cross sections show the characteristic shape which is expected for compound nucleus reactions. However, cross-section measurements only yield information about the energy spectra of the emitted particles, and agreement with values calculated from evaporation theory should not be interpreted to mean that the particle emission is isotropic. Bell and Skarsgard[15] and Jackson[16] studied the reactions $Bi^{209}(p,xn)Po^{210-x}$, and found agreement (for $x\geq 3$) with a simple evaporation theory using an energy-independent nuclear temperature. At incident particle energies well above the compound nucleus resonance, there was a small residual cross section attributed to direct interaction processes.

The angular distribution of neutrons with energies above 1 Mev from the bombardment of a thick bismuth target with 15-Mev deuterons and 30-Mev helium ions was measured by Allen et al.[17] After correction by Cohen,[18] the distribution was almost isotropic from the helium ion bombardment, but strongly peaked in the forward direction from the deuteron bombardment. The bombarding energies were too low for the reactions studied in the present paper to be contributors to the neutron yield.

Toms and Stephens[19] found that about 90% of the photoneutrons obtained from the bombardment of lead with 23-Mev bremsstrahlung fitted an evaporation spectrum with $T=1.35$ Mev. The angular distribution of neutrons with energies above 4 Mev was almost isotropic. Price,[20] however, found that only the low-energy (0–few kev) neutrons from the bombardment of bismuth with 22-Mev bremsstrahlung were isotropic.

Gugelot[21] measured the energy spectra of neutrons arising from the bombardment of gold and thallium with 16-Mev protons. The spectra could be fitted to the equation $P(E)\propto Ee^{-E/T}$ with $T=0.77$ Mev (gold) or 0.71 Mev (thallium).

All these experiments suggest that, in a variety of nuclear reactions, neutron emission from heavy nuclei is predominantly due to an evaporation mechanism. The present experiment confirms these observations by a new method.

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Recoil Studies of Heavy Element Nuclear Reactions. II*

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Angular distributions and ranges of recoils from the reactions $Bi^{209}(\alpha,2n)At^{211}$ and $Cm^{244}(\alpha,2n)Cf^{246}$ have been measured. At helium ion energies higher than about 10 Mev above the Q values of these reactions, the results are consistent with a reaction mechanism involving the emission of one or both neutrons in the forward hemisphere.

INTRODUCTION

During the course of investigations of the type reported in the preceding paper (Paper I), several reactions were found which gave recoil angular distributions markedly different from those predicted by the isotropic evaporation model. Two of these reactions, the reactions $Bi^{209}(\alpha,2n)At^{211}$ and $Cm^{244}(\alpha,2n)Cf^{246}$, have been studied in some detail in an effort to define the reaction mechanisms.

Recoil angular distributions alone were not sufficient to determine unambiguously the mechanisms. The $Bi^{209}(\alpha,2n)$ reaction gave angular distributions of the At^{211} product which, at the higher bombarding energies, were much narrower than those given by Monte Carlo calculations based on a simple evaporation model. (See Paper I for discussion of the Monte Carlo calculations); on the other hand, the $Cm^{244}(\alpha,2n)Cf^{246}$ angular distributions were substantially broader than those of the At^{211}. In order to gain further insight into the dynamics of these reactions, ranges of the recoiling product nuclei were measured. These ranges, when interpreted by means of range-energy relations, define the momentum of the recoiling nucleus along the beam axis; and hence, since the incident particle energy is known, the total momentum of the outgoing neutrons along the beam axis. This additional information allows one to investigate possible mechanisms in more detail.

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EXPERIMENTAL

The techniques used in making the recoil angular distribution measurements were similar to those described in Paper I. Curium targets were prepared by vaporization. The targets were alpha counted and the target thicknesses were calculated from the counting rates. Target thicknesses of about one microgram per square centimeter were found to be satisfactory from a scattering standpoint; however, because of the high temperatures required for curium vaporization, there was a tendency for films of foreign material to deposit on the target during the vaporization process. Such films often caused targets to have effective thicknesses several times greater than those indicated by the alpha counting method. In order to avoid being misled by angular distributions from such targets, the curium was vaporized on to thin bismuth targets. These targets were then bombarded, and only those giving angular distributions for the bismuth reaction products in agreement with data previously obtained from thin bismuth targets were used. This procedure made it possible to re-use curium targets until the bismuth product angular distributions indicated that scattering was taking place.

