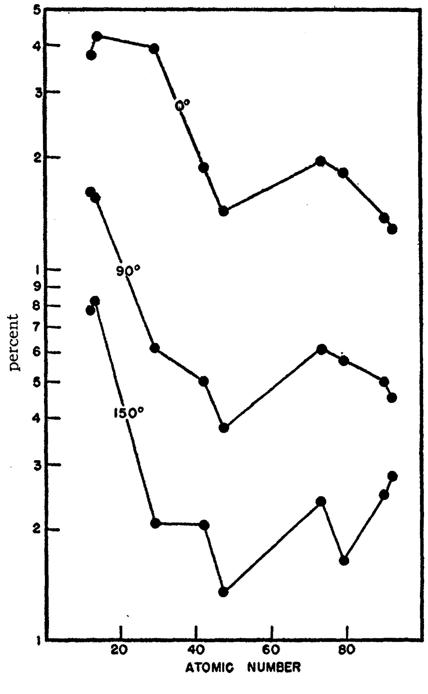

FIG. 12. Percent of all neutrons that are detected by silver. Lower decade of ordinate should be divided by ten so that ordinate scale covers 0.1 to 5.0 percent. See caption for Fig. 11.

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the outgoing particle is emitted before the energy has been shared with more than a small fraction of the nucleons in the nucleus. See R. M. Eisberg, Phys. Rev. **94**, 739 (1954).

[16] L. Wolfenstein, Phys. Rev. **82**, 690 (1951); W. Hauser and H. Feshbach, Phys. Rev. **87**, 366 (1952).

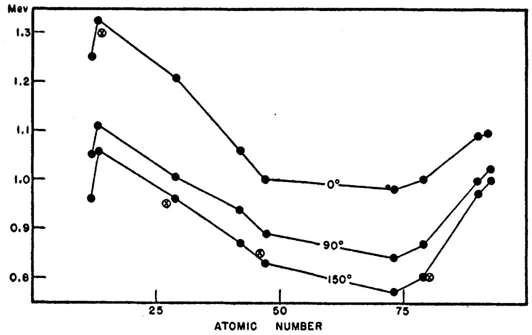

FIG. 13. Temperatures of neutron energy spectra at about 3-Mev neutron energy. Crosses are from reference 3.

has been suggested by Austern *et al.*[17] On the other hand, the general similarity between angular distributions from elements in the same mass region is almost suggestive of some sort of nonresonant process (e.g., charge exchange, diffraction). In the aluminum-detector data, the 0°/180° ratio is never larger than five, and in the light elements is generally less than two. This may indicate that an appreciable fraction of these neutrons are from compound nucleus interactions, although even from direct interactions lower energy angular distributions would be more isotropic. The rather small anisotropies in thorium and uranium are probably due to the fact that fission occurs with higher probability in these elements, and fission neutrons are emitted approximately isotropically. The smaller anisotropies in the lighter elements may be due to the fact that most noncompound nucleus interactions in these elements result in proton emission.[18]

Several interesting conclusions may be drawn from the yield data, in spite of the fact that it is generally somewhat rough. The very low neutron production in aluminum and magnesium is quite surprising, although it complements the data on $(p,pn)+(p,2n)$ cross sections[19] where essentially the same effect is found to be quite common on elements up to about mass 60.

The very high neutron production in thorium and uranium is undoubtedly due to fission. However, those elements also have the highest cross section for production of silver-detected neutrons. Since these neutrons exhibit a markedly forward angular distribution, they cannot originate from fission. The nearly isotropic angular distributions of the aluminum-detected neutrons from thorium and uranium indicate that these are principally fission neutrons. The ratio of "aluminum neutrons" to total neutrons is quite sensitive to the temperature[20] with which the neutrons are emitted, so that if the fission temperature is assumed to be 1.0 Mev, all other temperatures may be readily estimated. These estimates are shown in Fig. 13, and may be considered to be the average temperature at a neutron energy of about 3 Mev. Figure 13 also shows the temperatures found by Gugelot[3] at these neutron energies from 16-Mev proton-induced reactions. The agreement is excellent. It is important to note that, as has been pointed out in another paper,[18] the spectra at these energies are largely determined by "second" neutrons from (p,pn) and $(p,2n)$ reactions, and therefore are not too meaningful.

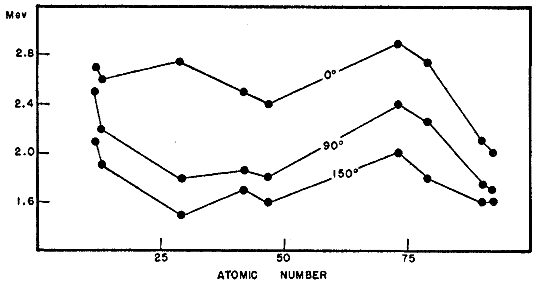

FIG. 14. Temperatures of neutron energy spectra at about 10-Mev neutron energy.

Of somewhat more interest is the temperature at higher neutron energies. A simple calculation reveals that the ratio of activities between the silver and aluminum should be very sensitive to the temperature of the neutron energy spectrum. This ratio has been measured for $T=2.5$ Mev with neutrons from the Be(d,n) reaction[21]; from that ratio, the values of T required to explain the ratios of silver to aluminum activities in this experiment were obtained and are shown in Fig. 14. These values are the temperatures of that portion of the spectrum between the energies detected by aluminum and silver, or roughly about 10 Mev. It is interesting to note that these temperatures agree with those obtained from Gugelot's inelastic proton scattering data at 10-Mev outgoing proton energy.

By use of Figs. 13 and 14, the energy spectrum may be roughly approximated, so that the absolute fraction of all neutrons detected by aluminum and silver may be estimated. This calculation was used to obtain the ordinates in Figs. 11 and 12. The relatively small fraction of all neutrons detected in these experiments indicates that compound nucleus formation may still be the principle interaction.

The author is greatly indebted to Mrs. M. K. Hullings for her invaluable assistance in parts of the counting and calculating.

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[17] Austern, Butler, McManus, and Sharp, Phys. Rev. 91, 453 (1953).

[18] B. L. Cohen and E. Newman (to be published).

[19] Cohen, Newman, and Handley (to be published).

[20] The term "temperature" is used here in a purely experimental sense, referring to the reciprocal of $\partial/\partial E[\log(N(E)/E)]$ where $N(E)$ is the energy distribution of the emitted neutrons.

[21] B. L. Cohen, Phys. Rev. 81, 184 (1951).