the thermal value. However, the background is no more than 14% of the signal at resonances over energies from 15 to 40 eV and, for most of this region, is less than 8% of the signal at resonances.

For easier comparison of the current Ag^{111}/Mo^{99} data with that obtained in the 1958 experiment, results obtained at resonances in the energy region 8.8 to 39 eV are plotted in Fig. 3 for both experiments.

DISCUSSION

In this experiment, values for the Ag^{111}/Mo^{99} activity ratio fell below the thermal value by as much as 50% at the following resonances (uncertain assignments indicated by parentheses): (8.8), 15.4, 16.1, 16.7, (18.0), 19.3, 21.1, 22.9, 23.6, (26.5), (27.8), (32.1), and (33.2) eV. Observed values were as much as 22% greater than thermal for the following resonances: 25.5, (34.4), 35.3, and 39.4 eV. An explicit contradiction of the earlier assignments of levels by symmetry occurs at 15.4 eV.

In both the 1958 data and the present data, there is some indication of a periodicity in the symmetry with changing energy. This effect, if it is real, may be an indication that the strongest resonances produce small bands which have a common spin or some other property influencing symmetry in fission.

Although the quantitative results for the Ag^{111}/Mo^{99} activity ratios are systematically lower than those reported in the earlier paper, due chiefly to improvements in the signal-to-background ratio, substantial confirmation of most of the qualitative results of the 1958 experiment is reassuring as to the reality of the effect. The simplest explanation of these results is that fission occurs in one or another of only two possible distributions. A more definitive answer will probably be obtained from similar measurements on Pu^{239}.

ACKNOWLEDGMENTS

We wish to acknowledge the assistance of Charles W. Fuller and Charles M. George (now deceased), who were responsible for engineering and field supervision; of Francis G. Berry, who was responsible for autoradiography; and of Donald F. Silver, Hillis J. Harrington, and George E. Jaynes, who devised precision methods for cutting the metal after bombardment.

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Excitation Functions for Tb^{149g} from Reactions between Complex Nuclei*

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Excitation functions have been measured for twelve reactions that produce 4.1-h Tb^{149g} from Tb compound nuclei. Projectiles were B^{10}, B^{11}, C^{12}, N^{14}, N^{15}, O^{16}, O^{18}, and F^{19}. Peak cross sections range from approximately 0.5% to approximately 7% of the calculated total reaction cross section. The excitation functions are well systematized by the simple assumption that only those compound nuclei of angular momentum less than $7.5\pm1.5\hbar$ are effective in these reactions.

I. INTRODUCTION

THE production of different isomeric states by nuclear reactions gives information about the effect of angular momentum on the decay of the initial compound nuclei. Huizenga and Vandenbosch have given a detailed discussion of the various factors that influence relative isomeric yields.[1] The basic assumption in these considerations is the preference for small changes in angular momentum associated with photon or neutron emission. Huizenga and Vandenbosch have used relative isomeric yields to obtain information about the spin dependence of the nuclear level density.[1]

A particularly favorable case of nuclear isomerism is that of Tb^{149}. The product 4.1-h Tb^{149g} can be identified by its α radiation, and isomeric transition from 4.0-min Tb^{149m} is very improbable.[2] We use yields of the low-spin state 4.1-h Tb^{149g} to obtain information about the angular momenta of the initial compound nuclei in (HI,xn)Tb^{149g} reactions (HI denotes a heavy ion, e.g., C^{12}, N^{14}, etc.).

