Excitation Functions for Alpha-Induced Reactions on Zinc-64†

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Eleven excitation functions for alpha-induced reactions on Zn^{64} have been measured up to incident energies of 41 Mev. The values of $\sigma(\alpha,p)/\sigma(\alpha,n)$ and $\sigma(\alpha,pn)/\sigma(\alpha,2n)$ in the region of maximum yield were found to be 1.7 and 9, respectively. Reactions involving alpha-particle emission account for about 20% of the total inelastic cross section at 40 Mev. The total inelastic cross section was found to agree with calculated values for $r_0=1.6\times10^{-13}$ cm. The competition between different reactions was analyzed in terms of the statistical theory by use of a level density expression of the form $W(E)=C\exp\{2[a(E-\delta)]^{\frac{1}{2}}\}$. Values of a ranging from 0.8 to 2.8 were required to fit the experimental results, indicating that the statistical theory is not completely applicable.

INTRODUCTION

NUCLEAR reactions induced by particles with incident energies less than 50 Mev have been the subject of considerable investigation in recent years. Two experimental approaches have been used in these studies. The first consists of the measurement of energy spectra and angular distributions of emitted particles[1–7]; the second, of the determination of excitation functions for these reactions.[8–19] One of the main purposes of these investigations has been to study the applicability of the statistical theory[20] to reactions in this energy region. According to the theory, the emission of different particles is assumed to proceed by successive evaporation from a compound nucleus,[21] and the shapes of the energy spectra of emitted particles, as well as the cross sections for different reactions, are determined in large measure by the value of the level density parameter, a. The latter may be related to the level density, $W(E)$, of the residual nucleus resulting from a particular nuclear reaction, by the familiar expression obtained from the Fermi gas model,[20] $W(E)=C\exp 2(aE)^{\frac{1}{2}}$. According to the model, a is proportional to the mass number of the residual nucleus in question. This prediction has been borne out by the previously mentioned experiments with moderate success at the very best. While the values of a obtained from several measurements of energy spectra do indeed exhibit the expected increase with mass number, other such measured spectra, as well as all excitation functions measured to date, appear to be consistent with $a\sim2$, regardless of mass number. The situation has been summarized by Igo and Wegner.[22] They point out that the low values of a may be partly due to the effect of direct-interaction processes. The latter lead to the emission of an excessive number of high-energy particles, which in turn result in low values of a that have no meaning in terms of the nuclear model. Ample evidence for direct-interaction processes has indeed been found in measurements of the angular distribution of emitted particles. Whereas an evaporation mechanism requires that particles be emitted with symmetry about 90°,[23] several experiments[1,2] have shown a large excess of particles emitted in the forward direction. Igo and Wegner[22] conclude, however, that even when only energy spectra of particles emitted in the backward hemisphere are studied, thereby minimizing the contribution of direct-interaction processes, anomalously low values of a are still obtained in several instances.

In view of these inconsistencies it seemed desirable to continue the study of low-energy nuclear reactions. Further excitation-function measurements appeared to be of interest since many of the previously mentioned studies were rather fragmentary. The excitation functions for the reactions of zinc-64 with alpha particles ranging in energy from 13 to 41 Mev were chosen for investigation in the present work since it is possible to measure eleven excitation functions, accounting for over 80% of the total inelastic cross section. The results were analyzed in terms of the statistical theory and values of a were obtained from the measured cross sections. In addition, it was possible to study the competition between neutron, proton, and alpha emission and to observe in several instances the effect of the

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TABLE I. Measured cross sections in millibarns for reactions of zinc-64 with alpha particles.

Reaction / Threshold (Mev) / E_α (Mev)	(α,γ)	(α,n) 9.4	(α,p) 4.5	$(\alpha,2n)$ 20.3	(α,pn) 16.2	$(\alpha,3n)$ 31.8	$(\alpha,p2n)$ 6.0	$(\alpha,2pn)$ 21.7	$(\alpha,\alpha n)$ 14.9	$(\alpha,\alpha 2n)$ 22.0	$(\alpha,\alpha pn)$ 19.4
10.5	0.2										
13.4	0.7	53.5	151								
15.1		112	320								
16.5	0.9										
16.9		215	450								
18.5		295	515								
18.9					1.0						
20.2		289	505								
20.7					19.0						
21.0									2.0		
21.8				0.02							
22.8									13.0		
23.0		180	403								
23.3				0.17	223						
25.4									48.2		0.04
25.7		82.5	246								
26.0								1.0			
27.2							0.9	4.5			
27.6											0.24
27.9									88.1		
28.3		51.0	128	27.4	577						
28.9							4.5				0.59
30.2				60.5	724				134	0.15	
31.0											2.0
31.4		22.2	63.2	79.3	753						
31.8									150	0.8	
32.4				85.2	742		25.3	70.0			
34.0						0.3					
34.4											9.5
34.7				82.2	698			170	159	3.5	
36.2							84.2				
36.6		7.0	20.0								
36.8								320			
37.5				73.3	562	4.3					
37.7									153	10.6	37.5
39.1							97.0	542			
39.6		3.5	9.0	60.4	440						
40.5									150	22.1	95.0
40.8						14.0		670			

even-even or odd-odd nature of the residual nucleus on the measured cross sections.

EXPERIMENTAL

The irradiations were performed with the deflected alpha-particle beam of the Brookhaven 60-inch cyclotron. A detailed description of the target assembly and Faraday cup used to monitor the beam intensity is given elsewhere.[24] The beam intensity varied between 0.2 and 1.0 microamperes. Irradiation times varied between 10 seconds and 6 hours. The initial alpha-particle energy was 41 Mev. The variation in the energy of the incident beam for different bombardments was monitored by determining with a beta proportional counter the gross activity of $Ga^{66}+Cu^{64}$ and $Ga^{67}+Zn^{65}$ induced in copper foils by alpha particles degraded to either 39 Mev or 25 Mev. The ratio of the production cross sections of $Ga^{66}+Cu^{64}$ and $Ga^{67}+Zn^{65}$ in this energy region is very sensitive to small changes in the energy of the incident beam.[25] In general, the incident energy varied by less than 0.3 Mev from run to run. In order to perform experiments at bombarding energies below 41 Mev, the beam was degraded in energy by use of aluminum absorbers. The curves of Aron *et al.*[26] were used to determine the energy of the degraded beam. The stacked-foil technique was used to irradiate between one and eight target foils in any one experiment. A total of 52 irradiations was performed in the course of this study.

