Differential Range Study of (α,xn) Reactions of Copper*

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The differential ranges of the $Cu^{63}(\alpha,n)$, $Cu^{65}(\alpha,2n)$, and $Cu^{65}(\alpha,3n)$ reaction products have been measured in the energy range of 14–40 MeV by means of the electrostatic collection technique. The results have been used to derive a range-energy relation for gallium recoils in H_2. Comparison with the Lindhard-Scharff-Schiott calculation indicates that the latter overestimates the importance of electronic stopping. The results have been compared with a Monte Carlo evaporation calculation and have been used to obtain the average values of the neutron and photon energies. The information obtainable from transformation of the recoil velocity distribution between the c.m. and laboratory systems has been explored. These various analyses all indicate that the differential ranges and angular distributions of the recoils yield mutually consistent results.

I. INTRODUCTION

THE determination of the recoil properties of the products of intermediate-energy nuclear reactions can provide valuable information about the nature of these reactions. Average projected range measurements often indicate whether or not a particular reaction involves the formation and subsequent decay of a compound nucleus. Angular-distribution data provide corroborative evidence about the reaction mechanism as well as specific information about the details of the evaporation process in those cases where a compound-nuclear mechanism has been established. Differential-range measurements provide a sensitive probe of the reaction mechanism and also give confirmatory information about the parameters governing the evaporation process. In addition, the results of these experiments can be used to derive a range-energy relation for the recoil products which may then be compared with theoretical predictions.

The present study is one of a series of investigations of the recoil properties of the products of the reactions of Cu^{65} and Cu^{63} with intermediate-energy He^4 and He^3 ions. In previous publications we have reported the results of average range measurements[1,2] and angular-distribution studies[3–5] for various (α,xn), (He^3,xn), $(\alpha,\alpha n)$, and (He^3,α) reactions. We have found evidence for both compound-nuclear and direct-interaction processes and have made comparisons with statistical-theory and distorted-wave calculations. The angular-distribution results for compound-nuclear reactions have been analyzed in detail in terms of the velocity and angular distributions of the reaction products in the c.m. system and have also been used to provide information about the partition of the available energy between the emitted particles and photons.

We report here the results of differential-range measurements for the recoil products of the $Cu^{63}(\alpha,n)$, $Cu^{65}(\alpha,2n)$, and $Cu^{65}(\alpha,3n)$ reactions at bombarding energies of 14–40 MeV. The ranges were determined by the electrostatic collection of the gallium recoils stopping in hydrogen gas, a technique that has been used by a number of workers.[6–11] The experimental details are discussed in Sec. II and the results presented in Sec. III. A range-energy relation for $Ga^{66,67}$ in hydrogen is derived in this section and compared with the calculation of Lindhard, Scharff, and Schiott (LSS).[12] In Sec. IV the differential ranges are compared with a spin-independent form of the statistical theory[13] and the sensitivity of the results to the value of the level-density parameter is explored. The data are also analyzed in terms of a previously developed[3] transformation procedure to yield the parameters governing the distribution of recoil evaporation velocities in the c.m. system. Finally, the average values of the total kinetic energy of the emitted neutrons and photons are determined in the manner first proposed by Simonoff and Alexander.[14] The results of these various analyses are compared with similar results previously obtained[3] for the same reactions from an analysis of angular-distribution data.

II. EXPERIMENTAL

A. Experimental Procedure

We have measured the differential ranges of Ga^{66} produced by the $Cu^{63}(\alpha,n)$ and $Cu^{65}(\alpha,3n)$ reactions and of Ga^{67} formed by the $Cu^{65}(\alpha,2n)$ reaction. The irradiations were performed with the 60-in. cyclotron at Argonne National Laboratory. The irradiation con-

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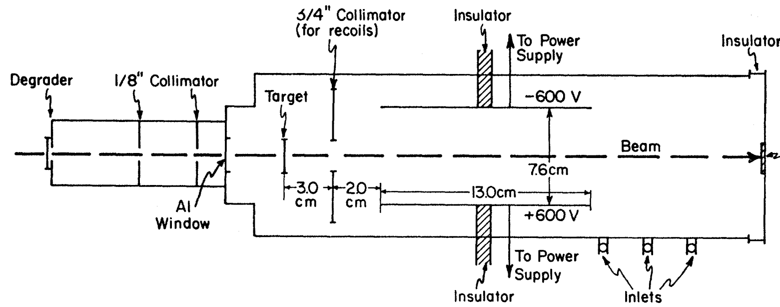


FIG. 1. Schematic diagram of the irradiation chamber.

ditions are best described by reference to Fig. 1, which shows a schematic diagram of the apparatus.

The deflected He^4-ion beam was first degraded to the appropriate energy with aluminum foils, collimated by two $\frac{1}{8}$-in.-diam apertures separated by 1–3 in. from each other, and then allowed to enter the hydrogen-filled irradiation chamber through a 0.002-in.-thick aluminum window. The energy of the incident beam was determined by means of a range-energy relation based on that of Bichsel *et al.*[15] for protons.

The irradiation chamber was of cylindrical shape, 18 in. long and 6 in. in diam. Upon entering the chamber the beam was recollimated by a $\frac{1}{8}$-in. aperture located just upstream from the target foil. The recoil products originating in the target are emitted in a forward cone and are slowed down by collisions with hydrogen molecules. The hydrogen pressure was adjusted so that the recoils stopped within 5–18 cm downstream from the target. Two brass plates, 13-cm long by 8-cm wide, were placed in this stopping region in a direction parallel to that of the beam. The plates were electrically insulated from each other and from the body of the chamber and were maintained at a potential of $\pm$600 V. The distance between the plates was normally 3 in. so that the usual field between the plates was 400 V/in.

Previous investigations using this technique have shown that those recoil products that retain a positive ionic charge at the end of their range will be attracted to the negatively charged plate. The range of the recoil projected along the beam direction will be given by the horizontal distance between the target and the collection point. In order to obtain this information the brass plates were covered with high-purity (99.999%) aluminum foil on which the recoil products were collected. After irradiation the aluminum foils were cut into 13 strips, each of which corresponded to a particular differential-range interval.

In most of the experiments a wide-angle collimator with a $\frac{3}{4}$-in. aperture was placed 3 cm downstream from the target. This collimator prevented recoils emitted at angles greater than 18° to the beam from entering the stopping region. Recoils emitted at these large angles were able to strike the collector plates prior to being stopped and so would have given rise to an erroneous short-range tail on the distribution. Our previous angular distribution measurements[3] indicate that less than 10% of the recoils are emitted at these large angles.

The targets for these experiments consisted of very pure (99.999%) silver foils, 0.0005-in. thick, on which copper had been electrodeposited to a thickness of 10-15 μg/cm^2. In the case of the measurements above 28 MeV, where the $(\alpha,2n)$ and $(\alpha,3n)$ reactions of Cu^{65} were investigated, the targets consisted of highly enriched[16] (99.7%) Cu^{65}. Targets of natural copper were used to measure the differential ranges of the Cu$^{65}(\alpha,2n)$ and Cu$^{63}(\alpha,n)$ reaction products below 26 MeV.

