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Distribution Measurements

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A Test of the Independence Hypothesis by
Angular Distribution Measurements*

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Angular distribution measurements of ^{137m}Ce and ^{137g}Ce from the reactions $^{136}\text{Ba}(^4\text{He}, 3n)$ and $^{137}\text{Ba}(^3\text{He}, 3n)$ have been made over the energy range from 33.3 to 43.6 MeV and from 20.3 to 32.5 MeV, respectively. The results have been analyzed to give the average total kinetic energy of the neutrons and photons emitted in the reactions. Comparison of these quantities at the same excitation energy and angular momentum of the compound nuclei shows that they are equal within the limits of error and thereby confirms the independence hypothesis. The observed differences in the average photon energies for reactions leading to ^{137m}Ce and ^{137g}Ce are quantitatively related to the difference in the average angular momentum of compound nuclei leading to each isomer.

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I. Introduction

In the preceding paper¹ it was shown that the $^{136}\text{Ba}(^4\text{He},3\text{n})$ and $^{137}\text{Ba}(^3\text{He},3\text{n})$ reactions proceed via compound nucleus formation at intermediate energies. The compound nuclear character of these reactions was established by average range measurements of the ^{137}Ce recoils and corroborated by excitation function and isomer ratio measurements.

Since both reactions involve the formation of the same compound nucleus they can be used to test the independence hypothesis, which states that the decay of a compound nucleus is independent of its mode of formation and depends only on its excitation energy and angular momentum. Previous tests of the independence hypothesis have involved the measurement of excitation functions^{2,3} and charged particle spectra.⁴ The present work is based on the measurement of the angular distributions of the $^{137\text{m,g}}\text{Ce}$ recoil nuclei. The angular distribution results can be analyzed by methods described elsewhere^{5,6} to give values of the average total kinetic energy of the emitted neutrons T_n and the average total energy dissipated by photons T_γ . If the decay of the compound nucleus is independent of its mode of formation, there should be no differences in the T_n or T_γ values from the ^3He and ^4He reactions provided the comparison is made at the same excitation energy and angular momentum of the compound nucleus. The study of reactions involving isomer formation is particularly useful in connection with this requirement since, as will be shown later, it permits the equalization of both these quantities in the formation of the low-spin product. Measurement of the angular distribution of isomeric products also gives information on the effect of angular momentum on the deexcitation process.

In this paper we present our results from a comprehensive study of the angular distributions of $^{137m,g}\text{Ce}$ from the ($^4\text{He}, 3n$) and ($^3\text{He}, 3n$) reactions over the compound nucleus excitation energy range from 33.7 to 46.8 MeV.

II. Experimental

The experimental procedure and apparatus for angular distribution studies have been described in previous reports from this laboratory.^{7,8} Thin targets are irradiated in an evacuated chamber. The product nuclei recoil out of the target in a forward cone and are stopped by a circular catcher foil placed a known distance from the target. The catcher foil is cut into concentric rings each of which corresponds to a particular angular interval and the radioactivity of each ring is assayed following chemical separation of cerium.

The targets were prepared from enriched ^{137}Ba (89.6%) and ^{136}Ba (97.2%).⁹ Thin targets, ranging from 5 to 15 $\mu\text{g}/\text{cm}^2$, were prepared by molecular plating.¹⁰ Fifty microliters of a solution containing 10 to 20 μg of $\text{Ba}(\text{NO}_3)_2$ were placed in a cell containing approximately 20 ml isopropyl alcohol. A rotating platinum electrode assembly was used to deposit $\text{Ba}(\text{NO}_3)_2$ on 1 mil Al foil of high purity (99.999%). The Al foil served as the cathode and the cell was operated at 250 V for approximately 1 hour.

The bombardments were carried out in the ^3He and ^4He ion beams of the Argonne National Laboratory 60-inch cyclotron. Irradiations lasted from 2 to 3 hours at beam intensities from 0.1 to 0.3 μamp . The beam was collimated before entering the chamber by 0.95 and 0.48 cm apertures.