The targets consisted of high-purity 0.0005-inch natural-zinc foils, or of zinc, enriched to 93% in Zn^{64},[27] plated on thin gold foils. The natural-zinc foils were used when there was no interference from the other zinc isotopes and when the beam energy was high enough so that the energy loss in the target foils was less than 1.2 Mev. In all other cases thin (1–2 mg/cm^2) targets of enriched Zn^{64} were used. The target foils were however thick enough to make the loss of recoils negligible. After irradiation the target foils were dis-

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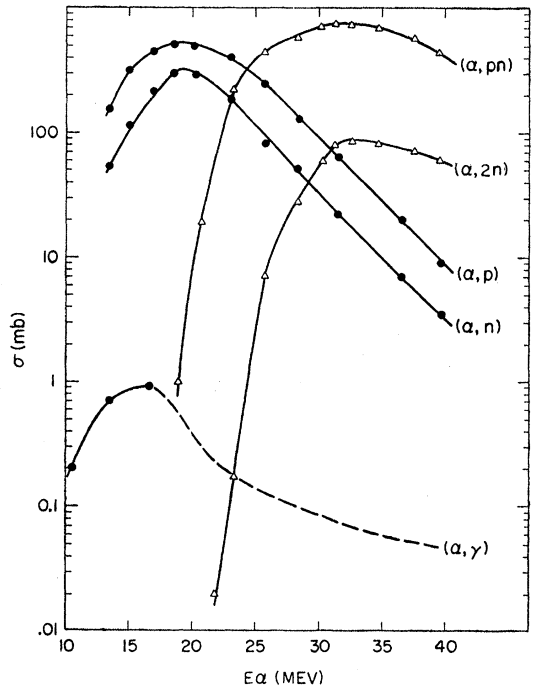

FIG. 1. Measured excitation functions for reactions involving emission of one or two nucleons and for radiative capture.

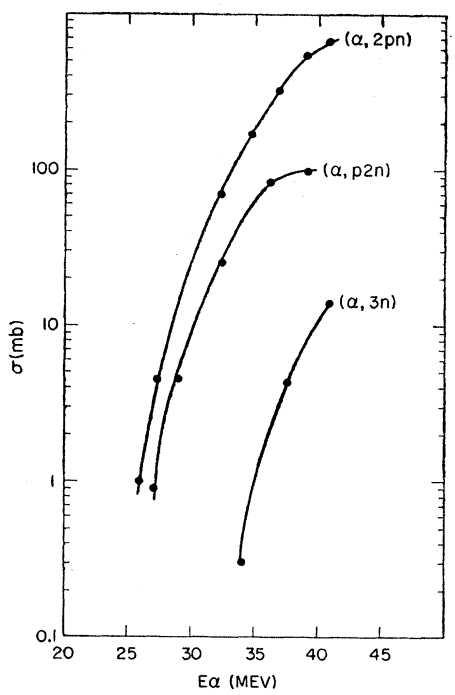

FIG. 2. Measured excitation functions for reactions involving emission of three nucleons.

solved in acid in the presence of carrier, and separation of the desired elements was carried out. The chemical yield was determined either by weighing the final samples or by subsequent spectrophotometric or polarographic determination. The disintegration rates of the samples were determined with beta proportional counters, 3-inch NaI scintillation counters, or deep-well NaI scintillation counters. In many cases the scintillation counters were connected to a 100-channel pulse-height analyzer and the decay of a particular photopeak was followed. Since only energetic beta emitters were counted on beta-proportional counters, no self-scattering or self-absorption corrections were applied in the comparison of different samples. In most cases, cross-section measurements were repeated and agreement to within 3 to 5% was obtained. The chemical procedures, counting techniques, and calibration methods used are presented in more detail in the Appendix.

RESULTS

A total of 80 cross sections was measured in this study. These are presented in Table I, together with the thresholds for the corresponding reactions. The latter were obtained from the masses of stable nuclides listed by Wapstra[28] coupled with the latest decay energy measurements.[29] The errors in the listed cross-section values are estimated to be of the order of 10%. This value is based on the results of duplicate experiments, which agreed to within 3 to 5%, and on an estimated error of 5 to 10% in the determination of counting efficiencies. The points on a given excitation function have a relative error of less than 5% with respect to each other. This is also the case for points on different excitation functions involving genetically related nuclides provided that advantage was taken of this relationship in the determination of counting efficiencies. The listed bombarding energies are most accurate for values close to the energy of the undegraded beam. The energy values below 15 Mev may be in error by over 1 Mev due to the magnification by the straggling process of small errors in the assumed value of the incident energy.

The excitation functions are plotted in Figs. 1–3. The (α,γ) excitation function was not measured above 17 Mev due to the interfering production of Ge^{68} by the $(\alpha,2n)$ reaction on Zn^{66}. The excitation function above this energy was estimated on the assumption that it has the same shape as the excitation function for the (α,γ) reaction on Ni^{58}.[30] The maximum cross section is of the same magnitude as the maximum cross sections measured for other alpha-induced radiative capture reactions.[30,31] The relative yield of the (α,p) and (α,n) reactions indicates that proton emission from excited Ge^{68} is about twice as probable as neutron emission. The (α,p) to (α,n) cross-section ratio has