Recoil range measurements were made in two ways: (1) differential range curves were obtained by stopping the recoils in a stack of thin plastic films, and (2) average ranges were determined by measuring the number of recoils escaping from very thick targets.

For the thin film absorption measurements, targets were prepared in the manner described in Paper I for the angular distribution studies. Targets of about one microgram of target material per square centimeter were used. The absorber films were made of VYNS plastic, prepared by stretching on a water surface in the manner described by Pate and Yaffe.[1] The films, which were 1 inch in diameter, were mounted on aluminum frames which were spaced in slots 0.030 inch apart. The film thicknesses were usually in the range from 5 to 15 micrograms per square centimeter. Depending on the film thicknesses, a stack of 15 to 20 films was used to stop the recoils. Film thicknesses were measured by the optical absorption method described by Pate and Yaffe,[1] by use of a Beckman Model DU spectrophotometer at a wavelength of 600 mμ, using the transmission-thickness calibration curve given by Pate and Yaffe. Results of successive measurements made on the same film were quite reproducible, and measurements made at different areas of the same film were usually in good agreement. Several films were weighed on a microbalance. The weights given by this method were in good agreement with those calculated from the transmission measurements. The film thicknesses as measured before the bombardment were probably accurate to within about one microgram per square centimeter.

The recoil target assembly used for the angular distribution measurements was also used for the recoil range studies. The carrier holding the plastic absorber films was placed close to the target; the first plastic film was about 0.5 cm from the target surface. The beam was collimated to $\frac{1}{4}$-inch diameter by means of a graphite collimator. The target chamber was evacuated to 50 to 100 microns pressure during the bombardment.

Beam intensities of 0.2 to 0.5 microampere were usually used; the duration of the bombardments was several hours. The passage of the beam through the plastic films caused them to become extremely fragile near the center; and there were very possibly dimensional and chemical changes in the plastic, so that the portion of the films that the beam passed through may have had a thickness appreciably different from that of the rest of the film. In general, the counting procedures were much the same as those previously described for the angular distribution studies. The yields of astatine or californium isotopes stopped in each film were measured by direct gross α-counting or α-pulse analysis of the films; no chemical separations were made.

The measurement of recoil ranges in thin plastic films is a difficult operation. In spite of all the precautions taken, the absolute values of the recoil ranges measured by this method were not consistently reproducible. The reason for this is not understood. However, useful information can still be derived from these recoil measurements because the ratio of the recoil ranges for the various reaction products in any one bombardment was quite reproducible. Consequently, the conclusions concerning reaction mechanisms that are inferred from the thin film recoil range data are based only on the relative recoil ranges for the various reaction products studied in a particular bombardment, rather than on the absolute recoil ranges themselves.

Targets for thick target range determinations were prepared by vaporizing bismuth on to 0.001-inch thick aluminum foil in vacuum. The thickness of the deposited bismuth was measured by weighing pieces of the aluminum foil before and after the vaporization. The targets contained 2.5 milligrams per square centimeter of bismuth. Variations in vaporization geometry gave rise to fluctuations in target thickness of about 3%. The targets, which were about 4 cm^2 in area, were cut from a large foil coated in a single vaporization. Eleven targets were arranged in a stack with a 0.001-inch aluminum catcher foil behind each target. The stack was exposed in vacuum to the full energy helium ion beam of the cyclotron, with the bismuth sides of the targets facing downbeam. The aluminum foils had a thickness of 7.3 milligrams per square centimeter. Each bismuth layer degraded the beam about 0.2 Mev. Range measurements were thus obtained from a single bombardment at 11 energies ranging from 46.6 Mev to 22.2 Mev. At energies where the excitation function of the reaction under investigation was very steep, it was necessary to apply a small correction to the measured recoil range, arising from the variation in cross section through the thickness of the target.

[1] B. D. Pate and L. Yaffe, Can. J. Chem. 33, 15 (1954).