An 0.32 cm collimator located just in front of the water cooled target holder further defined the beam. Before inserting the target foil, alignment experiments were performed to insure that the beam passed through the center of the target and catcher foils. The beam was degraded to the desired energy by placing Al foils in front of the collimators. The ^3He and ^4He energies were calculated from the incident energy and an appropriate range-energy relationship.¹¹

The catcher foil consisted of the same high-purity aluminum as the target backing. After irradiation it was cut into 10 concentric rings and Ce was separated from each foil by a previously described procedure.¹ The $^{137\text{m,g}}\text{Ce}$ disintegration rates were determined with thin NaI(Tl) detectors connected to single channel analyzers set to accept the La and Ce K-x-rays, as described in further detail in the preceeding paper.¹ The data were analyzed by a least squares program described in a previous report¹² from this laboratory.

The effects of target thickness and collimator size on the angular distribution were investigated for the ($^4\text{He}, 3n$) reaction. The target thickness effect was determined by irradiating targets of $5.5 \mu\text{gm/cm}^2$ and $10.0 \mu\text{gm/cm}^2$ at 39.2 and 39.0 MeV bombarding energy, respectively, with an 0.32 cm collimator at the target. The collimator effect was studied by irradiating a $12.0 \mu\text{gm/cm}^2$ target with an 0.16 cm collimator at 39.1 MeV. The resulting angular distributions of $^{137\text{m}}\text{Ce}$ are shown in Fig. 1. The various curves have been normalized to the same area. It is seen that there is no broadening of the angular distribution due to target thickness for the thin targets used in this experiment. The mean angles obtained for target thicknesses of $5.5 \mu\text{gm/cm}^2$ and $10.0 \mu\text{gm/cm}^2$ were 10.1° and 10.0° , respectively. The $^{137\text{g}}\text{Ce}$ angular distributions showed the same insensitivity to target thickness as those

of ^{137m}Ce . The mean recoil angle of ^{137m}Ce from the experiment with the 0.16 cm collimator was 10.8° . The widening of the angular distribution for a smaller collimator is opposite to the expected effect and must be due to an experimental uncertainty for this particular bombardment. A subsequent¹³ test of the collimation effect using the same apparatus and similar experimental conditions showed that collimation effects on the angular distributions of $^{137m,g}\text{Ce}$ are negligible.

III. Results

The angular distribution data, which are available from the authors on request, were used to obtain for each reaction the values of the mean laboratory recoil angle, $\langle\theta_L\rangle$, and those of $\langle\tan^2\theta_L\rangle$. The equations by which these quantities were calculated from the measured disintegration rates, D_i , are:

$$\langle\theta_L\rangle = \frac{\sum_i D_i \Delta\theta_i \langle\theta_i\rangle}{\sum_i D_i \Delta\theta_i} \quad (1)$$

and

$$\langle\tan^2\theta_L\rangle = \frac{\sum_i D_i \Delta\theta_i \langle\tan^2\theta_i\rangle}{\sum_i D_i \Delta\theta_i} \quad (2)$$

The summations are over the i rings of the catcher foil, $\Delta\theta_i$ is the angular interval corresponding to the i^{th} ring, and $\langle\theta_i\rangle$ and $\langle\tan^2\theta_i\rangle$ are the mean values of these quantities for the ring in question.

The results are summarized in Table I. The uncertainties in the $\langle\theta_L\rangle$ values are approximately 0.1° - 0.3° and arise from the statistical uncertainty in the disintegration rates, as well as minor errors in the chemical yield determinations and alignment procedures.