We have measured excitation functions for twelve different (HI,xn)Tb^{149g} reactions. The peaks of these excitation functions have values between approximately 0.5% and approximately 7% of the calculated total reaction cross section.[3] Recoil range studies previously presented give strong evidence that these reac-

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[1] J. R. Huizenga and R. Vandenbosch, Phys. Rev. **120**, 1305 (1960); R. Vandenbosch and J. R. Huizenga, *ibid.* **120**, 1313 (1960).
[2] R.D. Macfarlane, Phys. Rev. **126**, 274 (1962).
[3] T. D. Thomas, Phys. Rev. **116**, 703 (1959).

tions are essentially pure compound-nucleus reactions.[4] We are able to correlate all these excitation functions with the simple assumption that only those compound systems of angular momentum less than $7.5\pm1.5\hbar$ contribute to the production of Tb^{149g} in these reactions. We conclude that the 4.0-min Tb^{149m} is a very effective shield for inhibiting production of Tb^{149g} from compound nuclei of high spin.

II. EXPERIMENTAL PROCEDURES AND RESULTS

We have irradiated stacks of target and recoil catcher foils with beams from the Berkeley heavy-ion linear accelerator. Beam intensities were 0.05 to 0.20 μA. The beam was deflected through 11 or 30 deg, and collimated to a circle of $\frac{3}{8}$-in. diam. The target assembly served as a Faraday cup, and the charge collected was measured by an electrometer. Electron scattering into or out of the Faraday cup was inhibited by a permanent magnet.

The targets were thin layers (30 to 120 $\mu g/cm^2$) of $BaCl_2$, La, Ce, Pr, or Nd evaporated onto 0.00025-in. Al. The thickness of the deposits was determined by measurement of the weight and area of the target layer. Successive weighing of the La and Pr deposits showed that the weight increased with time (final weight approximately 20% greater than initial weight). Presumably the evaporated metallic layers were oxidizing rather slowly. No weight increase was observed for the Ce, Nd, and $BaCl_2$ deposits. Therefore, we infer that the oxidation of Ce and Nd was complete before the initial weighing and that $BaCl_2$ did not absorb water. We have assumed that the initial weights represent La and Pr metals, CeO_2, Nd_2O_3, and $BaCl_2$. Some isotopically enriched elements were used: 98.0% Ba^{138}, 99.6% Ce^{140}, 97.4% Nd^{142}, 97.3% Nd^{144}, and 96.2% Nd^{146}. The target foils were used many times.

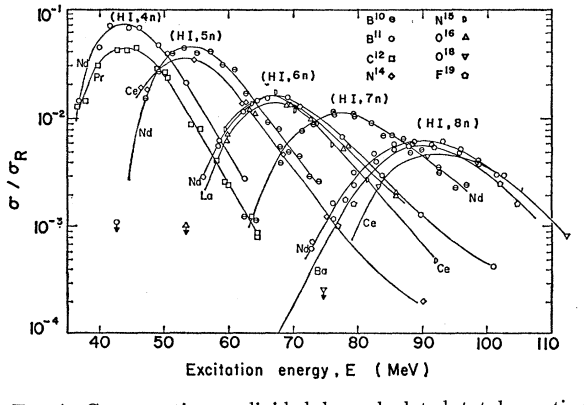

FIG. 1. Cross section σ divided by calculated total reaction cross section σ_R (reference 3) for (HI,xn)Tb^{149g} reactions. The various projectiles, targets, and reactions are indicated. Excitation energies were calculated from Seeger's mass formula; P. A. Seeger, Nucl. Phys. **25**, 1 (1961).

In some experiments a stack of thin Al catcher foils (approx 0.15 mg/cm^2) was used. In others, one or two thick Al catcher foils (approx 1.8 mg/cm^2, 99.8% purity) were used. Recoil range studies previously reported have shown that essentially all the Tb^{149g} (>98%) recoils out of the target layers into the catcher foils.[4] We measured the α radioactivity of the catcher foils with 2π ionization chambers. No chemical separation was required. Decay curves showed a pure 4.1-h half period of Tb^{149g} for approximately 16 h. Blank foils indicated that the activation of impurities in the catcher foils was negligible. No absorption correction for the α radiation was applied in experiments using the thinner catcher foils. Absorption corrections for the other experiments are described in the Appendix.