All the astatine recoil products in the catcher foils were measured by gross alpha counting and alpha pulse height analysis. The recoil products in the thick targets were measured by gross alpha counting. The astatine was then volatilized from the targets and condensed on cold platinum disks, which were then pulse analyzed to provide abundance ratios of the various astatine isotopes. The average range of any given recoil species is given by the expression

$$R_{\text{average}} = N_c T/(N_c + N_T), \tag{1}$$

where R_{average} is the average range in the target material for this recoil species, N_c is the number of recoils stopped in the catcher, N_T is the number of recoils remaining in the target, and T is the target thickness. Thick target ranges were not measured for the $Cm^{244}(\alpha,2n)Cf^{246}$ reaction because of the large amount of curium that would have been required.

It should be pointed out that the measured ranges are actually the components of the range along the beam axis. The angular distributions of the various recoils were sufficiently narrow and unvarying that these components are valid quantities for the comparison of one reaction with another.

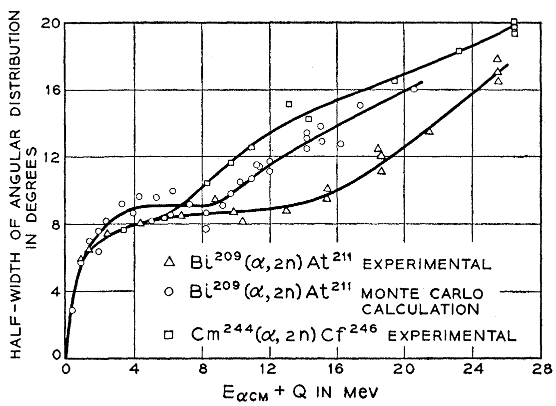

FIG. 1. Angular distribution half-width as a function of $E_{\alpha cm}+Q$ for the reactions $Bi^{209}(\alpha,2n)At^{211}$ and $Cm^{244}(\alpha,2n)Cf^{246}$.

RESULTS AND CONCLUSIONS

A plot of the half-width of the angular distribution versus energy is shown in Fig. 1 for the reactions $Bi^{209}(\alpha,2n)At^{211}$ and $Cm^{244}(\alpha,2n)Cf^{246}$. Both Monte Carlo and experimental results are shown. The Monte Carlo calculations shown in Fig. 1 were for the $Bi^{209}(\alpha,2n)$ reaction. At values of $E_{\alpha cm}+Q_2$ greater than about 10 Mev, the experimental half-widths for both reactions differ considerably from those given by the Monte Carlo calculations.

The difference in the angular distributions of the $Bi^{209}(\alpha,2n)$ reaction and the $Cm^{244}(\alpha,2n)$ reaction is probably due to the difference in the amount of energy which must be carried off by the neutrons. The fission

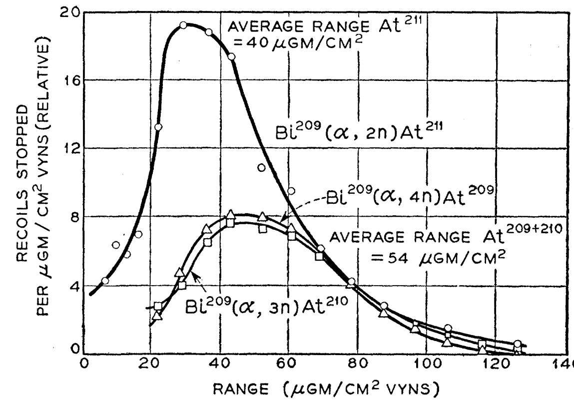

FIG. 2. Recoil differential range curves for the reactions $Bi^{209}(\alpha,2n)At^{211}$, $Bi^{209}(\alpha,3n)At^{210}$, and $Bi^{209}(\alpha,4n)At^{209}$ ($E_\alpha=43.6$ Mev).

activation energy for Cf^{246} is about 4.5 Mev[2]; and after emission of the second neutron, the residual excitation energy must be less than this amount if the nucleus is to survive. The binding energy of the last neutron in At^{211} is 7.7 Mev, and the At^{211} nucleus can therefore retain an excitation energy up to this amount and survive. In addition, the Q values for the two reactions differ by 1 Mev; as a result of these two effects, the neutrons must carry away a total of 4.2 Mev more energy to produce Cf^{246} than to produce At^{211}. The Cf^{246} angular distributions are therefore broader than the At^{211} angular distributions.

