Excitation Functions for Nuclear Reactions between Complex Nuclei. I. Neutron Emission

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The excitation functions have been determined for the formation of Ce^{134}, Ce^{135}, and Ce^{137m} by the reactions $Te^{128}+C^{12}$, $Te^{130}+C^{12}$, and $Sn^{124}+O^{16}$. The maximum cross sections were less than 0.6 b for all the reactions studied. The excitation functions appear to be displaced by 10 to 15 MeV higher in energy than expected for compound nucleus formation reactions of low angular momentum. This energy shift agrees with the "extra" energy measured by two other different techniques for the same Te+C system. This agreement provides good evidence that the effect of high angular momentum is to decrease de-excitation by neutron emission relative to that by gamma emission.

INTRODUCTION

CONSIDERABLE interest has been exhibited in the study of nuclear reactions induced by ions heavier than alpha particles for several reasons. First, such heavy ions can cause nuclear reactions which proceed via compound nucleus formation at high total excitation energies. Second, much larger amounts of angular momentum can be introduced into the compound system than possible with light projectiles at the same excitation energies. The importance of the effect of high angular momentum has been demonstrated for fission induced by carbon, oxygen, and neon ions in a variety of targets from uranium to the lanthanide elements.[1,2] We have studied radiochemically the excitation functions for a number of reactions proceeding solely by neutron evaporation in an attempt to assess the evidence for effects due to high angular momentum. The reactions studied produced Ce^{134}, Ce^{135}, and Ce^{137m} by C^{12} bombardment of Te^{126}, Te^{128}, and Te^{130} and by O^{16} bombardment of Sn^{124}. In a subsequent paper, the excitation functions for the formation of Ba^{133m}, Ba^{135m}, and La^{135} by the same systems will be reported.

A number of authors[3–6] have considered the possible effects of angular momentum on nucleon emission. In general, the arguments may be summarized in this fashion. As the excited nucleus formed in the reaction undergoes nucleon emission, a distribution in energy and in spin states results. For those nuclear states characterized by spins greatly different from the spins of the available states in the residual nucleus following another nucleon emission, the rate of nucleon emission may be strongly diminished by the centrifugal barrier. Since gamma-ray emission has no such strong restriction by the centrifugal barrier, it may compete very favorably even though there is still sufficient excitation energy for nucleon emission. In the treatment of compound nucleus reactions by calculations such as Jackson has formulated,[7] it is assumed that as long as the total residual excitation energy exceeds the binding energy of the next neutron the latter is evaporated in preference to gamma de-excitation. For reactions involving high angular momenta, this may not be a valid assumption. Consequently, the excitation functions might be shifted to higher energies corresponding to the amount of energy "tied up" as rotational energy. In addition, it might be expected that the excitation curves would be broader than normal due to the relatively wide range of spins which the compound nucleus will possess. Of course, this concept of rotational energy effects may be greatly oversimplified but, at present, data are lacking to provide a more sophisticated model.

Sikkeland, Thompson, and Ghiorso[8] have investigated the excitation functions of californium isotopes from C^{12} reactions with U^{238} and Pu^{242}. Thomas *et al.*[9] have determined the excitation functions for $Au^{197}+C^{12}$ and $Pt^{195,196,198}+N^{14}$ forming astatine isotopes. Both groups of workers found that the experimental excitation functions from all of these reactions agreed well with curves calculated by the Jackson model using temperatures of 1.0 to 1.5 MeV, the normal range for reactions of low angular momentum. The failure to observe the expected effects of high angular momentum in these cases can be explained by assuming that fission occurs preferentially for the high-spin states. Karamyan, Gerlit, and Myasoedov[10] studied the reaction $V^{51}(C,2n)Zn^{61}$. They found that the excitation function was broad and reached a maximum about 8 MeV higher than $Cu^{65}(p,2n)Zn^{64}$. Their results,