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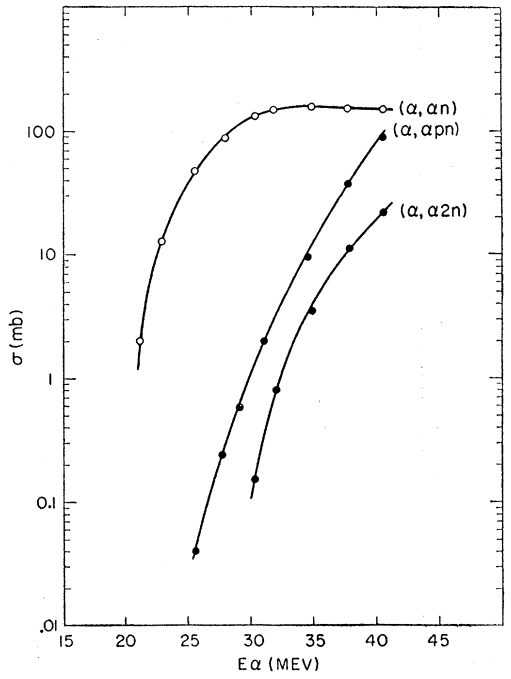

FIG. 3. Measured excitation functions for reactions involving alpha-particle emission.

also been measured for Fe^{54} and found to be about 3.[9] It is thus apparent that proton emission competes favorably with neutron emission in this mass region, at least in the case of neutron-deficient target nuclides. The ratio of the (α,pn) and $(\alpha,2n)$ cross sections in the region of maximum yield is about 9. The ratio of (α,pn) to $(\alpha,2n)$ cross sections has also been measured in a number of other cases in this mass region. A ratio of 60 was reported for Fe^{54},[9] 26 for Cr^{50},[14] 4 for Ni^{60},[18] 30 for Ti^{46},[32] and 2 for Ge^{70}.[33] While the (α,pn) reaction is in many instances favored by having a lower threshold than the $(\alpha,2n)$ reaction, the main effect appears to be due to differences in the level densities of the residual nuclei. The products of all the $(\alpha,2n)$ reactions mentioned are even-even nuclides while the corresponding products for the (α,pn) reactions are odd-odd nuclides and are thus expected to have a greater density of levels at low excitation energies.[34] In addition, several of the $(\alpha,2n)$ products occur at a closed shell or have an isotopic number of zero and these factors perhaps further reduce their yield. Some confirmation for this view is obtained from the large $(\alpha,2n)$ cross sections measured for V^{50} targets[35] and Cu^{65} targets,[36] where the resulting nuclides are not even-even.

The greater probability for proton rather than neutron emission is also demonstrated in the emission of three nucleons as witnessed by the fact that the ratio of cross sections for the $(\alpha,2pn)$ and $(\alpha,p2n)$ reactions at 40 Mev is approximately 6. It is of course possible that these cross sections include substantial contributions from triton and He^3 emission. An estimate of the triton contribution may be obtained from recent measurements of tritium yields in alpha-induced reactions for a number of target nuclei including zinc.[37] Comparison with the $(\alpha,p2n)$ cross sections reported in the present work is difficult since only the integrated yield of tritons produced by alpha particles ranging in energy from 48 Mev to the threshold of the (α,t) reaction was measured. A very rough comparison may be made by extrapolating the $(\alpha,p2n)$ excitation function of Fig. 2 to 48 Mev, and estimating the fraction of observed tritons in Gonzalez-Vidal's work that are actually emitted together with another particle and thus do not result in an (α,t) reaction. On this basis it is estimated that, even if the triton production observed by Gonzalez-Vidal is ascribed entirely to reactions with Zn^{64}, only about 10% of the Ga^{65} observed in the present experiments can result from (α,t) reactions.

The excitation functions for reactions involving alpha-particle emission are shown in Fig. 3. It is seen that substantial yields are found for these reactions. Similar results have been obtained recently for the corresponding reactions of Ni^{58} and alpha particles.[38] The $(\alpha,\alpha pn)$ and $(\alpha,\alpha 2n)$ excitation functions are rather similar in shape and relative yield to the (α,pn) and $(\alpha,2n)$ reactions on Ni^{60}.[18] The latter lead to the same residual nuclides as the reactions in question. The value of $\sigma(\alpha,pn)/\sigma(\alpha,2n)$ for Ni^{60} thus is about 4 to 5 for

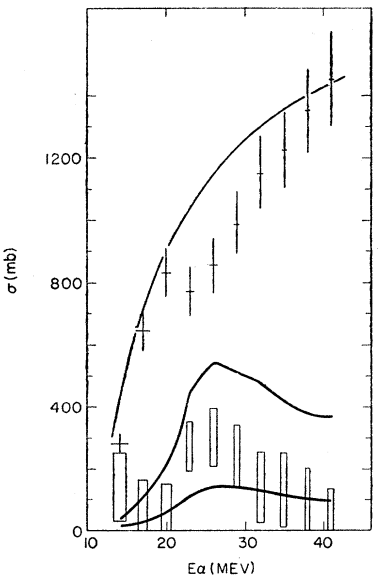

FIG. 4. Experimental values of the total inelastic cross section with their associated errors, and continuum-theory values for $r_0 = 1.6$ fermis; the vertical bars give the difference between the calculated and experimental values; the two heavy curves represent extreme values for the excitation functions of unmeasured reactions as estimated by the statistical theory.

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[37] J. Gonzalez-Vidal, University of California Radiation Laboratory Report UCRL-8330 (unpublished).
[38] Blann, Thomas, and Seaborg, Abstract of the American Chemical Society San Francisco meeting, 1958; F. Houck and J. M. Miller (private communication).

incident energies a few Mev above the corresponding thresholds, while the value of $\sigma(\alpha,\alpha pn)/\sigma(\alpha,\alpha 2n)$ obtained in the present work over the corresponding energy region is about 4. The $(\alpha,\alpha pn)$ and $(\alpha,\alpha 2n)$ excitation functions are shifted to higher energies relative to the corresponding excitation functions for the Ni^{60} target by some 10 to 15 Mev, reflecting in large measure the kinetic energy of the emitted alpha particles.