Since one would expect the conditions for isotropic neutron evaporation from a compound nucleus to be better met at high excitation energies, these results lead one to suspect that a mechanism other than compound-nucleus formation is starting to contribute to the $(\alpha,2n)$ reactions at about 30 Mev, which is at the peak of the $(\alpha,2n)$ excitation functions for both reactions.[3,4] Further

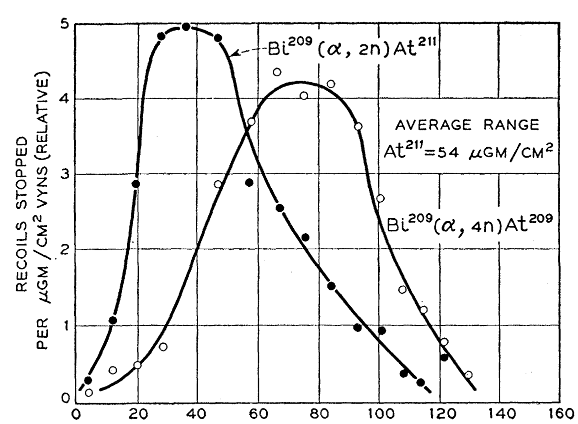

FIG. 3. Recoil differential range curves for the reactions $Bi^{209}(\alpha,2n)At^{211}$ and $Bi^{209}(\alpha,4n)At^{209}$ ($E_\alpha=46.5$ Mev).

[2] R. Vandenbosch and G. T. Seaborg, Phys. Rev. **110**, 507 (1956).
[3] A. Chetham-Strode, Jr., G. R. Choppin, and B. G. Harvey, Phys. Rev. **102**, 747 (1956).
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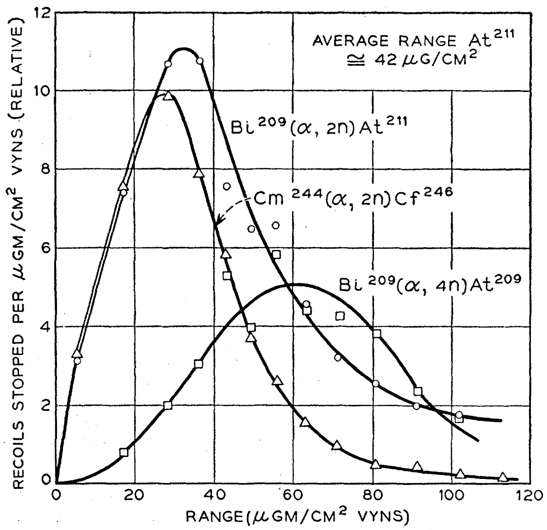

FIG. 4. Recoil differential range curves for the reactions $Bi^{209}(\alpha,2n)At^{211}$, $Bi^{209}(\alpha,4n)At^{209}$, and $Cm^{244}(\alpha,2n)Cf^{246}$ ($E_\alpha=46.5$ Mev).

evidence that a compound-nucleus mechanism begins to contribute to the $(\alpha,2n)$ reaction at helium ion energies above 30 Mev is supplied by the results of the thin target recoil measurements. Results of a single experiment in which the ranges of recoils were simultaneously measured for the $(\alpha,2n)$, $(\alpha,3n)$, and $(\alpha,4n)$ reactions of Bi^{209} with 43.6-Mev incident helium ions are shown in Fig. 2. There is a striking similarity between the differential range curves of the $Bi^{209}(\alpha,3n)$ and $Bi^{209}(\alpha,4n)$ reactions. The angular distribution studies in Paper I indicate that the $(\alpha,3n)$ and $(\alpha,4n)$ reactions of Bi^{209} both proceed by a compound-nucleus mechanism. The similarity in the differential range curves for these reactions is further evidence that both reactions proceed by the same mechanism. The peak of the range curve for either of these reactions may then be interpreted as the most probable range of recoils having the full momentum of the incident helium ion. The width of the curve is due to a combination of range straggling and deviations from the most probable recoil momentum caused by the momenta given to the recoils by the outgoing neutrons. The average range, which is what we wish to compare with the thick target range results, is the range which half the recoils exceed. The average range in most of the differential range curves is somewhat greater than the most probable range due to a long range "tail." As may be seen in Fig. 2, the average range for $Bi^{209}(\alpha,2n)At^{211}$ recoils produced by 43.6-Mev helium ions is considerably less than that for the $Bi^{209}(\alpha,3n)At^{210}$ and $Bi^{209}(\alpha,4n)At^{209}$ recoils. This can only mean that many of the At^{211} recoils have failed to absorb the full momentum of the incoming helium ion. In other words, a compound nucleus was not formed.