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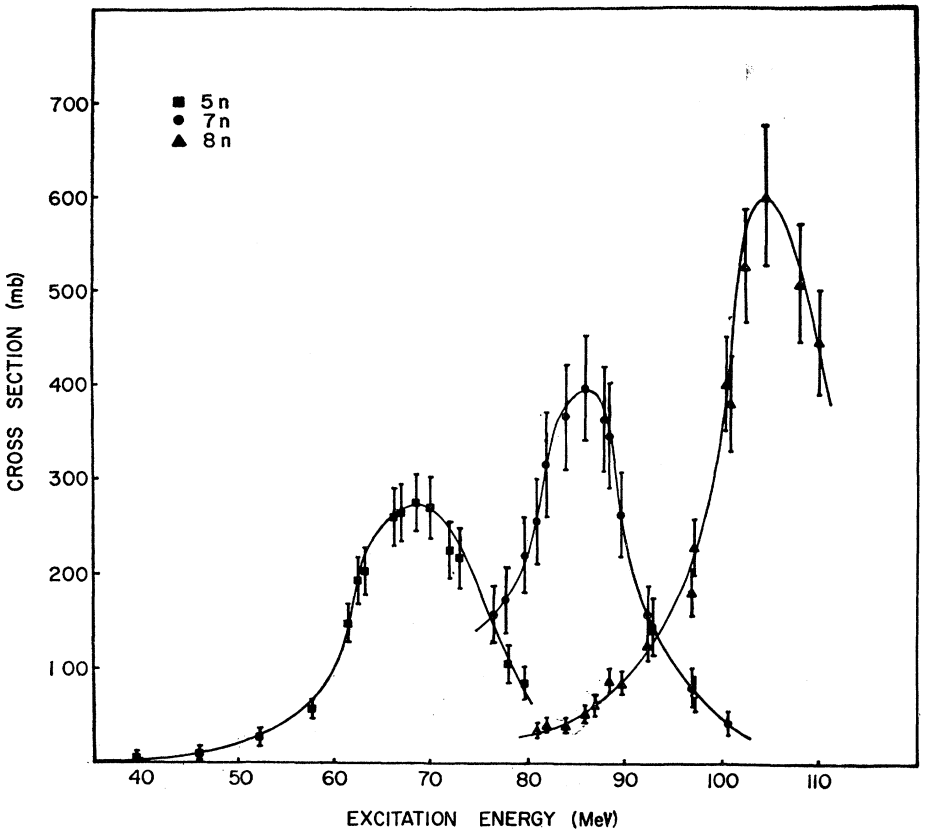

FIG. 1. Excitation functions of $Te^{130}+C^{12}$.

however, are not sufficiently extensive to draw conclusive results. Recently, Glover and Weigold[11] have suggested the presence of an angular momentum effect in their study of the reaction $Ni^{58}+n \rightarrow Co^{58}+p$.

Following our initial results, the Te+C system was investigated by two other techniques for angular momentum effects. The number and energy of the gamma rays emitted were determined by Mollenauer,[12] while the range and angular distribution of the recoil product nuclei were measured by Morton, Choppin, and Harvey.[13] The results of all three investigations are compared in the discussion.

EXPERIMENTAL

The tellurium targets were prepared by electrodeposition from a solution of 1.5–1.8 mg/ml of the separated isotope in $3M$ perchloric acid. A mirror plate of tellurium metal of 0.7 to 1.0 mg/cm^2 was obtained on 0.1-mil gold foil at a voltage of 1.2 V. The tin targets were prepared by vacuum vaporization of natural tin metal onto 0.25-mil aluminum foil. Targets of 0.4 to 0.5 mg/cm^2 of tin were used. The tellurium and tin targets were assembled in stacks of 12 with aluminum absorber foils either in front or in the stack as desired for energy degradation. The separated tellurium isotopes, Te^{126}, Te^{128}, and Te^{130}, were obtained from ORNL.