Prior to each irradiation an alignment experiment was performed in order to ensure that the beam passed through the target foil. This was accomplished by irradiating a Mylar foil placed in the target position for a few seconds and noting the position of the beam spot. The position of the target holder in the chamber was then appropriately adjusted. Prior to irradiation the chamber was evacuated and flushed with hydrogen gas a number of times. The chamber was then filled to the desired pressure as determined with a differential manometer. The desired pressures ranged from 40 to 120 Torr depending on the bombarding energy. The pressure was checked at the end of the irradiation and was usually within 2 Torr of the initial value.

The irradiations had a duration of 2–3 h and the beam current was usually maintained at 0.5 μA. The dependence of the differential range on the beam intensity was investigated and the results will be given in Sec. II B. The ion current passing through the collector plates was monitored throughout the irradiation. The current through the negative plate was approximately 20 μA and that through the positive plate about 60 μA, presumably reflecting the ejection of electrons from the target and window foils. The ion current was found to be proportional to the He^4-ion current. Under normal operating conditions the plate voltage was independent of the beam current and there were no electrical discharges in the chamber. Two of the irradiations performed in the course of this study were, for unknown reasons, plagued by numerous discharges. The differential-range curves obtained in these experiments were highly anomalous and were discarded.

Bryde *et al.*[7] have shown that recoils collected on aluminum foils in the fashion described above do not

[15] H. Bichsel, R. Mozley, and W. Aron, Phys. Rev. 105, 1788 (1957).

[16] Obtained from Oak Ridge National Laboratory.

TABLE I. Summary of experimental tests.

E_α (MeV)	Reaction	Plate	Collection efficiency (%)	Target thickness (μg/cm²)	Beam current (μA)	Pressure (Torr)	Field[a] (V/cm)	R_0 (μg/cm²)	ρ
20.1	(α,n)	Neg.	78	11	0.1	73.2	1200/7.62	75.5	0.336
20.1	(α,n)	Neg.	82	15	0.5	78.7	1200/7.62	76.2	0.331
20.1	(α,n)	Pos.	22	11	0.1	73.2	1200/7.62	90.6	0.423
20.1	(α,n)	Pos.	18	15	0.5	78.7	1200/7.62	88.5	0.406
20.1	(α,n)	Neg.		15	0.5	77.4	900/7.62	76.0	0.338
42.4	$(\alpha,2n)$[b]	Neg.		11	0.5	132.0	2400/22.8	130.0	0.400
42.4	$(\alpha,2n)$[c]	Neg.			0.5		1200/7.62	138.0	0.37

[a] Potential difference divided by distance between plates.
[b] These data were obtained with a newly constructed larger chamber.
[c] Values estimated by extrapolation of data obtained at 39.8 MeV.

adhere very strongly to the aluminum. The recoils can be removed from the foil in an irreproducible manner by careless handling. In order to prevent this undesirable recoil loss, the aluminum foil was carefully sprayed with acrylon immediately after irradiation. This procedure had the desired result of materially reducing the danger of recoil loss due to ruboff without, at the same time, affecting the differential range in any noticeable way.

The aluminum foils were then cut into strips as described above and gallium was radiochemically separated from each strip.[17] The production of gallium from direct activation of impurities in the aluminum foil or from impurities in the silver target backing was determined in one experiment and found to be completely negligible.

The activities of 9.5-h Ga^{66} and 78-h Ga^{67} were determined by means of β-proportional counters having a background of 0.5 cpm. The results were corrected for chemical yield and self-absorption. The decay curves were resolved by means of the CLSQ least-squares computer program.[18]

B. Tests of Experimental Procedure

It is not obvious in an experiment of this type that the horizontal distance between the target and the collection point of a recoil is necessarily equal to the projected range. A number of factors can affect this relationship and some of these have been investigated by previous workers.[7–11] Some of the effects that can give rise to incorrect results include scattering of the recoils in the target, formation and subsequent diffusion of neutral recoil atoms, diffusion of recoil ions prior to collection, recoil drifts due to convection currents, and inhomogeneities in the electric field.

The effect of target thickness has been investigated in our previous studies[3,5] of the angular distribution of recoils resulting from the interaction of copper with He^4 and He^3 ions. It was found that the distributions were independent of target thickness for targets of comparable thicknesses to those used in the present study. Since the angular distribution is at least as sensitive to the effect of scattering in the target as the differential range, it may be concluded that this represents a negligible source of error.

The shape of the electric field in an experimental arrangement similar to the present one has been investigated in some detail by Bryde *et al.*[7] These workers found that field inhomogeneities occurred only at the edges of the plates. Distortions in the range curve due to this effect may therefore be avoided by adjusting the hydrogen pressure so that the most probable range corresponds to the midpoint of the collection plates.

The gallium recoils are initially produced with a positive ionic charge. In order for the stopped recoils to be attracted along the lines of force it is necessary that they retain a positive charge. Neutral atoms are more subject to diffusion, and if negatively charged species are formed, they too must first go through a neutral stage. A measure of the formation of neutral atoms may be obtained from a comparison of the relative number of gallium recoils collected at the two plates. The difference in the shapes of the differential-range curves of recoils collected at the two plates gives evidence as to the effect of diffusion on neutral or negatively charged recoils. These results will be presented below.

The effect of diffusion on the width of the differential-range curve has been treated in detail by Alexander *et al.*[8,9,11] These authors point out that this effect depends on the distance the stopped recoils have to drift to reach the collection plates and on the plate voltage. The effect is best investigated by determining the differential range as a function of applied voltage and drift distance. The possibility of convection effects at high-beam currents can be investigated by measuring the range at various beam currents. The results of these tests are presented below.

It has been pointed out[8] that the differential ranges of reaction products formed in heavy-ion-induced compound-nuclear reactions tend to be Gaussian in shape. As will be seen in Sec. III, the same situation is found to hold in the present case. The results of the various experimental tests may therefore be conveniently summarized in terms of the parameters characterizing the Gaussian: the median range R_0 and the range-straggling parameter ρ ($\rho=\sigma/R_0$, where σ is the standard deviation).

[17] N. T. Porile and D. Morrison, Phys. Rev. **116**, 1193 (1959).
[18] J. B. Cumming, U. S. Atomic Energy Commission Report No. NASNS 3107, 1962, p. 25 (unpublished).

TABLE II. Gaussian analysis of differential-range results.