The results of these experiments are given in Table I and in Fig. 1. Data from the reaction $Nd^{142}(B^{10},3n)Tb^{149g}$ do not appear in Fig. 1 because of large uncertainties in the calculated total reaction cross section.[3] Measurements of relative values of the cross sections from a single experiment depend on only the relative target thicknesses and counting rates. We estimate that uncertainties from the above sources give standard deviations in the relative values of approx 10% for the Ce targets and approx 5% for all other targets. The reproducibility of the results from one experiment to another indicate that the relative beam intensities have a standard deviation of approx 5%. We estimate uncertainties in the following absolute quantities: (a) beam intensity, approx 10%; (b) target thickness, approx 10%; (c) absolute activity measurements, approx 5%; and (d) alpha branching ratio for 4.1-h Tb^{149g}, 20%.

The beam energies were calculated from the range-energy curves of Northcliffe[5] and an initial beam energy of 10.38 MeV/amu. The reproducibility of these excitation functions and those reported later leads us to believe that the day-to-day and month-to-month variations in the initial beam energy are less than $\pm\frac{3}{4}$%. However, no extensive study of this question has been made.

III. DISCUSSION

Our measurements show that the production of Tb^{149g} from $_{65}Tb$ compound nuclei is not a very probable process (see Table I and Fig. 1). This is in contrast to the high yields of Dy^{149}, Dy^{150}, and Dy^{151} from $_{66}Dy$ compound nuclei.[6] In those reactions producing Dy compound nuclei, a very large fraction f_n of the total reaction cross section leads to Dy final products. The dependence of f_n on excitation energy E (in MeV) for Dy can be described by the following empirical

[4] J. M. Alexander and D. H. Sisson, Phys. Rev. **128**, 2288 (1962) L. Winsberg and J. M. Alexander, Phys. Rev. **121**, 518 (1961).

[5] L. C. Northcliffe, Phys. Rev. **120**, 1744 (1960).

[6] J. M. Alexander and G. N. Simonoff, Lawrence Radiation Laboratory Report UCRL 10541 (unpublished).

TABLE I. Cross-section results.[a]

E_b (lab) (MeV)	σ (mb)	E_b	σ	E_b	σ
$Nd^{142}(B^{10},3n)Tb^{149g}$		$Pr^{141}(C^{12},4n)Tb^{149g}$		$Nd^{144}(B^{10},5n)Tb^{149g}$	
52.9	5.69	8.51	1.32	78.6	4.92
48.2	18.7	80.3	3.42	75.2	7.92
43.0	39.6	75.2	9.90	71.8	13.7
38.5	4.70	69.8	25.1	68.1	23.6
...	...[b]	64.6	36.7	64.2	38.0
41.6	27.6	58.3	16.9	60.0	41.5
35.6	<0.4	52.0	<0.1	55.9	34.2
...	...	...	...	51.5	9.95
49.1	21.3	83.9	1.86	46.7	<0.4
44.1	40.9	78.2	5.27	...	...
39.0	2.36	69.5	26.4	80.2	4.68
...	...	63.1	32.7	77.0	7.48
55.4	4.19	56.0	6.11	73.6	12.4
50.9	13.0	...	...	69.9	20.8
45.7	37.2	85.0	1.25	66.2	36.9
40.5	11.8	79.6	3.42	62.2	46.7
		74.0	10.4	58.1	43.0
$Nd^{142}(B^{11},4n)Tb^{149g}$		68.2	27.8	53.8	20.6
62.8	25.2	61.4	29.9		
58.2	47.1	54.7	4.65		
53.2	51.6	...	...		
48.4	22.5	74.9[c]	15.6		
42.5	<0.13	68.6[c]	35.8		
...	...	62.4[c]	28.0		
72.6	4.26				
68.5	8.57				
59.8	41.8				
55.2	58.2				
50.3	43.3				
45.0	4.30				
$Ce^{140}(N^{14},5n)Tb^{149g}$		$Nd^{144}(B^{11},6n)Tb^{149g}$		$La^{139}(O^{16},6n)Tb^{149g}$	
114.9	<0.38	112.8	0.96	94.2	13.8
98.3	1.99	100.8	2.75	...	...
90.9	6.77	87.6	12.5	127.8	3.16
83.9	16.2	80.5	23.6	119.0	4.04
75.6	32.1	74.9	25.6	110.1	9.02
66.6	10.8	71.1	17.7	100.3	18.1
56.8	<0.05	67.0	7.51	89.4	6.62
...	...	...	...	...	...