The experimental excitation functions may be summed in order to estimate the magnitude of the total inelastic cross section. The value obtained in this fashion constitutes a lower limit since there are a number of reactions leading to stable nuclides which were not investigated. The total inelastic cross section may be calculated according to continuum theory,[39] and is shown in Fig. 4 for a nuclear radius parameter, r_0, of 1.6 fermis (1.6×10^{-13} cm). The corresponding experimental values are plotted together with their estimated uncertainties. The experimental values are in general smaller than the calculated values, particularly between 22 and 32 Mev where the former fall short by as much as 300 millibarns from the continuum-theory values. The vertical bars represent the difference between the calculated and experimental values and thus constitute a rough excitation function for the unmeasured reactions. It is possible to estimate values for the cross sections of the unmeasured reactions by use of the statistical theory, as shown in the following section. The excitation functions estimated in this fashion are given in Fig. 5. The cross sections for these reactions are probably good to within a factor of two. The maximum and minimum values of the cross sections for all unmeasured reactions, obtained by assuming that the estimated cross sections may be in error by a factor of two, are given by the two heavy lines in Fig. 4. It is seen that the estimated cross sections overlap at all energies with those obtained from the difference between the experimental and calculated values for the total inelastic cross section. If r_0 is taken as 1.5 or 1.7 fermis, the fit obtained in this comparison is considerably poorer than for $r_0=1.6$ fermis. The two most significant unmeasured excitation functions are seen to be those for the $(\alpha,\alpha\gamma)$ and $(\alpha,2p)$ reactions.

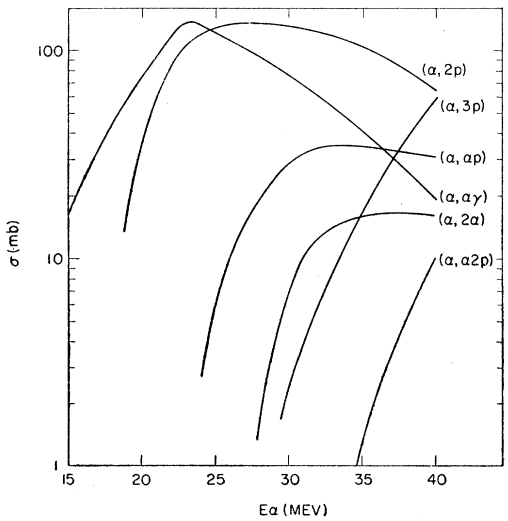

FIG. 5. Excitation functions for unmeasured reactions as estimated by use of the statistical theory.

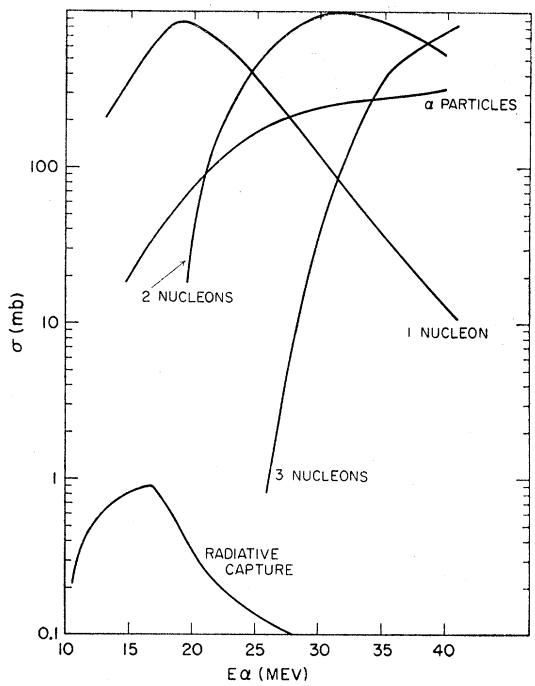

FIG. 6. Summary of measured and estimated excitation functions.

At the lowest bombarding energies used in this study, reactions involving the emission of at most one nucleon are the only ones that are observed. As the energy of the incident alpha particles is increased, reactions involving successively the emission of two and three nucleons become energetically possible and compete with each other as well as with all other reactions. This competition is pictured in Fig. 6 where the excitation functions for the emission of one, two, or three nucleons are plotted. Both measured and estimated excitation functions are included and the occurrence of (α,d) or of (α,t) and (α,He^3) reactions is assumed to be equivalent to the emission of two or three nucleons, respectively. Measured and estimated values for reactions involving alpha-particle emission are given as a separate curve regardless of the total number of particles emitted. It is seen in Fig. 6 that a particular set of reactions goes through a maximum as the thresholds of the reactions involving emission of one more nucleon are passed. It can be seen that reactions involving the emission of alpha particles constitute a substantial fraction of the total inelastic cross section ranging from an estimated 5% at 15 Mev, to about 20% at 40 Mev.

[39] M. M. Shapiro, Phys. Rev. **90**, 171 (1953).

DISCUSSION

The experimental results may be compared with values predicted by the statistical theory.[20] According to the theory, the cross section for an (α,x) reaction is given by

$$\sigma(\alpha,x)=\sigma_c(\alpha)f_x/\sum_j F_j,$$

where $\sigma_c(\alpha)$ is the cross section for formation of the compound nucleus by an alpha particle, f_x is the emission function for particle x over a particular energy interval, and F_j is the emission function for particle j over the total energy interval. In this calculation F_j and f_x have been evaluated for protons, neutrons, and alpha particles. The emission functions for deuterons, tritons, and He^3 were found to be about an order of magnitude smaller and were consequently neglected. The emission functions are given by

$$\left.\begin{matrix} f_x \\ F_x \end{matrix}\right\} = 2M_x(2I_x+1)\int_{\epsilon_x^{\min}}^{\epsilon_x^{\max}} \epsilon_x\sigma_{cx}(\epsilon_x)W(\epsilon_x)d\epsilon_x,$$

where M_x and ϵ_x are the reduced mass and kinetic energy in the center-of-mass system and I_x is the spin of particle x; $\sigma_{cx}(\epsilon_x)$ is the inverse cross section for formation of the compound nucleus by particle x with energy ϵ, and $W(\epsilon)$ is the level density of the residual nucleus. The level density of the residual nucleus was assumed to be of the form