Reaction	E_α (MeV)	E_R (MeV)	Target thickness (μg/cm²)	R_0 (μg/cm²)	ρ	ρ_s	ρ_n	$E_{c.m.}+Q$ (MeV)	T_n (MeV)	T_γ (MeV)
(α,n)	14.1	0.83	10.9	51.2	0.273	0.075	0.262	7.9	3.8	4.1
	18.5	1.09	7.9	66.8	0.258	0.070	0.248	12.0	4.5	7.5
	25.2	1.48	11.0	82.8	0.322	0.064	0.315	18.3	9.9	8.4
	25.7	1.51	13.8	90.8	0.356	0.064	0.346	18.8	12.1	6.7
$(\alpha,2n)$	18.5	1.04	7.9	64.8	0.248	0.071	0.238	2.8	3.7	<0
	25.2	1.42	11.0	82.2	0.272	0.065	0.264	9.1	6.2	2.9
	25.7	1.45	13.8	88.3	0.273	0.065	0.264	9.6	6.3	3.3
	28.6	1.61	9.5	102.8	0.248	0.063	0.240	12.3	5.8	6.5
	28.9	1.63	9.6	99.8	0.266	0.063	0.258	12.6	6.8	5.8
	34.8	1.96	10.0	109.2	0.306	0.059	0.300	18.2	11.1	7.1
	39.8	2.24	12.6	118.0	0.342	0.056	0.337	22.9	16.0	6.9
$(\alpha,3n)$	28.6	1.58	9.5	102.0	0.203	0.063	0.193	1.6	3.7	<0
	28.9	1.60	9.6	95.6	0.222	0.063	0.213	1.9	4.6	<0
	34.8	1.93	10.0	110.2	0.242	0.059	0.234	7.4	6.6	0.8
	39.8	2.21	12.6	120.5	0.258	0.057	0.252	12.2	8.8	3.4

The results of the various experimental tests are summarized in Table I. The first two rows describe the effect of beam intensity on the differential range of the (α,n) reaction product at 20.1 MeV. It is seen that an increase in current from 0.1 to 0.5 μA has no discernible effect on either the median range or the width of the distribution. Evidently convection effects are of little significance at the indicated beam currents. It is also seen that approximately 80% of the activity collected at the two plates is found on the negative plate. Although the total number of Ga^{66} recoils produced in the irradiation was not determined directly, it can be estimated from the known cross section[17] that the activity collected at the two plates represents at least 90% of the total activity. We conclude from these facts that over 70% of the gallium recoils retain a positive ionic charge at the end of their range and are therefore attracted to the negative plate.

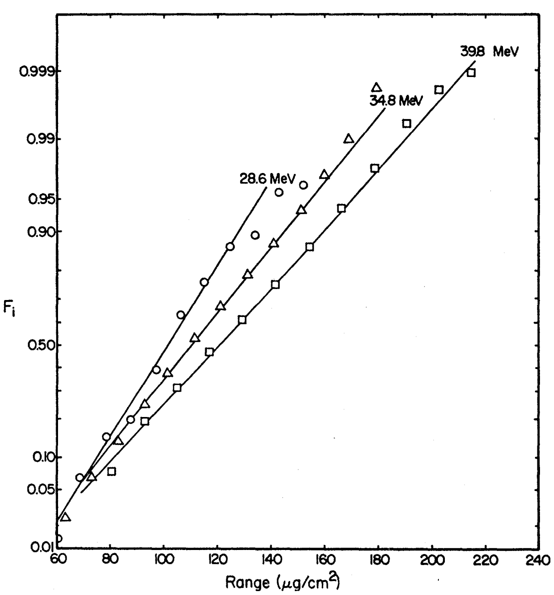


FIG. 2. Probability plot of the differential ranges of the $(\alpha,3n)$ reaction products. F_i is the fraction of the total activity found in the ith strip.

The effect of neutralization of the ionic charge on the differential ranges is summarized in rows 3 and 4 of Table I. These are the results for the (α,n) reaction at 20.1 MeV obtained from recoils collected at the positive plate. It is seen that both R_0 and ρ are significantly larger than the comparable values obtained from the negative plate. We conclude that neutralization and the possible subsequent formation of negative ions do indeed perturb the differential range. Insofar as neutralized recoil products are also collected on the negative plate, an error will be introduced in the results. In view of the magnitude of the difference between the two types of distributions and the relative number of recoils collected at the two plates the errors in ρ and R_0 will be less than 5%. In fact, the actual errors are likely to be substantially smaller than this value since the activity on the positive plate includes the contribution from negatively charged as well as neutral recoils.

The effect of the plate voltage may be seen from a comparison of the results given in rows 2 and 5. A reduction in the potential difference between the plates from 1200 to 900 V at a constant distance of 3 in. (7.6 cm) has no effect on the differential range.

The effect of the distance between the plates is shown in columns 6 and 7. It is seen that an increase in the distance between the plates from 3 to 9 in. (7.6 to 23 cm) leads to a slight increase in width and decrease in median range, both effects being less than 10%. It should be stated, however, that the interpretation of this result is somewhat uncertain for a number of reasons. First, as the distance was increased from 3 to 9 in. there was a concomitant decrease in field strength from 158 to 105 V/cm. We do not believe that this is a serious complication because it has already been seen that the results are independent of field strength in the range of 118–158 V/cm. Of greater importance are the facts that the results for the 1200-V potential difference had to be extrapolated from 39.8 to 42.4 MeV. Also, the results for the 2400-V potential difference were obtained with a different chamber in which all recoils, including those emitted at angles larger than 18°, were allowed to enter the collection region. These various differences in conditions make this particular comparison less secure, although it may still be concluded that the differential ranges are essentially independent of plate distance under the conditions of our experiment.

In summary, we would like to conclude that, after extensive testing of the experimental method, conditions have been found under which the differential ranges appear to be independent of the irradiation parameters and therefore reflect the intrinsic distribution of recoil ranges. The present findings are in the main consistent with the results of previous investigations,[7–9,11] although there are significant differences associated with the identity of the recoil product. The results presented in Sec. III were all obtained under the same conditions: The potential difference was 1200 V, the distance between the plates was 3 in., and the beam intensity was 0.5 μA. Differential ranges were only determined for recoils collected at the negative plate.

III. RESULTS

A. Differential Ranges

The results of our experiments are summarized in Table II in terms of the previously defined Gaussian parameters. The latter were obtained from a probability plot of the fraction of the total activity collected up to a given distance from the target versus distance. An example of this type of plot is illustrated in Fig. 2, which shows the data for the $Cu^{65}(\alpha,3n)$ reaction. The median range R_0 is obtained as the distance at which 50% of the activity has been accumulated and the straggling parameter ρ is determined by the slope of the line. The measured recoil distances d were converted to the more convenient superficial density units $\mu g/cm^2$ by means of the equation

$$R_0(\mu g/cm^2) = d(cm) \times 89.88 \times (273/T) \times P/760, \quad (1)$$

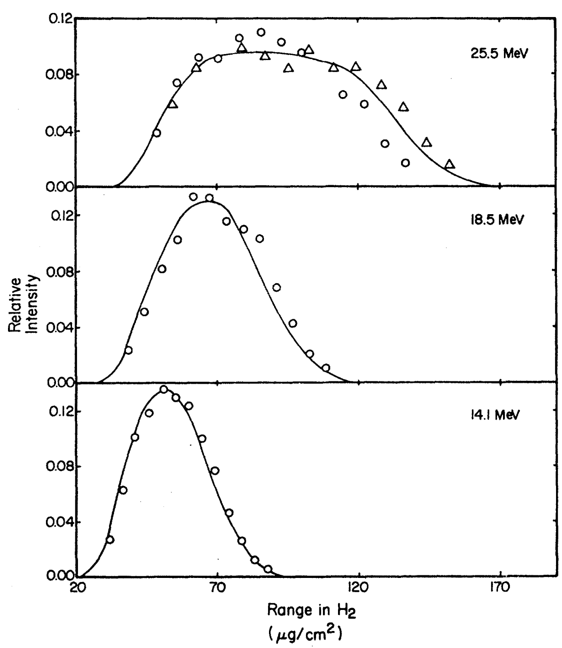


FIG. 3. Differential ranges of the $Cu^{63}(\alpha,n)$ reaction product. The solid curves are the result of the Monte Carlo evaporation calculation described in Sec. IV A. The two sets of points (O,Δ) refer to two separate experiments at essentially the same bombarding energy.