E_b (lab) (MeV)	σ (mb)	E_b	σ	E_b	σ
$Ce^{140}(N^{14},5n)Tb^{149g}$		$Nd^{144}(B^{11},6n)Tb^{149g}$		$La^{139}(O^{16},6n)Tb^{149g}$	
106.4	<0.55	94.2	5.99	103.7	14.5
84.4	17.0	78.4	28.0	93.1	14.3
67.6	11.2	64.4	3.48	82.2	0.86
				...	...
		$Ce^{140}(N^{15},6n)Tb^{149g}$		118.4	3.48
		120.0	1.1	109.4	8.46
		99.0	14.6	99.7	18.5
		92.6	25.3	89.4	7.78
				77.9	1.06
		84.0	9.95		
		...	...		
		125.0	<0.95		
		108.3	5.09		
		102.2	9.98		
		95.8	19.0		
		88.5	18.1		
$Nd^{146}(B^{10},7n)Tb^{149g}$		$Nd^{146}(B^{11},8n)Tb^{149g}$		$La^{139}(O^{18},8n)Tb^{149g}$	
102.6	5.08	111.5[c]	6.87	147.4	1.79
98.0	7.20	108.5[c]	8.52	131.0	7.97
95.2	10.2	105.5[c]	10.6	122.4	8.75
92.0	12.6	100.8[c]	12.0	133.9	4.26
88.9	13.0	97.8[c]	12.4	104.2	<0.4
84.0	19.4	94.5[c]	11.8		
80.7	19.6	91.3[c]	9.29	$Ba^{138}(F^{19},8n)Tb^{149g}$	
77.3	15.0	88.0[c]	6.07	137.6	4.78
71.9	6.01	84.5[c]	3.08	125.8	9.69
67.9	1.56	81.1[c]	1.31	111.9	2.32
64.0	<0.97	77.6[c]	<0.75	96.5	<0.08
...	...	...	...	...	...
100.9	4.93	112.8	6.89	151.6	1.16
98.0	6.54	108.5	9.19	121.4	9.39
93.4	9.74	105.6	11.5	109.1	1.39
90.3	13.4	102.7	13.4	95.6	<0.09
87.1	16.8	97.9	12.7	...	...
84.0	20.1	94.5	10.6	140.6	3.35
80.7	20.8	91.5	7.84	110.4	2.42
75.3	12.4	88.0	4.66	97.1	<0.09
71.8	8.32	84.5	2.17		
65.8	1.57	81.0	1.11		

[a] A value of 10% was used for the alpha branching ratio of 4.1-h Tb^{149g} [L. Winsberg, Bull. Am. Phys. Soc. 3, 406 (1958)].

[b] Different experiments are separated by dotted lines.

[c] These energies may be systematically in error by 1 to 2 MeV.

relationship[6]:

$$f_n=(\tfrac{1}{2})^{(E-35)/65} \quad \text{for} \quad 45<E<105 \text{ MeV}. \qquad (1)$$

It is reasonable to suppose that f_n for $_{65}Tb$ is not very different from that for $_{66}Dy$ compound nuclei. Therefore, we assume that the cross sections for 4.0-min Tb^{149m} are much greater than those for 4.1-h Tb^{149g}. Indeed, Macfarlane has observed that the ratio of Tb^{149m} to Tb^{149g} production increases very rapidly with increasing energy.[2] From this observation he concludes that Tb^{149m} has higher spin than Tb^{149g} and that isomeric transition is improbable.