$$W(\epsilon_x)=c\exp\{[4a(E^*-S_x-\delta-\epsilon_x)]^{\frac{1}{2}}\},$$

where c is a constant assumed to be the same for all residual nuclei under consideration; a is the level density parameter; E^* is the initial energy of excitation of the compound nucleus; S_x is the binding energy of x. This expression differs from the level density expression obtained from the Fermi gas model by the introduction of the energy term δ in the exponent. The latter is introduced in order to account for the greater density of levels near the ground state of an odd-odd nucleus.[34] In the present calculation the effect of a closed shell on the level density was also included in the parameter δ. δ thus is zero for an odd-odd nucleus for which there are no shell effects, and is greatest for an even-even nucleus at a closed shell. The values of δ for odd-mass nuclei lie between those for even-even and odd-odd nuclei. The inverse cross section was taken as

$$\sigma_{cx}(\epsilon)=\pi R^2\left(1-\frac{K_xB}{\epsilon_x}\right),$$

where $R=r_0A^{\frac{1}{3}}$ if x is a nucleon and $R=r_0A^{\frac{1}{3}}+1.21\times10^{-13}$ if x is an alpha particle. B is the Coulomb barrier for protons, and K_x is a constant taken as $K_n=0$, $K_p=0.7$, and $K_\alpha=1.66$.[40] The advantage of the above approximation for the inverse cross section is that its use permits an integration in closed form. The integration was performed between the following limits:

$$\epsilon_x^{\max}=E^*-S_x-\delta;$$

$$\epsilon_x^{\min}=K_xB \text{ for evaluation of } F_x;$$

$$\epsilon_x^{\min}=E^*-S_x-(S_l+K_lB) \quad \text{if} \quad K_xB\leq E^*-S_x-(S_l+K_lB);$$

$$\epsilon_x^{\min}=K_xB \quad \text{if} \quad K_xB>E^*-S_x-(S_l+K_lB) \quad \text{in the evaluation of } f_x.$$

Here l is the second particle to be evaporated and its identity is determined by the smallest value of S_l+K_lB. The only difference between f_x and F_x thus is that the former refers to the emission of only one particle, while the latter refers to the total emission of particles of type x.

The same emission functions are applicable for the case of multiple particle emission except for the fact that the initial nucleus no longer has a unique excitation energy E^* but a spectrum of excitation energies U. f_x is then calculated for each value of U for which secondary emission is energetically possible and the result is weighted by the probability, $P(U)$, of forming the starting nucleus for the evaporation process under discussion with excitation energy U. In order to obtain the desired cross section, the weighted emission function is summed over all allowed values of U. The cross section for a reaction involving the emission of two particles, $\sigma(\alpha,xy)$, is thus given by

$$\sigma(\alpha,xy)=\sigma_c(\alpha)\sum_U F(U)f_y(U)/\sum_k F_k(U),$$

where $F_k(U)$ now is the total emission probability for all particles k for an initial excitation energy U. $P(U)$ is given by

$$P(U)=\frac{2M_x(2I_x+1)}{\sum_j F_j}\int_{E^*-S_x-(U+\Delta U)}^{E^*-S_x-U}\epsilon_x\sigma_{cx}(\epsilon x)W(\epsilon_x)d\epsilon_x,$$

if x is the first evaporated particle. F_j is the total emission function defined previously. ΔU was in general taken as 1 Mev. In those cases where there are two different evaporation paths leading to the same residual nucleus, the total emission function was obtained from the addition of the emission functions for the two separate reactions. Thus in the case of the (α,pn) reaction it was found that approximately equal contributions were obtained from reactions involving the initial evaporation of a neutron and a proton.

The emission functions for reactions involving the evaporation of three nucleons were readily obtained by subtraction of the emission functions for only one nucleon, and for one nucleon followed by a second nucleon or an alpha particle, from the total emission functions for protons and neutrons, F_p+F_n. The validity of this procedure hinges on the fact that there is no appreciable competition from reactions involving

[40] K. J. LeCouteur, Proc. Phys. Soc. (London) **A63**, 259 (1950).

the emission of four particles for the bombarding energies under consideration.

It is also possible to calculate an emission function for photons, for comparison with the (α,γ) cross sections. The expression for f_γ, given in the same units as f_x, is[41]

$$f_\gamma=\frac{2I_\gamma+1}{c^2}\int_{\epsilon_\gamma{}^{\min}}^{\epsilon_\gamma{}^{\max}}\epsilon_\gamma{}^2\sigma_{c\gamma}(\epsilon_\gamma)W(\epsilon_\gamma)d\epsilon_\gamma,$$

where the inverse cross section is obtained from the magnetic dipole and electric quadrupole absorption cross sections as given by calculated values.[20] It is not clear whether the contribution from electric dipole absorption should be included in a calculation of the cross section for the inverse reaction since the characteristic dipole oscillation which occurs in the absorption process may not take place during emission. In any case, the contribution of the electric dipole cross section to the total absorption cross section is small for photons below 16 Mev, and any error in neglecting this contribution is small.

The calculation is expected to be most accurate for bombarding energies well above the threshold of a particular reaction. The assumption that the inverse cross section for charged particles is zero for incident energies below KB and the consequence that charged particles are not evaporated with energies less than KB only become reasonable when kinetic energies much larger than KB are possible. In addition, the inverse cross section for neutrons is energy dependent and is larger than πR^2 for low-energy neutrons. Another source of error near threshold lies in the assumption that there are no excited levels in a residual nucleus for excitation energies less than δ. The over-all effect of the approximations in the calculation is that the values of a necessary to fit the data are somewhat too small. This follows, for instance, from the fact that low-energy neutrons are emitted with somewhat greater probability than would follow from an inverse cross section of πR^2, thereby enhancing the probability of multiple particle emission for a given bombarding energy.