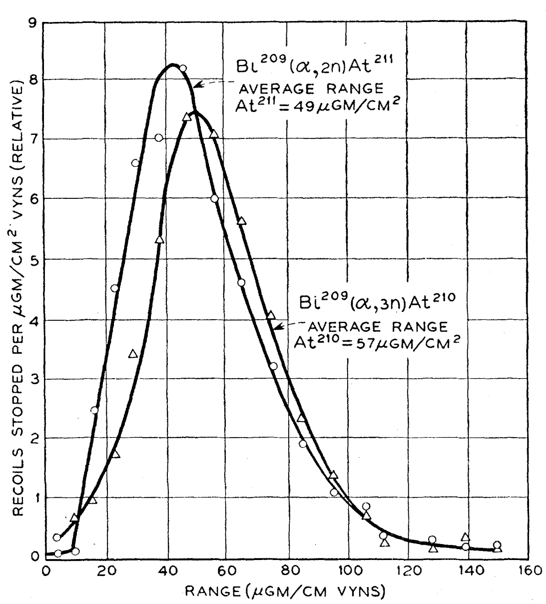

FIG. 5. Recoil differential range curves for the reactions $Bi^{209}(\alpha,2n)At^{211}$ and $Bi^{209}(\alpha,3n)At^{210}$ ($E_\alpha=38.6$ Mev).

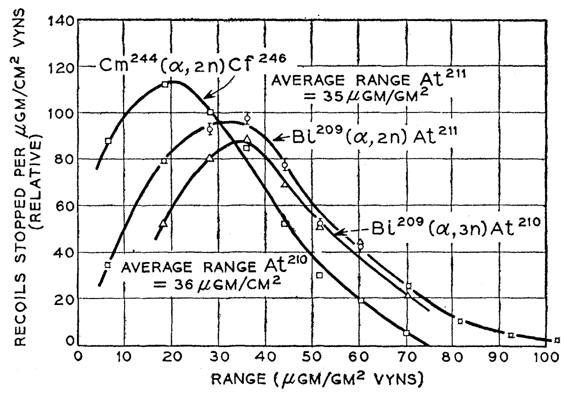

FIG. 6. Recoil differential range curves for the reactions $Bi^{209}(\alpha,2n)At^{211}$, $Bi^{209}(\alpha,3n)At^{210}$, and $Cm^{244}(\alpha,2n)Cf^{246}$ ($E_\alpha=33.0$ Mev).

According to the range-energy relation derived by Bohr[5] and Lindhard and Scharff[6], the ranges in light stopping media of recoils of the mass and energy studied in this work should be proportional to their energies. This type of range-energy relation is also supported by calculations based on experimental results of Baulch and Ducan,[7] Leachman and Atterling[8]; Sikkeland and Ghiorso[9]; Harvey, Wade, and Donovan[10]; and Morton, Valyocsik, Donovan, and Harvey.[11] If, then, the recoil range is proportional to its energy, it is possible to

[5] N. Bohr, Kgl. Danske Videnskab. Selskab. Mat.-fys. Medd. 18, No. 8 (1948).

[6] J. Lindhard and M. Scharff, Kgl. Danske Videnskab. Selskab. Mat.-fys. Medd. (to be published).

[7] D. L. Baulch and J. F. Duncan, Australian J. Chem. 10, 112 (1957).

[8] R. B. Leachman and H. Atterling, Arkiv Fysik 13, 101 (1957).

[9] T. Sikkeland and A. Ghiorso, University of California Radiation Laboratory (unpublished data, 1958).

[10] B. G. Harvey, W. H. Wade, and P. F. Donovan, University of California Radiation Laboratory (unpublished data, 1958).

[11] J. R. Morton, E. W. Valyocsik, P. F. Donovan, and B. G. Harvey, University of California Radiation Laboratory (unpublished data, 1958).

calculate the energy of the lower range $(\alpha,2n)$ recoils from the ratio of the ranges of the $(\alpha,2n)$ recoils to those of the $(\alpha,3n)$ or $(\alpha,4n)$ recoils since these last two reactions must proceed by a compound-nucleus mechanism on the basis of the results discussed above.