The probability of isomeric transition (I.T.) for 4-min Tb^{149m} has not been determined. However, we can infer that this I.T. branching ratio is less than 0.5% by the following argument. Excitation functions have been measured[6] for many reactions of the type (HI,xn)Dy^{149}, Dy^{150}, and Dy^{151}. These excitation functions all have very similar shapes and magnitudes when plotted as in Fig. 2. In Fig. 2 we compare the excitation functions for the re-

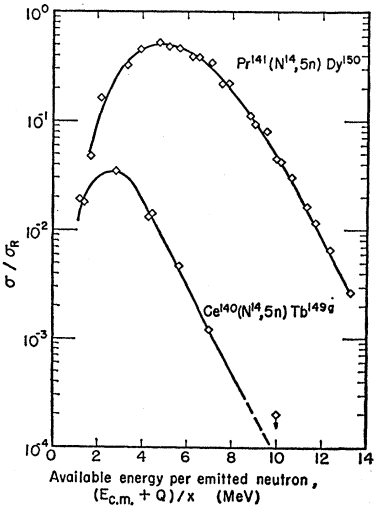

FIG. 2. Cross section σ divided by total reaction cross section σ_R (reference 3) vs available energy per emitted neutron $(E_{c.m.}+Q)/x$ for two (HI,xn) reactions.

actions $Ce^{140}(N^{14},5n)Tb^{149g}$ and $Pr^{141}(N^{14},5n)Dy^{150}$. We assume that the striking differences in shape and magnitude of these excitation functions are due to the formation of 4-min Tb^{149m}. If this assumption is correct, then the cross section for 4-min Tb^{149m} must be at least 200 times that for 4.1-h Tb^{149g} at 10.0 MeV per emitted neutron (see Fig. 2). Also, the I.T. branching ratio for 4-min Tb^{149m} must be less than 1/200.

The results in Fig. 1 show marked differences between the two reactions $(HI,4n)Tb^{149g}$ and also the three reactions $(HI,8n)Tb^{149g}$. Very close correlation has been observed in similar plots for $(HI,xn)Dy$ reactions.[6]

Let us consider the possibility that the Tb compound nuclei of high spin preferentially decay to 4.0-min Tb^{149m} and that only those compound nuclei of low spin decay to 4.1-h Tb^{149g}.

In order to calculate the cross sections for producing both isomeric states, we need the following information: (a) the total reaction cross section σ_R; (b) the distribution function P_l, which describes the probability of populating various states of angular momentum l of the original compound nuclei; (c) the relative probabilities of decay by photon emission, neutron emission, and charged-particle emission $\Gamma_\gamma:\Gamma_n:\Gamma_c$; (d) the change in the angular-momentum spectrum associated with the emission of particles or photons; and (e) the spins of the final products.

Our experiments give excitation functions for producing the low spin isomer of Tb^{149}. We do not attempt a detailed calculation of the shape of these excitation functions, but we do present a correlation of the results that strongly suggests that only those compound nuclei of low angular momentum ($<7.5\pm1.5\,\hbar$) are effective in these reactions.[7]

We treat each of the foregoing points as follows: (a) Barrier-penetration cross sections have been calculated by T. D. Thomas for a square-well nuclear potential.[3] These calculations give values of the total reaction cross section σ_R. (b) These calculations also indicate that the angular momentum spectrum P_l can be rather well approximated for our purposes by the classical approximation