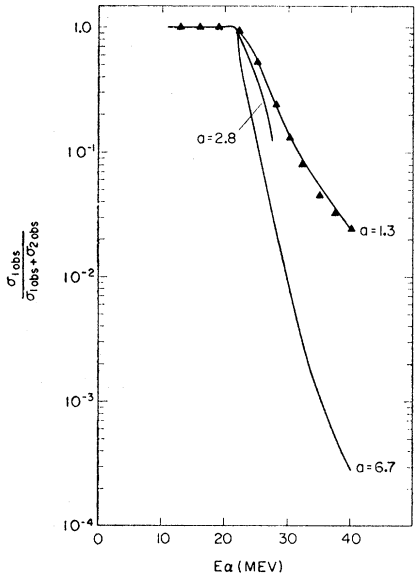

FIG. 7. Competition between reactions involving emission of one and two nucleons. Ordinate: $[\sigma(\alpha,p)+\sigma(\alpha,n)]/[\sigma(\alpha,p)+\sigma(\alpha,n)+\sigma(\alpha,pn)+\sigma(\alpha,2n)]$. The triangles are taken from the experimental curves; the solid curves are calculated values for $a=1.3$, 2.8, and 6.7.

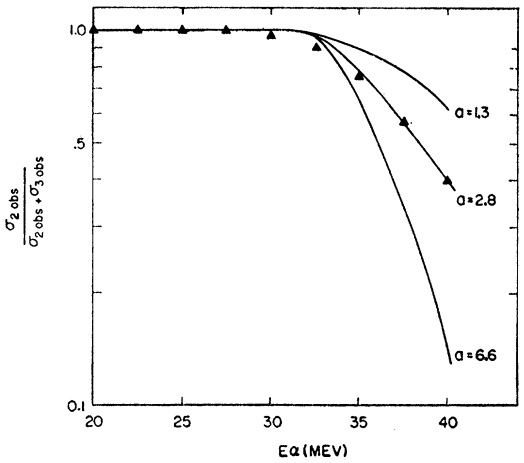

FIG. 8. Competition between reactions involving emission of two and three nucleons. Ordinate: $[\sigma(\alpha,pn)+\sigma(\alpha,2n)]/[\sigma(\alpha,pn)+\sigma(\alpha,2n)+\sigma(\alpha,3n)+\sigma(\alpha,2np)+\sigma(\alpha,n2p)]$. The triangles are taken from the experimental curves; the solid curves are calculated values for $a=1.3$, 2.8, and 6.6.

The values of a and δ used in the calculations were left as adjustable parameters to be determined by comparison with the experimental results. It was found that the calculated ratio for the emission of one or two nucleons, or of two or three nucleons, depended very strongly on the value of a, but was rather insensitive to the values of δ. On the other hand, the ratio of (α,p) to (α,n) cross sections, or of (α,pn) to $(\alpha,2n)$ cross sections, varied strongly with δ, but had a much weaker dependence on a. The two parameters could thus be determined almost independently. As a first approximation, the shell and pairing energies given by Cameron[42] were used as the values of δ. These were then adjusted until a fit with the experimental results was obtained.

The comparison of experimental and calculated values is given in Figs. 7–9. The ratio of measured cross sections for reactions involving the emission of one or two nucleons agrees with calculated values for $a=1.3\pm0.5$ over the entire energy range. The quoted error applies at 25 Mev and becomes smaller at higher energies. The corresponding ratio for reactions involving the emission of two or three nucleons agrees with calcu-

[41] D. E. Fisher, Oak Ridge National Laboratory Report ORNL-2535, 1958 (unpublished).

[42] A. G. W. Cameron, Atomic Energy of Canada United Report AECL-443, 1957 (unpublished).

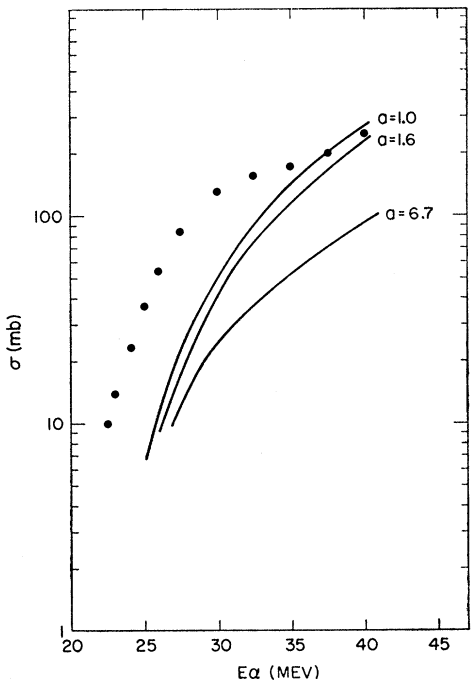

FIG. 9. Comparison of measured and calculated cross sections for reactions involving alpha-particle emission; ●—points from the experimental curves; solid lines—calculated excitation functions for $a=1.0$, 1.6, and 6.7.

lated values for $a=2.8\pm0.3$ over the entire energy range. The experimental cross sections at 40 Mev for reactions involving alpha-particle emission agree with calculated values for $a=1.3\pm0.3$. As the bombarding energy is lowered, the values of a required to fit the experimental data become considerably smaller. The value of a obtained from a comparison with the calculations of the (α,γ) and $(\alpha,n)+(\alpha,p)$ cross sections at 13–16 Mev is 0.8 ± 0.3. In view of the assumptions and approximations made in the calculations, the above values of a are only approximately correct, tending to be somewhat on the low side.