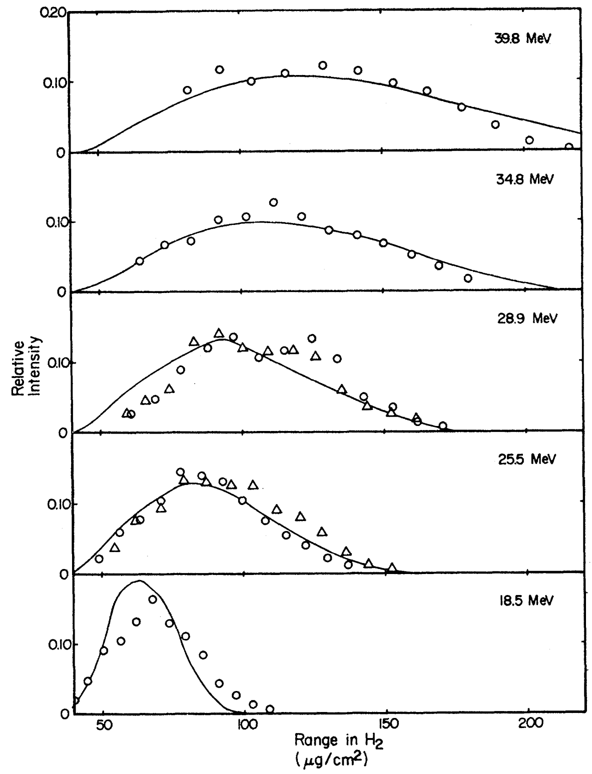


FIG. 4. Differential ranges of the $Cu^{65}(\alpha,2n)$ reaction product. See Fig. 3 for details.

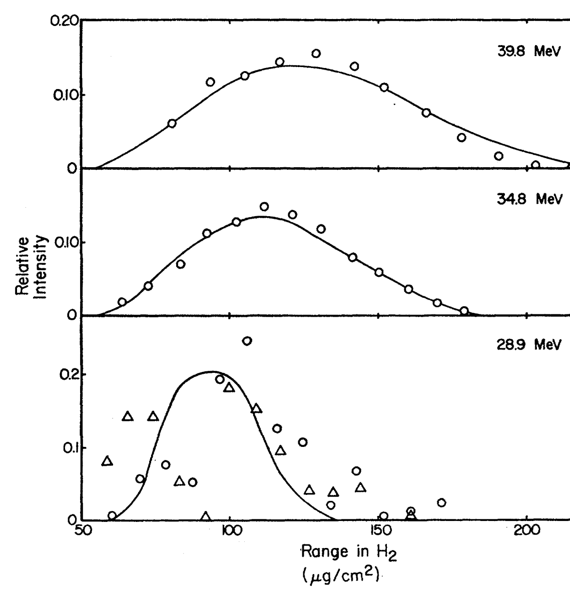
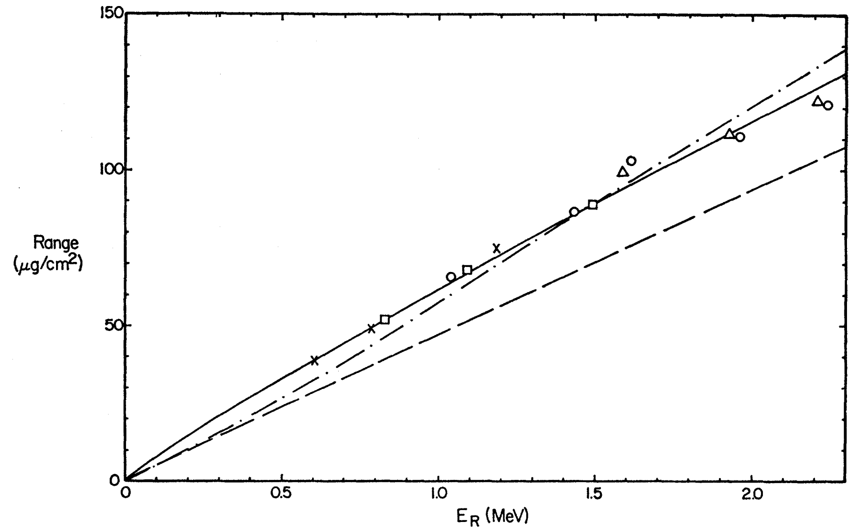


FIG. 5. Differential ranges of the $Cu^{65}(\alpha,3n)$ reaction product. See Fig. 3 for details.

FIG. 6. Range-energy relation for $Ga^{66,67}$ in H_2. The experimental points refer to the following reactions: △, $(\alpha,3n)$; ○, $(\alpha,2n)$; □, (α,n); and ×, (α,n) from Bryde *et al.* (Ref. 7). Solid curve is a least-squares fit to data; dashed curve is the LSS calculation for $k=0.132$; dot-dashed curve is the LSS calculation for $k=0.07$.



where T is the absolute temperature of the hydrogen gas and P is the pressure in Torr. The above equation is based on the reasonable assumption of ideal gas behavior of hydrogen over the narrow temperature range of 0–25°C and for pressures up to 1 atm.

The results shown in Fig. 2 and summarized in Table II have been corrected for energy loss in the target by adding half the hydrogen equivalent of the target thickness to the measured distances. The relative stopping powers of hydrogen and copper for gallium recoils were obtained from the LSS calculation.[12] This correction only amounted to about 1.5 $\mu g/cm^2$ of H_2. The plots shown in Fig. 2, as well as similar plots for the other reactions, confirm that the differential ranges are approximately Gaussian in shape.

The differential ranges of the (α,n), $(\alpha,2n)$, and $(\alpha,3n)$ products are plotted in Figs. 3–5, respectively, for the various bombarding energies. The actual data may be obtained from the authors on request. It can be seen both from these plots and from the corresponding Gaussian parameters that both the recoil range and the width of the distribution increase with bombarding energy. The first of these trends is a consequence of the proportionality of recoil energy and bombarding energy for compound-nuclear reactions and the second, of the increasing energy of the evaporated neutrons relative to that of the incident α particle.

An estimate of the random errors in the results may be obtained from the agreement between experiments performed at essentially the same bombarding energy. It is seen in Table II that there are several such cases. On the average, the standard deviations in duplicate R_0 and ρ values are about 4%. These uncertainties are consistent with the random errors associated with the activity measurements, chemical-yield determinations, and temperature and pressure determinations.