$$P_l dl=(2l/l_{\max}{}^2)dl \quad \text{for} \quad l \ l_{\max}, \tag{2}$$

$$P_l dl=0 \quad \text{for} \quad l>l_{\max}, \tag{3}$$

and

$$l_{\max}{}^2=2\mu(E_{\text{c.m.}}-V)R^2\hbar^{-2}, \tag{4}$$

where μ is the reduced mass, V is the Coulomb barrier, and R is the sum of the radii of the colliding nuclei. We consider the spin of the target nucleus to be negligible and identify the orbital angular momentum l of the collision partners with the total angular momentum of the compound nucleus. (c) We will not attempt to calculate $\Gamma_\gamma:\Gamma_n:\Gamma_c$ because this requires a description of the spin dependence of nuclear level density for conditions in which the classical approximation is not valid.[8,9] Instead, we assume that f_n for Tb compound nuclei is very similar to that for Dy [see Eq. (1)]. (d) The effect of change in the angular momenta accompanying neutron and photon emission is approximated by the dashed line in Fig. 3 as discussed in the next paragraph. (e) Macfarlane has assigned spins 5/2 and 11/2 to Tb^{149g} and Tb^{149m}, respectively.[2]

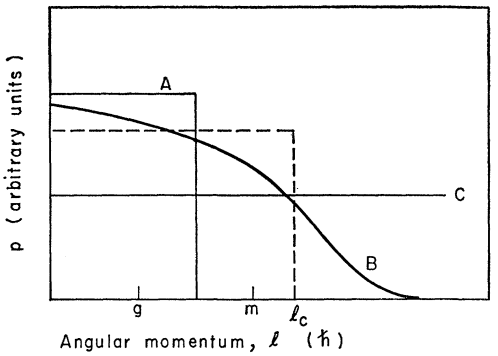

FIG. 3. The relative probability p of forming Tb^{149g} from Tb compound nuclei of angular momentum l, at a given excitation energy. The spins of the ground and metastable states of Tb^{149} are denoted by g and m. Curve A is for the limiting case of infinitely strong preference for minimal spin change. Curve C is for all spin changes equally probable. Curve B is for strong favoring of small spin changes and is approximated by the dashed rectangle of base l_c.

Consider the various angular-momentum states of compound systems of a given excitation energy. The relative probability p of forming Tb^{149g} as a function of l is shown in Fig. 3. Curve A is the limiting case of infinitely strong preference for zero or minimal spin change. Curve C is the limiting case if all possible spin changes are equally probable. Curve B is a schematic representation of the assumption that small spin changes ($\lesssim 4\hbar$) are strongly favored in each step of the de-excitation cascade. If small spin changes per emitted particle are sufficiently strongly favored, and curve B, therefore, has a relatively sharp break, then we can approximate the curve by a rectangle of base l_c (the dashed curve in Fig. 3) as follows:

$$p=\text{const} \quad \text{for} \quad l<l_c, \tag{5}$$

and

$$p=0 \quad \text{for} \quad l>l_c. \tag{6}$$

The absolute value of p must depend on excitation energy, but if the dependence on l is sharp, then the value of l_c may vary only slowly with excitation energy. We test for this possibility by trying to select one value of l_c that will systematize our results for the different reactions. If one value of l_c can be selected, then only that part of the reaction cross section $\sigma(<l_c)$ leading

[7] A very similar discussion of these results was presented by T. D. Thomas at the Gordon Research Conference for Nuclear Chemistry, June 1962.

[8] T. Ericson, in *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1960), Vol. 9, p. 425.

[9] J. R. Grover, Phys. Rev. **127**, 2142 (1962); **128**, 267 (1962).