The target stacks were bombarded for 2 to 5 h in the HILAC accelerator of the Lawrence Radiation Laboratory (Berkeley) with beams of C^{12} and O^{16} ions whose initial energy was 10.2 MeV/nucleon. The range-energy curves compiled by Hubbard were used to calculate the average bombarding energy of each foil in a stack.[14] The beam intensities reported by the HILAC group for each bombardment were used for calculation. The accuracy of such beam intensity measurements is considered to be $\pm 10\%$ for the HILAC. Following bombardment, the targets were shipped via air to Tallahassee.

The chemical separation of the cerium isotopes from the target was accomplished using a procedure which was basically similar to that given by Hicks.[15] The final cerium counting samples were obtained as cerium oxalate nonahydrate. Since the cerium isotopes, Ce^{134}, Ce^{135}, and Ce^{137m}, decayed primarily by electron capture, the counting and determination of the absolute cross

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[12] J. F. Mollenauer, Phys. Rev. **127**, 867 (1962).
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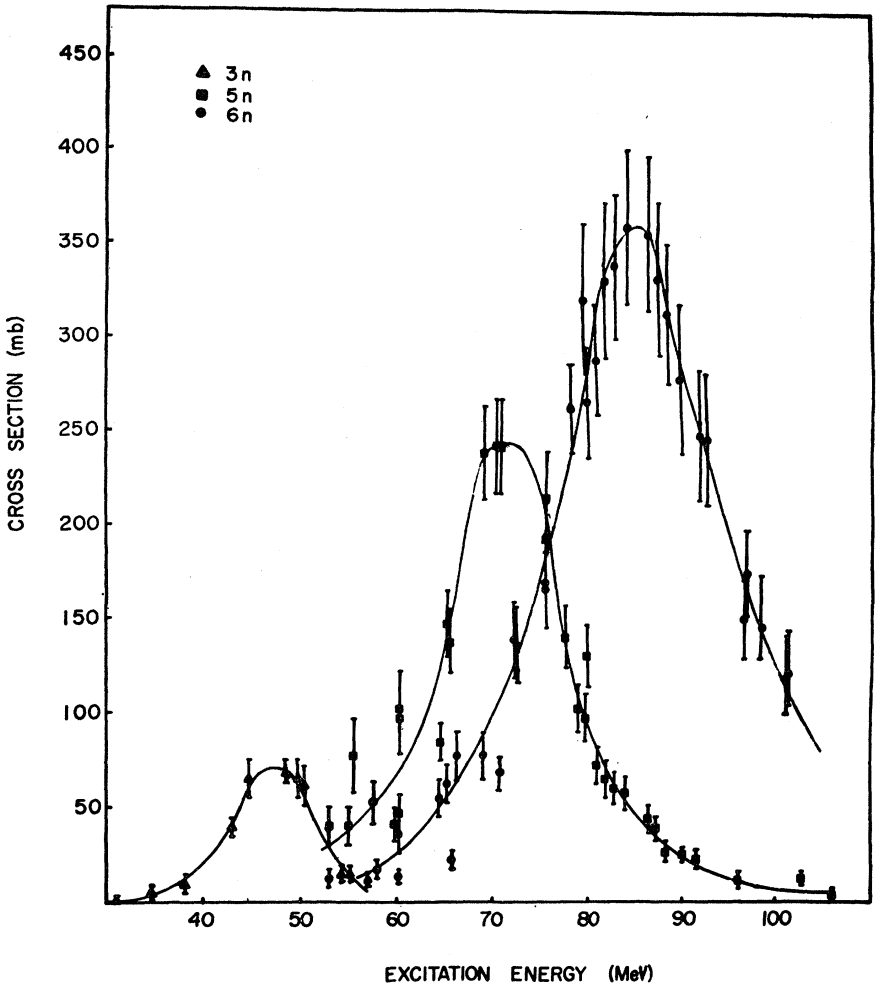

FIG. 2. Excitation functions of $Te^{128}+C^{12}$.

sections was accomplished by measurement of the x-ray spectra. The equipment used to measure the x-ray spectra was a NaI(Tl) scintillation crystal used in conjunction with a TMC 256-channel pulse-height analyzer.