B. Range-Energy Relation

The measured range-straggling parameter is principally made up of contributions from the evaporation of nucleons ρ_n and from the inherent straggling in the stopping process ρ_s. If both these contributions are treated as Gaussian, then the relation between them is[14]

$$\rho^2=\rho_n^2+\rho_s^2. \qquad (2)$$

In order to proceed with the interpretation of the results it is necessary to extract the value of ρ_n from the measured value of ρ. The values of ρ_s may be obtained from the LSS theory.[12] The calculated values depend on the relative importance of nuclear and electronic stopping. Although the theory predicts the relative contribution from these two processes via the value of the electronic stopping parameter k, the absolute calculation of ρ_s has been shown to disagree with experiment.[9] It seems more reasonable to determine an empirical value of k by comparison of the experimental range-energy relation with LSS and use this value to determine ρ_s. We turn, accordingly, to a discussion of the range-energy relation for gallium recoils in hydrogen.

The recoil energy E_R may be obtained from the energy of the incident helium ion E_b by the relation

$$E_R=E_bA_bA_R/(A_b+A_T)^2, \qquad (3)$$

where A_b, A_R, A_T refer to the masses of the bombarding particle, recoil product, and target, respectively. The values of E_R are summarized in Table II. The measured ranges are the projections of the actual ranges along the beam direction. Although the compound nuclei are initially moving parallel to the beam, the evaporation of particles introduces a small transverse component in the motion of the recoil products. The true range R is therefore related to the measured range by $R=R_0/\langle\cos\theta_L\rangle$, where θ_L is the laboratory recoil angle. Since the angular distributions of the recoil products formed in the reactions of present interest have been measured,[3] the correction can be made. It amounts to approximately 1.5%.

The experimental range-energy relation, corrected for the above effect, is shown in Fig. 6. We include the data obtained by Bryde *et al.*[7] for the $Cu^{63}(\alpha,n)$ reaction,

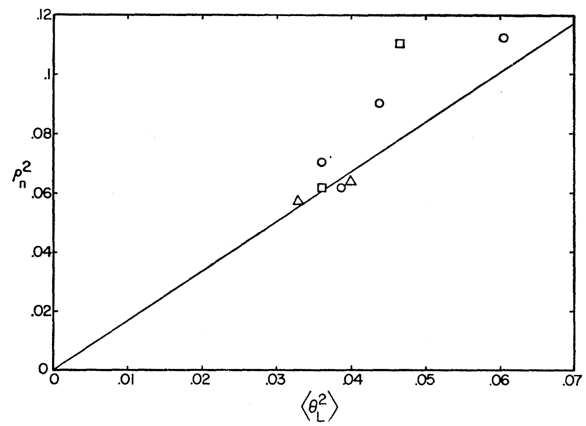


FIG. 7. Dependence of ρ_n^2 on $\langle\theta_L^2\rangle$. The experimental points refer to the following reactions: △, $(\alpha,3n)$; ○, $(\alpha,2n)$; □, (α,n). The solid line is obtained from Eq. (6).

Their results are seen to be in excellent agreement with our values. If it is assumed that the range-energy relation is of the form $R_{(\mu g/cm^2)}=bE^{N/2}{}_{(\mathrm{MeV})}$, the values of the constants b and N may be determined by a least-squares fit to the data. The results are $b=61.70\pm1.01$ μg/cm^2 and $N=1.816\pm0.048$. The solid curve drawn through the points in Fig. 6 is the result of this fit.

The range-energy curve predicted by the LSS theory[12] is given by the dashed line in Fig. 6. This curve has been obtained with a value of the electronic stopping parameter $k=0.132$, which is appropriate to the particular combination of recoil and stopping atoms of present interest. The LSS calculation predicts the values of the recoil-path length in terms of the reduced-range variable ρ. The latter was converted to the reduced range projected along the direction of motion of the recoil in the manner prescribed by LSS and the range was then obtained by the use of the appropriate ρ-R conversion factor. The ranges obtained in this fashion have to be corrected for the effect of evaporation. The isotropic emission of nucleons increases the recoil range by the factor[19] $[1+\frac{1}{6}(N^2+N-2)(V/v_{\mathrm{CN}})^2]$, where V and v_{CN} are, respectively, the c.m. velocity that the product nucleus acquires as a result of evaporation and the velocity of the compound nucleus, and N is the previously defined exponent in the range-energy relation. The value of $(V/v_{\mathrm{CN}})^2$ is in turn related[14] to the experimentally determined value of ρ_n so that the correction can be made in terms of quantities determined exclusively in the present experiment. Since it turns out that ρ_n is nearly equal to ρ, the correction can be made without difficulty in spite of the fact that ρ_n cannot be determined until the corrected theoretical range-energy relation has been obtained. The ratio of the corrected theoretical range to that calculated in the absence of evaporation is obtained in terms of the experimentally determined value of N as

$$R_{\mathrm{corr}}/R=1+0.47\rho_n^2. \tag{4}$$

[19] L. Winsberg and J. M. Alexander, Phys. Rev. **121**, 518 (1961).

The correction amounts to approximately 3% and the LSS ranges shown in Fig. 6 have been increased by this much.

It is seen that the LSS ranges are significantly smaller than the experimental values. One possible interpretation of this discrepancy is that the calculation overestimates the importance of electronic stopping for 1–2-MeV recoils. Adopting this point of view, it is then possible to determine the value of k that gives the best agreement with the experimental curve. It is found that the LSS curve for $k=0.07$ gives the best fit to the data, as shown by the dot-dashed curve in Fig. 6. Even in this case it appears that the calculated curve has a greater slope than the experimental one, suggesting that electronic stopping has a somewhat stronger energy dependence than predicted by the theory. Similar conclusions have been drawn by Gilat and Alexander[9] for dysprosium ions stopping in a number of gases, although their results for hydrogen were in reasonably good agreement with the theory.

It seems reasonable to assume that the LSS theory is generally satisfactory provided that the parameter k is adjusted to fit the range data. Proceeding on this basis, it is then possible to obtain from LSS the value of the straggling parameter ρ_s for the empirically determined value of k. The values of ρ_s obtained in this fashion are summarized in Table II. The desired values of the nuclear-straggling parameter ρ_n are obtained from Eq. (2) and are listed in Table II. It is seen that most of the range straggling is due to the effect of evaporation, while the stopping process contributes very little to the width of the differential range. As pointed out by Alexander *et al.*,[8] it is precisely this fact that allows information about the evaporation process to be extracted from differential ranges in hydrogen.