These results are rather disturbing from the viewpoint of the statistical model. The products of the reactions studied in the present work cover a range of only a few mass numbers and the value of a determined from these reactions should therefore be essentially the same in all cases. Furthermore, the calculated values of a are considerably lower than $A/10$, and in some cases significantly lower than $A/20$. The Fermi gas model, on the other hand, indicates that a should be approximately equal to $A/10$ or $A/20$, depending on the details of the nuclear model. This discrepancy may be explained if it is assumed that some of the reactions in question proceed predominantly by a direct-interaction mechanism. In this case the values of a calculated for these reactions lose their meaning and it becomes quite possible to calculate different values of a for different reactions. Several excitation functions for reactions proceeding predominantly by a direct-interaction mechanism have been measured in the heavy-element region.[8,10] These excitation functions in general exhibit an initial rise followed in many cases by either a broad plateau or a broad peak. The sharp peaking that is characteristic of reactions proceeding by an evaporation mechanism is not present in these cases since the emitted particles usually have high energies and the residual nuclei are thus less susceptible to further particle emission. An examination of the excitation functions determined in the present work shows that the $(\alpha,\alpha n)$ reaction has an excitation function with a shape similar to those of reactions proceeding by a direct interaction mechanism. This behavior may then account for the poor fit of the calculated excitation functions for reactions involving alpha-particle emission for any given value of a. The excitation functions for the (α,p) and (α,n) reactions show a fairly sharp peaking. If it is assumed that these reactions proceed exclusively by a direct interaction mechanism at 40 Mev and that this mechanism leads to constant cross sections over the energy range under consideration, then it follows that a direct-interaction process can account for only a few percent of the (α,p) and (α,n) reactions below 30 Mev. It is consequently very surprising that the same value of a can account for these reactions over the entire energy range.

It is of interest to consider the evidence of energy-spectra measurements in this connection. The two experiments that may be compared to some extent with the present work are the (α,α')[1] and (α,p)[2] energy spectra for bombardment of copper with 40-Mev alpha particles. The angular distribution measurements in the (α,α') experiment indicate that over 90% of the alpha particles are emitted in the forward direction and are thus mostly due to direct-interaction processes. The energy spectrum of the outgoing alpha particles is very flat, supporting the interpretation in terms of this mechanism. These results are consistent with the interpretation of the $(\alpha,\alpha n)$ reaction in terms of a direct interaction mechanism. The energy spectra of protons emitted at 150° were measured in the (α,p) experiment and found to be consistent with $a=1.4$. Since the contribution of direct-interaction processes should be minimized at such large angles relative to the incoming beam, it may well be the case that very low values of a cannot be blamed exclusively on the occurrence of such processes but may instead reflect a partial excitation of the nucleus.

The experimental and calculated values for the ratio of cross sections for the (α,p) and (α,n) reactions, and for the (α,pn) and $(\alpha,2n)$ reactions, are shown in Fig. 10. The calculated values are very sensitive to $\Delta\delta$, the difference in the values of δ for the residual nuclei under comparison. Thus in the case of the (α,p) and (α,n) reactions agreement in the region of maximum yield was obtained for $\Delta\delta=0.8$ Mev, while the choice of $\Delta\delta=0$ would have led to a value for $\sigma(\alpha,p)/\sigma(\alpha,n)$ of only 0.9 at 20 Mev. The value of $\Delta\delta$ obtained from the

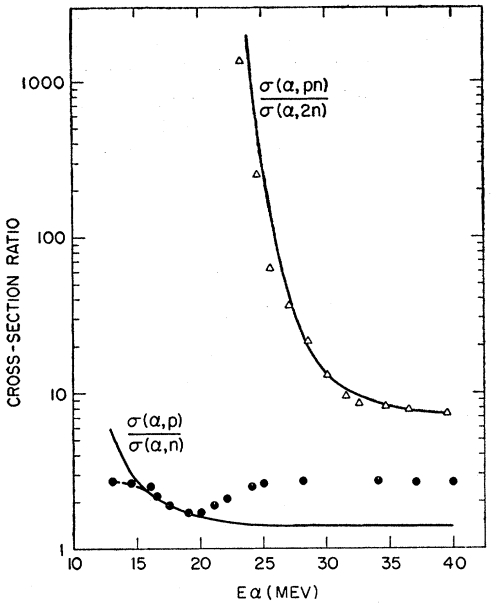

FIG. 10. Experimental and calculated values of $\sigma(\alpha,p)/\sigma(\alpha,n)$ and $\sigma(\alpha,pn)/\sigma(\alpha,2n)$, as a function of bombarding energy. The respective symbols for values from the experimental curves are ● and △. The dashed line is calculated on the assumption that there are levels between the ground state and characteristic level of the residual nuclei.

pairing and shell energies given by Cameron[42] is 0.33 Mev. The calculated ratio is not very sensitive to the value of a. The use of $a=6.7$ thus affects the calculated ratio by about 25%. In the case of the (α,pn) and $(\alpha,2n)$ reactions, the experimental and calculated ratios are in agreement for $\Delta\delta=2.85$ Mev. This rather large energy difference is due to the odd-odd and even-even nature of the residual nuclides in question. The value of $\Delta\delta$ obtained from Cameron's table[42] is 3.33 Mev. Cameron has also listed values for the pairing energy only.[43] The values of $\Delta\delta$ obtained from the latter for Ge^{66} and Ga^{66} and for Ge^{67} and Ga^{67} are 2.80 and 0.10 Mev, respectively. It is thus difficult to conclude from the present data whether shell effects should be included in the determination of the characteristic level for nuclear reactions. Examination of relative yields at a closed shell should be more informative because of the large value of $\Delta\delta$ due to shell effects when one of the two nuclides in question occurs at a closed shell. In either case it is seen that there is fairly good agreement between the pairing energy values obtained from cross-section data and from the more conventional procedure through mass-formula calculations.

The experimental ratios shown in Fig. 10 are seen to exhibit an initial decrease with increasing bombarding energy which is due to two factors. First, the energy thresholds of the (α,p) and (α,pn) reactions are lower than the corresponding thresholds for the (α,n) and $(\alpha,2n)$ reactions and the ratios must hence decrease from an initially infinite value. Second, the (α,p) reaction is affected by the competition of reactions involving further nucleon emission at a lower energy than the (α,n) reaction, because of the lower threshold of the $(\alpha,2p)$ reaction. Similarly, the (α,pn) reaction is affected by the competition of the $(\alpha,2pn)$ reaction at energies where there is as yet no reaction involving the emission of three nucleons competing with the $(\alpha,2n)$ reaction. The (α,n) reaction is, of course, also affected by further competition and when the $(\alpha,2n)$ reaction begins to compete effectively with it, the ratio of $\sigma(\alpha,p)/\sigma(\alpha,n)$ is seen to increase. The corresponding ratio for the (α,pn) and $(\alpha,2n)$ reactions does not exhibit a similar increase because of the very weak competition from the $(\alpha,3n)$ reaction. The increase in the value of $\sigma(\alpha,p)/\sigma(\alpha,n)$ is not duplicated by the calculation, because the latter does not duplicate threshold behavior particularly well.