The radioactivity in the 12 counting samples produced in each bombardment was determined by integration of the x-ray peak. The activity due to each isotope in each counting sample was obtained from graphical analysis of the decay curves.

Graphical analysis was possible in this work even though the half-lives are relatively close because the three isotopes investigated were produced at different excitation energies. In no case were more than two activities produced in a single counting sample. The 72-h (Ce^{134}) and the 22-h (Ce^{135}) activities were easily resolved from each other. In the case of the resolution of the 22-h (Ce^{135}) and the 35-h (Ce^{137m}) difficulty was encountered only in the overlap region of the two excitation functions as can be seen in Figs. 1 and 2 for Te^{128} and Te^{130} targets. This difficulty was greatly reduced by checking the Ce^{137m} excitation curve based on the x-ray spectra against that from the 255-keV internal transition gamma spectra.

However, in the case of Ce^{135} (22 h) which decays to La^{135} (19 h), the graphical method of resolution required correction of the measured activity for grow-in of La^{135}. Since the time between the chemical separation of Ce from La and counting of the Ce samples was very short compared to the half-lives of the isotopes involved, the correction was small (of the order of 0.94 to 0.97 of the measured cerium activity).

Since a number of bombardments were carried out on each of the systems studied, the calculation of the excitation function for a given isotope was simplified by normalization of the data on that isotope from different bombardments to that of one bombardment. The cross section for production of each isotope was then calculated in the normal manner from the absolute disintegration rate at end of bombardment. The absolute disintegration rates were obtained by application of correction factors for fluorescent yield,[16] total efficiency

[16] H. L. Hagedoorn and A. H. Wapstra, Nucl. Phys. **15**, 146 (1961).

factor,[17] window absorption, isotopic abundance, and K x-ray decay abundance.[18]

An error analysis of the results of this investigation showed that there was approximately 12% error in determination of the cross sections at the maximum in the excitation function curve and about 15 to 30% error in the cross sections in the leading and trailing edges of the curves. However, it should be noted that this analysis has not taken into account the possibility of error in the decay schemes used. Also, the counting equipment was not calibrated for the specific decaying species measured. There is also an uncertainty in the bombarding energy of about 2%.

TABLE I. xn excitation functions for $Te^{130}+C^{12}$.

5n			7n			8n		
E (MeV)	σ (mb)	ϵ_σ (mb)	E (MeV)	σ (mb)	ϵ_σ (mb)	E (MeV)	σ (mb)	ϵ_σ (mb)
39.5	6		76.5	157	±33	80.9	34	±5
45.9	9		77.9	173		82.0	40	
52.2	26		79.7	219		83.9	40	
57.7	58	±8	80.9	256		85.9	53	
61.6	146		82.0	316		87.0	63	
62.5	193		83.9	366		88.4	87	
63.3	203	±24	85.9	389	±54	89.8	85	
66.2	259		88.0	363		92.6	125	±17
67.1	265		88.4	346		93.0	131	
68.4	275	±30	89.8	262	±45	96.9	180	
70.1	269		92.6	157		97.3	228	
72.1	224		93.0	143		100.5	399	±52
73.3	218		96.9	79		100.9	379	
77.9	105	+19	97.3	73	±17	102.5	525	
79.7	84		100.8	42		104.5	596	±75
						108.1	504	
						108.6	444	±56

The length of time between end of bombardment and separation (26 to 30 h) plus the difficulties of unique detection of its radiations prevented us from obtaining data on Ce^{137g} production. Macfarlane has recently published data on production of isomeric states in heavy-ion induced reactions.[19]

RESULTS

The calculated cross sections as a function of energy are given in Tables I, II, and III for the $Te^{128}+C^{12}$, $Te^{130}+C^{12}$, and $Sn+O^{16}$ systems.