The mean angles are plotted as a function of the available energy, $E_{\text{cm}} + Q$, in Fig. 2. The Q values were determined from the nuclidic masses compiled by Mattauch et al.¹⁴ and E_{cm} is the center of mass bombarding energy. The various trends displayed by these data are readily explainable. The gradual increase of $\langle \theta_L \rangle$ with energy arises from the fact that the recoil velocity due to neutron emission increases more rapidly with energy than the velocity of the compound nucleus. For a given value of the available energy the $\langle \theta_L \rangle$ from ^3He irradiations are some 3° - 5° larger than those for ^4He . This difference is merely a consequence of the larger forward momentum imparted to the compound nucleus in ^4He bombardments. The $\langle \theta_L \rangle$ of $^{137\text{m}}\text{Ce}$ are uniformly larger than those of $^{137\text{g}}\text{Ce}$, the difference at a given energy amounting to close to 1° . This difference reflects differences in the partition of the available energy between neutrons and photons and is discussed in more detail below.

IV. Discussion

The experimental results may be analyzed to yield the values of T_n and T_γ . The mean squared velocity imparted to the residual nucleus as a result of isotropic neutron evaporation $\langle V^2 \rangle$ may be related⁶ to $\langle \tan^2 \theta_L \rangle$ by the expression

$$\langle \tan^2 \theta_L \rangle = \sum_{n=1}^{\infty} \frac{2\langle V^{2n} \rangle}{(2n+1)v_{\text{CN}}^{2n}} \quad (3)$$

The quantity v_{CN} is the velocity of the compound nucleus, obtained from the bombarding energy and conservation of momentum. In turn $\langle V^2 \rangle$ is related⁵ to the average total kinetic energy of the emitted neutrons by

$$\langle V^2 \rangle = \frac{8T_n}{(A_T + A_b + A_R)^2} \quad (4)$$

where the masses of the target, bombarding, and product

nuclei are denoted respectively by A_T , A_b , and A_R . Finally, T_Y is obtained as the difference between the total available energy and T_n :

$$T_Y = (E_{cm} + Q) - T_n \quad (5)$$

The values of $\langle V^2 \rangle$ were calculated by solving Eq. 3 by an iteration procedure. Three terms in the expansion were required to obtain $\langle V^2 \rangle$ within 1% accuracy. The use of more than one term requires the assumption that $\langle V^{2n} \rangle = \langle V^2 \rangle^n$.

The results of this analysis are summarized in Table 1. The uncertainties in the values of T_n and T_Y were estimated from the differences in the results obtained from bombardments at approximately the same energy as well as the total statistical uncertainty of the counting data. The mean uncertainty is 0.2 MeV for ^{137m}Ce and 0.6 MeV for ^{137g}Ce .

The validity of the independence hypothesis may be tested by comparing the values of T_n or T_Y derived for the two reactions at the same value of the available energy. Before presenting this comparison it is necessary to consider the differences in the angular momentum of the two systems. Since the reaction products are isomeric we are particularly interested in calculating the average angular momentum of those compound nuclei that deexcite to each isomeric state, denoted by $\langle J_c^m \rangle$ and $\langle J_c^g \rangle$.

The isomer ratio program described in the preceding paper¹ was used to perform this calculation. The basic steps of the calculation involve the evaluation of the angular momentum distribution of the compound nucleus, the residual nucleus formed after the emission of each neutron, and that formed after the emission of each but the last

γ -ray. At this point in the program the yield of excited nuclei with $J < 7/2$ is assigned to ^{137g}Ce , that of nuclei with $J > 7/2$ to ^{137m}Ce while that of nuclei with $J = 7/2$ is divided equally between the two products.