The calculated values for the two ratios in question are considerably larger than the experimental values at the lowest energies. This effect is attributed to the assumption that there are no levels between the ground state of a nucleus and the characteristic level at which the level density expression takes hold, at an energy δ above the ground state. As a result the effective thresholds of all reactions in question are increased by an energy δ and the calculated ratio increases too rapidly as the thresholds are approached. It is possible to obtain better agreement with the experimental ratios by including the levels between the ground state and the characteristic level in the calculation. The emission function then has the form

$$f_x'=f_x+2M_x(2I_x+1)\int_{\epsilon_x=E^*-S_x-\delta}^{\epsilon_x=E^*-S_x}\epsilon_x\sigma_{cx}(\epsilon_x)Kd\epsilon_x,$$

where K is a constant related to the number of levels between the ground state and the characteristic level. Agreement was obtained for $\sigma(\alpha,p)/\sigma(\alpha,n)$ for $K\sim 10$–20 for both residual nuclides, as indicated by the dashed line in Fig. 10.

ACKNOWLEDGMENTS

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[43] A. G. W. Cameron, Can. J. Phys. **36**, 1040 (1958).

APPENDIX

A. Chemical Procedures

The procedures were adapted from previous radiochemical procedures.[44] Only the basic steps are listed. In practice, these steps were used a varying number of times.

Germanium.—$GeCl_4$ was distilled from 6*N* HCl solution saturated with Cl_2. The distillate was collected in water and GeS_2 was precipitated with H_2S from 3*N* HCl.

Gallium.—Germanium was removed by fuming with HCl. A 7*N* HCl solution was extracted twice with isopropyl ether. The ether fraction was washed with 7*N* HCl and gallium was back-extracted with water. The *p*H was adjusted to 7 and the oxinate was precipitated.

Zinc.—Gallium was extracted with isopropyl ether from 7*N* HCl. The aqueous residue was purified on a Dowex 1 column with 2*N* HCl. Zinc was eluted with dilute NH_4OH.

Copper.—Germanium was removed by fuming. CuCNS was precipitated from 0.5*N* HCl with SO_2 and NH_4CNS.

B. Counting and Calibration Procedures

280-day Ge^{68}.—Annihilation radiation of gallium daughter in equilibrium with Ge^{68} measured with a 3-inch NaI crystal connected to a 100-channel pulse-height analyzer; disintegration rate obtained from comparison with annihilation radiation of a calibrated Na^{22} source counted in the same geometry. In both cases the positrons were allowed to annihilate in aluminum absorbers placed on either side of the source. The relative counting rate of the annihilation radiation from Ga^{68} and Na^{22} was found to be independent of geometry. The Na^{22} source was calibrated by 511-511-γ coincidence measurements. It was assumed that Ga^{68} decayed by positron emission in 85% of the disintegrations.[29]

20-min Ge^{67}.—Allowed to decay to gallium daughter; the latter was chemically separated and counted.

2.5-hr Ge^{66}.—Same as Ge^{67}.

1.5-min Ge^{65}.—Gross activity counted in a deep-well scintillation counter; annihilation radiation determined with a 3-inch NaI crystal connected to a 100-channel pulse-height analyzer and compared with standard Na^{22} source for calibration purposes; positron emission in 95% of the disintegrations was assumed.[45]

78-hr Ga^{67}.—90+92 kev and 182+206 kev γ-ray photopeaks measured on 100-channel pulse-height analyzer with a 3-inch NaI crystal; counting efficiency obtained from published curves for 3 inch crystal[46]; geometry measured by use of a collimating lead baffle. The unconverted intensities of the (90+92)-kev and (182+206)-kev γ rays were taken as 0.44/disintegration and 0.27/disintegration, respectively.[47] Separate determinations using the two sets of γ rays agreed to within 10%.

9.4-hr Ga^{66}.—Gross activity determined with β proportional counter; annihilation radiation measured with a 3-inch crystal connected to a 100-channel pulse-height analyzer and compared with standard Na^{22} source for calibration purposes; positron emission in 66% of the disintegrations was assumed.[29]

15-min Ga^{65}.—This activity accounts for the entire yield of Ga^{65} [48]; gross activity counted on β proportional counter; the disintegration rate of Zn^{65} daughter was determined for calibration purposes.

245-day Zn^{65}.—1.12-Mev γ ray measured with a 3-inch crystal connected to a 100-channel pulse-height analyzer and compared with the 1.28-Mev γ ray of a standard Na^{22} source for calibration purposes; correction for differences in counting efficiency was made by use of the published efficiency curves[46]; the intensity of the 1.12-Mev γ ray was taken as 0.44/disintegration.[29]

38.3-min Zn^{63}.—Gross activity counted on β proportional counter; annihilation radiation measured with a 3-inch crystal connected to a 100-channel pulse-height analyzer and compared with standard Na^{22} source for calibration purposes; positron emission in 93% of the disintegrations was assumed.[29]

9.3-hr Zn^{62}.—Allowed to reach equilibrium with copper daughter before start of counting; annihilation radiation followed with a 3-inch crystal connected to a 100-channel pulse-height analyzer; disintegration rate obtained from observed counting rate by means of an experiment in which Cu^{62} was separated and counted.

9.7-min Cu^{62}.—Gross activity measured in a deep-well scintillation counter; annihilation radiation determined with a 3-inch crystal connected to a 100-channel pulse-height analyzer and compared with standard Na^{22} source for calibration purposes; positron emission in 99% of the disintegrations was assumed.[29]

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[48] D. L. Morrison and N. T. Porile, Phys. Rev. **113**, 289 (1959).