Nuclear temperatures of the experimental excitation functions were calculated by the expression

$$T=(E^*-\sum_{i=1}^{x} B_i)/2x,$$

where E^* is the center-of-mass excitation energy corresponding to the maximum in the excitation function for the reaction $A(\text{H.I.},xn)B(\text{H.I.}=C^{12}\text{ or }O^{16})$ and B_i is the neutron binding energy. This assumes that each neutron carries off an average of $2T$ MeV as kinetic

TABLE II. xn excitation functions for $Te^{128}+C^{12}$.

6n			5n			3n		
E (MeV)	σ (mb)	ϵ_σ (mb)	E (MeV)	σ (mb)	ϵ_σ (mb)	E (MeV)	σ (mb)	ϵ_σ (mb)
53.0	12	±4	53.0	41	±11	31.5	0.3	
55.4	14		55.1	41		35.1	5	±1
58.0	18		55.5	77		38.3	10	
60.3	14		57.5	53		42.9	38	±6
60.3	42		59.8	41		44.7	64	
64.4	55	±10	60.3	48		48.4	68	±8
65.3	62		60.3	97	±19	49.8	65	
65.8	22		60.3	102		50.2	62	
66.2	77		64.4	84		54.3	15	±4
69.0	77		65.3	147	±18	55.2	14	
70.8	68		65.6	137		57.5	12	±4
72.2	139	±19	69.0	237				
72.6	137		70.3	241	±25			
75.4	164		70.8	241				
75.4	169		75.4	214				
78.1	261		75.4	193				
79.7	265	±34	77.6	140				
79.5	320		78.8	102	±13			
80.8	287		79.5	97				
81.8	329		79.7	129				
82.7	338		80.8	73				
84.0	358	±46	81.8	65				
86.3	356		82.7	60	±8			
87.2	331		84.0	58				
88.2	312		86.6	44				
89.5	278		87.2	38				
91.4	247	±34	88.2	26				
92.3	245		90.0	25				
96.4	150		91.4	22	±4			
96.8	174		95.9	11				
97.8	152		102.8	12				
100.9	119		106.0	4	±1			
101.2	121	±17						
102.8	93							
106.0	60							
106.4	60	±9						
110.1	22							

energy. The excitation energies were calculated from the bombarding energies using the masses and neutron binding energies in Cameron's tables.[20] The nuclear temperatures calculated in this fashion are listed in Table IV. The excitation functions are shown in Figs. 1, 2, and 3 where they are presented as a function of the

TABLE III. xn excitation functions for $Sn^{124}+O^{16}$.

5n			6n		
E (MeV)	σ (mb)	ϵ_σ (mb)	E (MeV)	σ (mb)	ϵ_σ (mb)
36.5	45	±9	36.5	60	±14
44.9	60		44.8	75	
52.4	75		52.4	100	
59.5	152		59.5	150	
63.1	192	±28	63.1	225	±38
66.6	190		66.6	298	
69.3	164		69.7	326	
73.3	116		73.3	329	
75.5	100	±17	75.5	333	
79.5	82		79.5	333	±47
80.8	70		80.8	330	
85.2	46		85.2	313	
90.5	16		90.5	269	
91.4	13	±3	91.4	232	
95.4	3		95.4	190	
96.7	1		96.7	145	±27
			100.3	111	
			102.0	89	
			104.7	71	
			107.4	52	
			111.8	29	
			116.2	20	
			120.2	12	±3

[17] S. H. Vegors, L. L. Marsden, and R. L. Heath, AEC Research and Development Report IDO-16370 (1958) (unpublished).

[18] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington 25, D. C.), NRC 61-2, 1961.

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[20] A. G. W. Cameron, Can. J. Phys. **35**, 1021 (1957).