We are now in a position to perform one additional test of the experimental method. Simonoff and Alexander[14] have shown that for a given nuclear reaction both ρ_n^2 and $\langle\theta_L^2\rangle$, the mean-square recoil angle, are directly proportional to $\langle V^2\rangle/v_{\mathrm{CN}}^2$ provided that the angular distribution of the recoils is isotropic in the c.m. system. Specifically, the desired relations are

$$\rho_n^2=N^2\langle V^2\rangle/3v_{\mathrm{CN}}^2 \tag{5}$$

and

$$\langle\theta_L^2\rangle=2\langle V^2\rangle/3v_{\mathrm{CN}}^2. \tag{6}$$

It follows that

$$\rho_n^2=N^2\langle\theta_L^2\rangle/2. \tag{7}$$

Our previous angular-distribution results[3] were shown to be consistent with isotropic evaporation and, moreover, provided values of $\langle\theta_L^2\rangle$ for the reactions of present interest. We can accordingly calculate values of ρ_n^2 by means of Eq. (7) and the experimentally determined values of N and $\langle\theta_L^2\rangle$ and compare them with the values given in Table II. If the experimental values of ρ have a contribution from instrumental effects, this should be reflected in values of ρ_n^2 larger than those obtained from Eq. (7). Figure 7 shows a plot of ρ_n^2

versus $\langle\theta_L^2\rangle$. It is seen that many of the experimental points do indeed lie above the line obtained by means of Eq. (7), suggesting that there may be some instrumental broadening of the differential ranges that was not apparent from the tests of the technique. Alternatively, the values of ρ_s may be larger than predicted by LSS, although previous measurements[9] make it unlikely that the theory can be wrong by the required factor of 2, particularly in view of the adjustment in the value of k. Note, however, that the smallest values of ρ_n^2, which should be most sensitive to broadening, are in good agreement with Eq. (7). We are therefore unable to draw a firm conclusion from this analysis, although the possibility of instrumental broadening or underestimation of ρ_s values should be kept in mind in the discussion that follows.

IV. DISCUSSION

A. Comparison with Monte Carlo Evaporation Calculations

The differential ranges have been compared with a spin-independent version of the statistical theory. The calculation was based on the code of Dostrovsky *et al.*[13] as modified by Porile[3,20] to keep track of the momentum of the residual nucleus.

In order to apply this calculation to the evaluation of differential ranges it was necessary to introduce several modifications into the code. The momentum and kinetic energy of the residual nucleus were determined in the manner described before.[3,20] Following particle evaporation, the kinetic energy of the product was converted to its range in hydrogen by means of the experimentally determined range-energy relation (Fig. 6). The range R_0 was then corrected for straggling on the assumption that this process led to a Gaussian dispersion in range along the direction of motion of the recoil product. The probability of obtaining a range R for a given value of R_0 was thus evaluated by the equation

$$P(R)=\exp[-(R-R_0)^2/2\rho_s^2R_0^2], \qquad (8)$$

using the experimentally determined values of ρ_s given in Table II. Values of R were obtained from Eq. (8) by the choice of two random numbers. The values of R were then projected along the beam direction for comparison with experiment.

The calculation was performed at the energies for which experimental results were available. The calculation was programmed for the Purdue 7094 computer and 10 000–20 000 iterations were performed at each energy for a given value of the level-density parameter. Because experimental results were only available for (α,xn) reactions, the calculation only considered the emission of neutrons. This restriction materially speeded up the computation without introducing any distortion in the results.

[20] N. T. Porile and S. Tanaka, Phys. Rev. 135, B122 (1964).

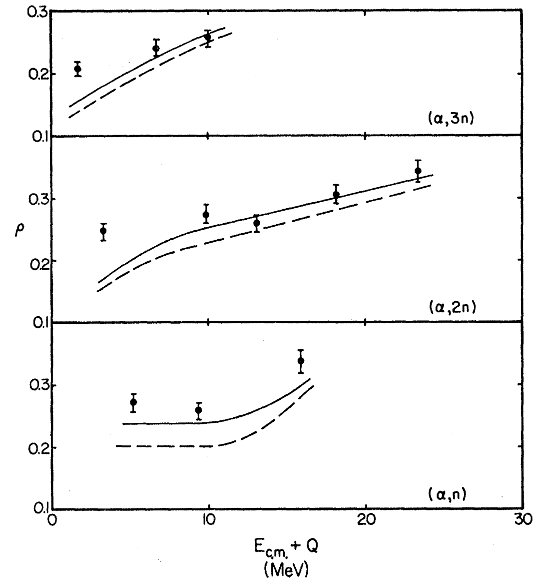


FIG. 8. Dependence of the width parameter ρ on the available energy. The curves are the results of a Monte Carlo calculation; —— $a=A/30$, - - - $a=A/10$, and the experimental points are also shown.

Since differential-range measurements have not heretofore been compared with the statistical theory, it seemed of interest to determine the sensitivity of the results to the assumed value of the level-density parameter. Although the calculation is based on the use of a highly approximate level-density formula, $\Omega(E)=C\exp[2(aE)^{1/2}]$, it is still of interest to determine the effect of this parameter on the results. We have accordingly calculated the differential ranges for $a=A/10$, $a=A/20$, and $a=A/30$, where A is the mass number of the residual nucleus resulting from a particular evaporation step.

The results of the calculation are summarized in Fig. 8. This is a plot of the calculated value of the straggling parameter ρ versus the energy available for neutron emission, $E_{\text{c.m.}}+Q$. Curves are included for $a=A/10$ and $a=A/30$. It is seen at a given bombarding energy that the width of the differential curve increases as a decreases. This dependence is a consequence of the inverse relation between the level-density parameter and the neutron kinetic energy, and the proportionality between the latter and the width of the differential range. Note, however, that the calculated width parameter is not particularly sensitive to the value of a. A factor of 3 variation in a thus affects the value of ρ by only about 10–20%. In addition, the calculated mean range is completely insensitive to the level-density parameter. It may thus be concluded that differential-range measurements are not particularly useful for obtaining information about the level-density parameter, unless the experimental uncertainty in ρ can be reduced to the 1% level.

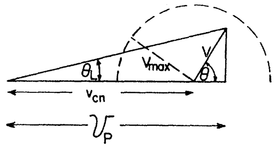


FIG. 9. Velocity diagram for transformation between c.m. and laboratory systems.

The experimental values of ρ are also included in Fig. 8. It is seen that, of the various curves under consideration, the one for $a=A/30$ gives the best agreement with experiment. Some of the data actually suggest an even smaller value of a but this is ruled out by the angular distributions[3] and excitations functions[13,17] of these same reactions. The calculated differential ranges obtained with $a=A/30$ are shown as the solid curves in Figs. 3–5. The curves have been normalized in area to the experimental data. The over-all agreement in the shape of the curves is seen to be very good.

B. Transformational Analysis of Differential Ranges

In a previous study[3] a procedure was developed that permitted the determination of the distribution of recoil-evaporation velocities in the c.m. system from the angular distribution of the recoil product. This procedure was based on the assumption of a Gaussian distribution of the velocity of the recoil product in the c.m. system. The angular distribution in this system was transformed to the laboratory for comparison with experiment. The best fit between experiment and calculation, as determined by a χ^2 test, then served to fix the parameters governing the Gaussian velocity distribution. The more straightforward analysis, in which the laboratory angular distribution is directly transformed to the c.m. system, could not be performed because of the double-valued nature of the transformation.