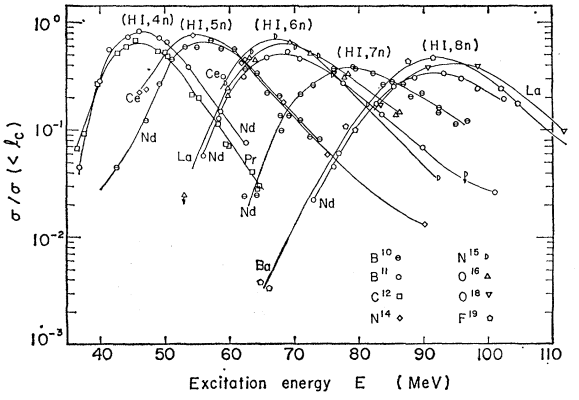

FIG. 4. Cross section σ for Tb^{149g} divided by cross section $\sigma(<l_c)$ for forming compound systems of angular momentum less than l_c. The radius parameter is 1.5 F and l_c is $8\hbar$.

to $l<l_c$ contributes to the production of Tb^{149g}. Using the classical approximation, we have

$$\sigma(\ l_c)=(l_c/l_{\max})^2\sigma_R. \tag{7}$$

We use two criteria for testing these assumptions and selecting a value of l_c: (a) The values of $\sigma/\sigma(\ l_c)$ (σ denotes cross section for Tb^{149g}) shall be less than unity but greater than f_n for Dy [see Eq. (1)]. (b) For a given value of x, the different (HI,xn)Tb^{149g} reactions shall have very similar dependence of $\sigma/\sigma(\ l_c)$ on excitation energy. The physical meaning of criterion (a) is as follows: charged particle emission (from nuclear evaporation and noncompound nucleus reactions) is less probable for Tb systems of relatively low spin than for Dy systems of a very wide range of angular momenta. Criterion (b) is the classical condition for independence of decay probability on mode of formation of the compound system.

In Fig. 4 we show values of $\sigma/\sigma(<l_c)$ for the (HI,xn)Tb^{149g} reactions, where l_c was taken as $8\hbar$ and the radius parameter was taken as 1.5F. This procedure has essentially removed the decreasing trend of the fractional cross sections with x. Also, the reactions forming the compound nuclei Tb^{153} and Tb^{157} are brought into closer correlation. The result for any other value of l_c is obtained simply by multiplying all curves in Fig. 4 by a factor proportional to l_c^2. Our two criteria would be reasonably well satisfied within the various errors by l_c values of $7.5\pm1.5\hbar$. The small value of l_c coupled with $l_{\max}$ values that vary from 7 to 90 implies that curve B in Fig. 3 is relatively sharp. In other words, the probability must be small for a compound system of very high spin to decay to the low-spin isomer of Tb^{149}.

In summary, this correlation gives evidence that 4.0-min Tb^{149m} "shields" 4.1-h Tb^{149g} from production by high-spin compound nuclei.

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APPENDIX

In many of these experiments Tb^{149g} recoil atoms were stopped in 0.00025-in. Al foils and the radioactivity of these foils was measured by 2π ionization chambers. Absorption corrections (approximately 5% to approximately 20%) for the α radiation were applied as follows.

We assume that all recoil atoms were formed at the center of the target layer (target thickness in mg/cm^2 is denoted by W) and recoiled along the beam direction a distance given by the average range R_0. The average range values were taken from the range-energy curve in reference 4. We assume that the fraction of the α radiation that is absorbed is given by d/r, where d is the average distance from the surface of the foil to the Tb^{149g} atoms, and r is the effective range of the 3.95-MeV α particles. If the radioactivity was detected from the surface of the catcher foil that faced the target, then we have

$$d=R_0-0.54W/2,$$

where 0.54 is an estimate of the stopping power of the target layer relative to that of Al.[4] If the α radiation from the other face was measured, then we have

$$d=t-R_0+0.27W,$$

where t is the thickness of the catcher foil.

The value of r was determined to be 4.04 mg/cm^2 Al by separate measurements of the α radiation from each surface of several foils. The ratio Q of the counting rates from the two faces of a given catcher foil is given by

$$Q=[1-(R_0-0.27W)/r][1-(t-R_0+0.27W)/r]^{-1}.$$

The two different expressions for d were used. The effective range r of the α particles represents that thickness of Al required to reduce the energy of the α particles below the threshold for detection. The threshold values for all counters were set by requiring equal counting rates for thick U metal standards.