Range curves for the products of the $(\alpha,2n)$ and $(\alpha,4n)$ reactions of Bi^{209} measured at an incident helium ion energy of 46.5 Mev are shown in Fig. 3. Another measurement at this same energy, this time including, in addition, the $Cm^{244}(\alpha,2n)Cf^{246}$ reaction, is shown in Fig. 4. Although the absolute ranges have not been reproduced, the relative ranges have. In order to compare the range of californium recoils with that of astatine recoils, the californium range should be multiplied by about 1.1 to correct for the $Z^{-\frac{2}{3}}$ dependence of range on nuclear charge.[5]

Differential range curves measured at 38.6 Mev and 33.0 Mev are shown in Figs. 5 and 6. The average range for the $(\alpha,2n)$ reaction product gradually approaches

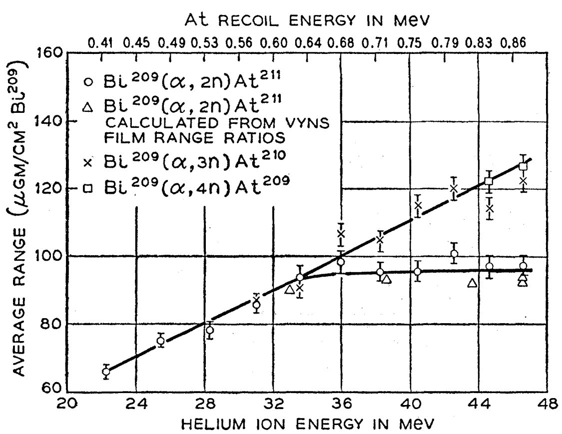

FIG. 7. Thick target ranges as a function of incident helium ion energy for the reactions $Bi^{209}(\alpha,2n)At^{211}$, $Bi^{209}(\alpha,3n)At^{210}$, and $Bi^{209}(\alpha,4n)At^{209}$.

that of a compound-nucleus reaction as the incident particle energy is lowered. This approach takes place at a higher energy for the $Bi^{209}(\alpha,2n)$ reaction than for the $Cm^{244}(\alpha,2n)$ reaction, as might be expected from the angular distribution data shown in Fig. 1. The discrepancy between the experimental $(\alpha,2n)$ angular distribution data and the Monte Carlo results at the lower energies ($E_{\alpha cm}+Q_2<7$ Mev) is just about at the limit of the estimated errors in the half-widths (about $\pm1°$ for the Monte Carlo results and about $\pm\frac{1}{2}°$ for the experimental results).

Results of the thick target range measurements are shown in Fig. 7. Included for comparison are the recoil range data from the thin film measurements for the $Bi^{209}(\alpha,2n)$ reaction. Only the ratios of ranges of $(\alpha,2n)$ product to $(\alpha,3n)$ or $(\alpha,4n)$ product were used; each ratio was multiplied by the appropriate $(\alpha,3n)$ or $(\alpha,4n)$ product thick target range in bismuth to give the range of $(\alpha,2n)$ recoils in bismuth. The results are in good agreement with the thick target range data. The recoil ranges of the products of the compound-nucleus reactions $Bi^{209}(\alpha,3n)$ and $Bi^{209}(\alpha,4n)$ plotted as a function of the incident particle energy establish the range-energy relation which is needed in order to calculate the energies of the $(\alpha,2n)$ recoils. The range-energy relation is seen from Fig. 7, to be quite linear. The errors indicated in Fig. 7 are due to the $\pm3\%$ fluctuations in target thickness, which was mainly responsible for the observed scatter of the points.

The range measurements for both Cm^{244} and Bi^{210} targets show that the $(\alpha,2n)$ reactions involve preferential emission of neutrons in the forward direction, but it is not possible to decide unambiguously whether only one, or both, neutrons are involved. Comparison of the experimental and calculated angular distributions of At^{211} leads to the same conclusion. Attempts to analyze the results by the use of average values for the recoil angles and momenta fail, probably because recoils having average momentum are not often emitted at the average angle. The surprising result is that the assumptions of the simple compound nucleus model begin to break down at energies as low as 30 Mev, when the cross sections for the $(\alpha,2n)$ reactions are near their maximum values. An unambiguous solution of the kinematic problem probably requires the measurement of the momenta of recoils as a function of their angle.

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