In this subsection a similar analysis of the differential range is developed. A somewhat similar analysis of angular distribution and differential-range data based on a Maxwellian distribution of evaporation velocities has been recently reported by Ewart and Kaplan.[21] We refer to Fig. 9, which summarizes the vector relationships of interest. The projection of the laboratory velocity along the beam direction is denoted by $\mathcal{V}_p$, and V and v_{CN} have already been defined. Let θ be the recoil angle in the c.m. system and θ_L that in the laboratory system. These various quantities are related by the expression

$$\cos\theta=(\mathcal{V}_p-v_{\text{CN}})/V. \tag{9}$$

The maximum value of V is determined by the kinematics of the reaction and is given by

$$V_{\max}=\frac{[8n(E_{\text{c.m.}}+Q)]^{1/2}}{A_b+A_T+A_R}, \tag{10}$$

where A_b, A_T, and A_R are the masses of the bombarding particle, target, and recoil nucleus, respectively, and there are n nucleons emitted in the reaction. The range of allowed $\mathcal{V}_p$ values obviously has to obey the relation

$$(v_{\text{CN}}-V_{\max})\leq\mathcal{V}_p\leq v_{\text{CN}}+V_{\max}, \tag{11}$$

and v_{CN} is always larger than $V_{\max}$ for the reactions of present interest.

A given value of $\mathcal{V}_p$ can result from a combination of various values of V and θ and the desired distribution is obtained by integrating over the distributions of V and θ:

$$P(\mathcal{V}_p)=2\pi\int_{V_{\min}}^{V_{\max}}\int_{\theta_{\min}}^{\theta_{\max}}P(V)W(\theta)\sin\theta d\theta dV \times\delta\left[\theta=\cos^{-1}\left(\frac{\mathcal{V}_p-v_{\text{CN}}}{V}\right)\right], \tag{12}$$

where the distribution of evaporation velocities is, as before,[3] assumed to be Gaussian and characterized by a most probable velocity V_0 and a width parameter C:

$$P(V)=[V_0C(2\pi)^{1/2}]^{-1}\times\exp[-(V-V_0)^2/2(V_0C)^2]. \tag{13}$$

The angular distribution of recoil products in the c.m. system has been shown[3] to be isotropic for the reactions of present interest, so that $W(\theta)=1$. The δ function in Eq. (12) shows that there is a contribution to $P(\mathcal{V}_p)$ only when Eq. (9) is satisfied. Keeping this condition in mind, Eq. (12) can be rewritten as

$$P(\mathcal{V}_p)=\frac{(2\pi)^{1/2}}{V_0C}\int_{|\mathcal{V}_p-v_{\text{CN}}|}^{V_{\max}}\left[\frac{V^2-(\mathcal{V}_p-v_{\text{CN}})^2}{V^2}\right]^{1/2}\times\exp\left(-\frac{(V-V_0)^2}{2(V_0C)^2}\right)dV. \tag{14}$$

The evaluation of Eq. (14) was performed by numerical integration on the 7094 computer. A solution accurate to within 1% was obtained by computing the integrand at intervals of $V_{\max}/200$. The distribution of projected velocities was usually evaluated for 20 equally spaced values of $\mathcal{V}_p$. This distribution was then directly compared with the experimental distribution of projected velocities, $P(\mathcal{V}_p)_{\text{expt}}$. The agreement between the two distributions was determined by means of a X^2 test, where

$$X^2=\sum\frac{[P(\mathcal{V}_p)_{\text{calc}}-P(\mathcal{V}_p)_{\text{expt}}]^2}{P(\mathcal{V}_p)_{\text{calc}}}. \tag{15}$$

The experimental distribution of projected velocities was obtained from the measured projected differential range in the following manner. As shown in Sec. III A, the distribution of projected ranges can be approximated by a Gaussian. It is now assumed that the projected range is related to the projected velocity by the experimental range-energy relation derived in Sec.

[21] A. Ewart and M. Kaplan, Phys. Rev. 162, 944 (1967).

III B. This assumption introduces a negligible error ($\sim$1%) since the difference between the actual and the projected ranges was shown to be only 1.5%. The Gaussian distribution of projected ranges, corrected for straggling, then transforms to the following distribution of projected velocities:

$$P(\mathcal{V}_p)_{\text{expt}}d\mathcal{V}_p=[R_0\rho_n(2\pi)^{1/2}]^{-1} \times\exp-\left[\frac{b'\mathcal{V}_p{}^N-R_0}{\sqrt{2}R_0\rho_n}\right]^2 Nb'\mathcal{V}_p{}^{N-1}d\mathcal{V}_p, \quad (16)$$

where $N=1.816$, as given by the empirical range-energy relation, and the constant b' is related to the proportionality constant b in this relation by $b'=(A_R/2)^{N/2}b$.

The comparison between experiment and calculation for a particular reaction at a given bombarding energy is thus performed by evaluating Eq. (16) with the experimental values of R_0, ρ_n, N, and b'. Equation (14) is evaluated for particular values of the velocity-distribution parameters V_0 and C. The values of the latter are varied in a systematic manner and the value of χ^2 is determined by means of Eq. (15). The best values of V_0 and C then are those giving the smallest value of χ^2.

A typical result of this analysis is shown in Fig. 10. This is a plot of χ^2 versus C for various values of V_0 obtained from the comparison with the differential range of the (α,$3n$) reaction product at 39.8 MeV. It is seen that, for a broad range of V_0 values, there is a particular value of C which results in a minimum χ^2. Unfortunately, a number of the curves have minima

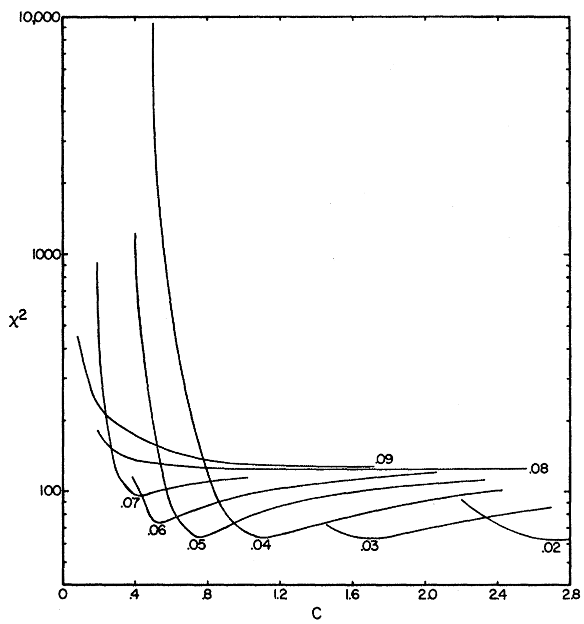


FIG. 10. Results of the parameter search for the (α,$3n$) reaction at 39.8 MeV. The calculated values of χ^2 are plotted as a function of C for different values of V_0.