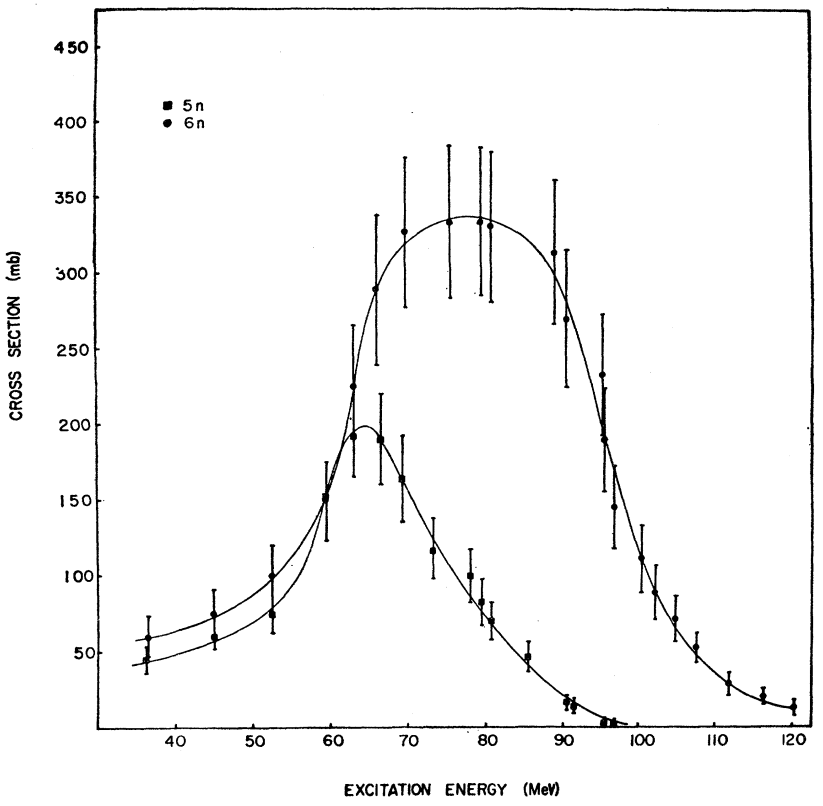

FIG. 3. Excitation functions of $Sn^{124}+O^{16}$.

excitation energy in the c.m. system. The experimental points are the result of many bombardments for each target system.

The excitation functions for $Sn^{124}+O^{16}$ appear to be quite different in appearance than the Te+C excitation functions. Since natural tin was used as the targets Sn^{122} was present in 4.8% abundance compared to the 6.1% abundance for Sn^{124}. Using the excitation functions for $Te^{128}(C,3n)Ce^{137}$ and $Te^{128}(C,5n)Ce^{135}$ and an estimated (H.I.,4n) curve, an estimation was made of the contribution of the $Sn^{122}(O^{16},3n)$ reaction to the Ce^{135} formation and of the $Sn^{122}(O^{16},4n)$ reaction to the Ce^{133} formation. Even though this is only a rough estimate, it is clear that these reactions contribute in appreciable fashion only to the leading (low-energy) side without changing the position of the maxima. This correction causes the leading edges for the Sn^{124} reactions to drop more sharply and, hence, to resemble more the Te+C reactions. However, the curves are still much broader than the corresponding Te+C curves and the temperatures are lower.

TABLE IV. Nuclear temperatures (in MeV) for the Cerium isotope excitation functions.

System	3n	4n	5n	6n	7n	8n
Te^{130}	...	...	2.80	...	2.00	2.40
Te^{128}	2.00	...	2.50	2.60	...	...
Te^{126}	...	2.75	...	...	...	...
Sn^{124}	...	...	1.9	2.0	...	...

DISCUSSION

Jackson and others[21,22] have pointed out that the nuclear temperature required to calculate curves which agree with experimental data for systems of low angular momentum is frequently on the order of 1.0 to 1.5 MeV. If it is assumed that the temperature of systems of little or no angular momentum is 1.2 MeV, then the expected energies corresponding to the maxima in the excitation functions for these systems can be calculated. The difference between these peak energies and the experimental values can be interpreted as a measure of the energy shift due to angular momentum effects. The energy shifts calculated with this 1.2-MeV temperature are given in Table V.