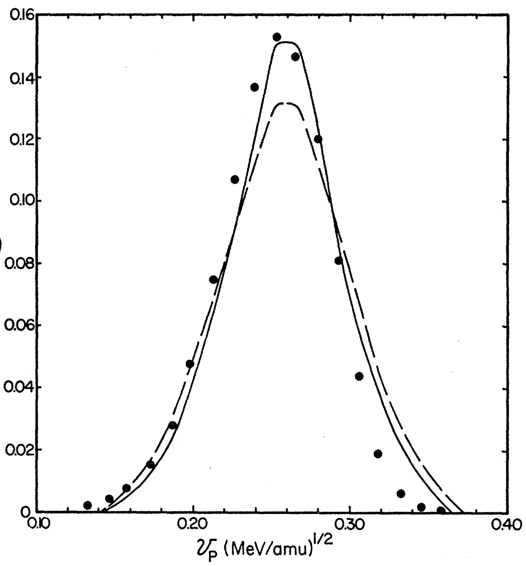


FIG. 11. Comparison of experimental and calculated projected velocity distributions of the (α,$3n$) reaction product at 39.8 MeV. The points are obtained from the measured differential range. Solid curve is one of the best-fitting calculations, $V_0=0.04$ and $C=1.1$; dashed curve is obtained with $V_0=0.06$ and $C=0.7$, the parameters giving the best fit to the angular distribution.

yielding values of χ^2 that are within 1% of each other and so result in equally good fits to the data. For instance, it is impossible to choose between the combinations $V_0=0.02$, $C=2.7$ and $V_0=0.05$, $C=0.75$. While the analysis thus restricts the choice of Gaussian evaporation parameters, it does not unambiguously define them. This conclusion is in marked contrast with the results of a similar transformational analysis of the angular distribution of the reaction product.[3] It was found in that analysis that the χ^2 test was much more sensitive and defined a unique combination of V_0 and C. For instance, the values $V_0=0.06$ (MeV/amu)$^{1/2}$ and $C=0.7$ constituted the best choice for the (α,$3n$) reaction at 39.8 MeV. The present results are somewhat inconsistent with this value, as shown by the comparison of the experimental and calculated distributions of the projected velocity given in Fig. 11. The curve obtained with these parameters gives a poorer fit to the data than one of the curves yielding the smallest value of χ^2, e.g., $V_0=0.04$, $C=1.1$. This discrepancy is not surprising in view of the 5–10% uncertainties in both types of experiments. It may therefore be concluded that the same distribution of recoil velocities can account for both the angular distribution and the differential range. The latter is, however, less sensitive to the values of the two parameters characterizing the distribution.

C. Average Neutron and Photon Energies

The average total kinetic energy in the c.m. system of the emitted neutrons, T_n, and the average total energy of the emitted photons, T_γ, may be obtained

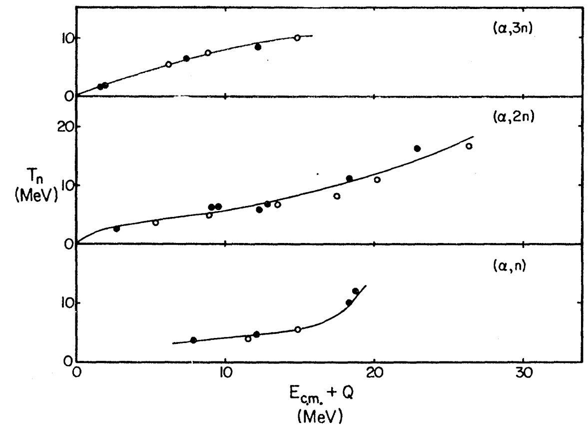


FIG. 12. Energy dependence of T_n. ●, obtained from differential-range data; ○, obtained from angular-distribution data (Ref. 3). In those cases where the analysis gives $T_n > E_{c.m.}+Q$, the points have been plotted at $T_n = E_{c.m.}+Q$.

from the differential ranges in the manner proposed by Simonoff and Alexander.[14] If the angular distribution of recoils in the c.m. system is isotropic, T_n is related to ρ_n by the expression

$$T_n = \frac{3E_b A_b (A_b + A_T + A_R)^2 \rho_n^2}{4N^2 (A_b + A_T)^2}, \tag{17}$$

where E_b is the bombarding energy. The values of T_γ are obtained from the equation

$$T_\gamma = E_{c.m.} + Q - T_n. \tag{18}$$

The values of $E_{c.m.}+Q$, T_n, and T_γ are summarized in Table II. The dependence of T_n on the available energy is shown in Fig. 12. It is seen that in all cases the values of T_n increase with the energy available for neutron emission. The values of T_γ also show an initial increase but appear to level off at a value of 7–8 MeV, indicating that γ-ray emission does not compete with neutron evaporation in the bombarding energy range of interest. It may be noted in Table II that in a few instances negative values of T_γ are obtained at the lowest energies. These values naturally have no physical significance but are merely a reflection of the experimental uncertainties.

We have previously[3] obtained the values of T_n and T_γ for these same reactions from the angular distribution of the recoil products. Figure 12 includes the values of T_n derived from the angular distributions. Note that these values were actually obtained on the basis of a somewhat more accurate formulation[22] than that used in the present analysis. Because of this difference the angular-distribution points should be lowered by about 4% to make them strictly comparable to the present values. It is seen that the two sets of points are in good agreement in all cases, confirming the assumption that $W(\theta)=1$.

[22] M. Kaplan and V. Subrahmanyam, Phys. Rev. **153**, 1186 (1967).

V. CONCLUSIONS

The differential ranges of the (α,xn) reactions of Cu^{63} and Cu^{65} have been compared with a spin-independent Monte Carlo evaporation calculation, used to derive the distribution of recoil velocities in the c.m. system, and analyzed to yield the values of T_n and T_γ. The Monte Carlo calculation is in satisfactory agreement with the data for a value of the level-density parameter $a = A/30$, both with respect to the energy dependence of the differential ranges and to the values of the straggling parameter. The comparison points out that the differential ranges are only slightly sensitive to the value of the level-density parameter.

A procedure has been developed whereby an assumed velocity distribution in the c.m. system is transformed to yield a laboratory distribution that may be compared with experiment. The results of this comparison determine the values of the Gaussian parameters V_0 and C characterizing the velocity distribution. It is found that while the analysis restricts the values of V_0 and C to certain permissible combinations, it does not define them uniquely. This is in contrast to the angular distribution of the recoil products, where a similar analysis does permit a specific choice of V_0 and C.

The values of T_n and T_γ obtained from the ranges were compared with similar values previously derived from the angular distributions and found to be in good agreement with the latter. This agreement confirms the assumption that the angular distribution of the recoils in the c.m. system is isotropic. The magnitude of the T_γ values indicates that γ-ray emission does not compete with neutron evaporation in the de-excitation of compound nuclei produced by 40-MeV α particles.

The differential-range studies yield information about the evaporation process that is, in general, less accurate than similar information derived from angular-distribution data. This follows from the fact that the measured widths have to be corrected for straggling and also from the need for an accurate range-energy relation. The present study has shown that the two experiments yield mutually consistent results.

The differential ranges were used to derive a range-energy relation for gallium in hydrogen. Comparison with the theoretical LSS relation indicates that the theory overestimates the importance of electronic stopping for recoils of 0.5–2 MeV.

ACKNOWLEDGMENTS

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