Morton *et al.*[13] have compared the experimental angular distributions for Ce^{137m} from the $Te^{130}(C,5n)$ reaction with distributions calculated by a Monte Carlo method based on the compound nucleus and statistical models. In order to fit the calculations to the experimental data, it was necessary to assume that a portion of the excitation energy was unavailable to neutron

[21] R. Vandenbosch, T. D. Thomas, R. A. Glass, and G. T. Seaborg, Phys. Rev. **111**, 1358 (1958).

[22] R. Vandenbosch and J. R. Huizenga, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, p. 284.

TABLE V. Energy shifts for the Cerium (xn) product excitation functions.

System	$3n$ (MeV)	$4n$ (MeV)	$5n$ (MeV)	$6n$ (MeV)	$7n$ (MeV)	$8n$ (MeV)
Te^{130}	...	...	17	...	11	18
Te^{128}	13	...	13	16	...	...
Te^{126}	...	13	...	...	...	...
Sn^{124}	...	...	7	10	...	...

evaporation. This energy was presumably removed by gamma de-excitation. For a nuclear temperature of 1.2 MeV, the "excess" gamma energy was about 16 MeV at 99 MeV excitation energy.

Mollenauer's measurement of the gamma rays from the Te+C system at 99-MeV average excitation energy gave a total gamma-ray energy of 12.2 MeV. However, this assumed a 2 b total cross section. Assuming 1.5 b to be a better estimate based on our excitation functions for (C,xn), (C,pxn), and (C,$\alpha xn+2pyn$), the gamma energy value is more likely about 16 MeV. This implies an "excess" gamma energy of approximately 10 MeV.

Thus, we can compare the excess energy determined by Morton (16 MeV), Mollenauer (10 MeV), and by us (15 MeV) for an excitation energy of 99 MeV. Mollenauer's results may be in disagreement due to the fact that he measured the total gamma-ray energy for all processes occurring for Te+C at that energy, whereas only Te(C,5n)Ce^{137m} was studied in the other two cases. Considering the inherent differences, the three methods are in reasonable agreement and provide evidence that the "extra energy" effect in heavy ion induced reactions arises from an increase in the ratio of the rate of gamma ray de-excitation to the rate of neutron emission, and it is a strong presumption that this effect is connected in some way with the high angular momentum introduced.

Pik-Pichak has calculated that evaporated neutrons on the average carry off less than one $\hbar$ unit of angular momentum. After initial evaporation of several neutrons and the removal of a large fraction of the original excitation energy, subsequent neutron evaporation is hindered by the small probability of residual nuclear states of high angular momentum. Gamma emission removes less excitation energy per event and more angular momentum; so it is favored. In support of this model, Mollenauer found approximately twice as much energy removed by gamma emission in carbon ion as in alpha bombardments.

The difference between the Sn+O and the Te+C systems is more difficult to explain. It cannot be ascribed to large differences in angular momentum between Te+C and Sn+O since this difference is believed to be rather small over the range of bombarding energies.[23] Unfortunately, this difference was not known at the time of the studies by Morton and Mollenauer. Qualitatively, we can say that there is a difference in the relative population of the spectrum of spin states of Ce^{140} at the same excitation energy even though the classically calculated angular momenta have very similar values. Mollenauer found preference for quadrupole emission in the systems other than Te+C, whereas the latter proceeded predominantly by dipole transitions. Comparison of the type of gamma emission for Te+C and Sn+O might be quite instructive in understanding their difference. Wildermuth[24] has suggested the difference may be explained by the fact that in the formation of the compound system two different nuclear clusters are formed. Even though the total angular momentum was the same, the two original clusters could result in different relative population of spin states in the spin spectrum of the compound nucleus.

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[23] T. D. Thomas, Phys. Rev. **116**, 703 (1959).
[24] K. Wildermuth (private communication).