Since the angular momentum distribution of the compound nucleus is modified by neutron and photon emission and since the isomer assignment is based on this modified distribution, it is difficult to make a direct determination of $\langle J_c^m \rangle$ and $\langle J_c^g \rangle$. In order to check the sensitivity of these quantities to the deexcitation process, approximate values of $\langle J_c^m \rangle$ and $\langle J_c^g \rangle$ were determined by apportioning the angular momentum distribution between the two isomers in the manner outlined above at each step of the deexcitation process. The results for the $(^4\text{He}, 3n)$ reaction at a compound nucleus excitation energy of 42.8 MeV are summarized in Table II. The three entries refer, respectively, to the values obtained by apportioning the angular momentum distribution of the compound nucleus, that of the residual nucleus resulting from the emission of three neutrons, and that following the γ -ray cascade. It is seen that $\langle J_c^g \rangle$ is virtually independent of the method of calculation. On the other hand, the values obtained for $\langle J_c^m \rangle$ decrease as the deexcitation process proceeds. This result is a consequence of the shift of the angular momentum distribution of the compound nucleus towards lower values by the emission of neutrons and photons. The values of $\langle J_c^g \rangle$ do not reflect this trend because the low spin of ^{137g}Ce restricts its formation to the lower end of the angular momentum distribution, whose shape is rather insensitive to the effect of neutron and photon emission. Of the three methods of calculating $\langle J_c^m \rangle$ it is

clear that the first one overestimates its value while the third one underestimates it. Therefore method 2 was chosen as the best approximation to a calculation of $\langle J_c^m \rangle$ and $\langle J_c^g \rangle$.

The results of this computation are plotted in Fig. 3 for both ^3He and ^4He bombardments. It is seen that the same values of $\langle J_c^g \rangle$ are obtained for both reactions. Therefore, if a comparison of T_n and T_γ for ^{137g}Ce formation is made at the same excitation energy of the compound nuclei, the average angular momentum of the latter will also be equal. The T_n and T_γ values for the two reactions are plotted in Fig. 4. Within the limits of error, the results for ^3He and ^4He bombardments can be seen to fall on the same curve. This finding confirms the independence hypothesis for the reactions leading to ^{137g}Ce .

In the case of ^{137m}Ce formation Fig. 3 shows that $\langle J_c^m \rangle$ is larger for ^4He bombardments by 1 to 3 $\hbar$ units. One may therefore expect a difference in the T_n or T_γ values. The magnitude of this difference can be estimated on the assumption that the photon energy arises from the dissipation of the rotational energy of the compound nucleus. The rotational energy is obtained from the rigid rotor model as

$$E_{\text{rot}}^m = \frac{\langle J_c^m \rangle (\langle J_c^m \rangle + 1) \hbar^2}{2 \mathcal{J}_r} \quad (6)$$

where $\mathcal{J}_r$ is the rigid-body moment of inertia. The expected difference in T_n or T_γ values is then obtained as the difference between the rotational energies obtained for ^3He and ^4He bombardment. This difference ranges from about 0.2 MeV for $(E_{\text{cm}} + Q) = 6$ MeV to 0.6 MeV for $(E_{\text{cm}} + Q) = 20$ MeV.

The values of T_n and T_Y for reactions leading to ^{137m}Ce are shown in Fig. 5. The width of the line drawn through the points corresponds to the difference in rotational energies and is thus a measure of the expected difference between the ^3He and ^4He points. Although the experimental points do scatter somewhat more than expected from the estimated uncertainties it does appear that the T_Y values from the ^4He experiments are slightly larger than those from the ^3He ones, in agreement with the theoretical estimate. Our conclusion is that the reactions leading to ^{137m}Ce also confirm the independence hypothesis.

A more sensitive test of the relation between angular momentum and energy dissipated in photon emission than is afforded by the data in Fig. 5 can be obtained from a comparison of the T_n or T_Y values associated with the formation of ^{137m}Ce and ^{137g}Ce in the same reaction. As indicated in Fig. 3, the differences between $\langle J_c^m \rangle$ and $\langle J_c^g \rangle$ for a given projectile range from 3 to 10 $\hbar$ units so that the expected effect is much larger. As before, the expected difference in photon energies is obtained by application of Eq. 6 to the data shown in Fig. 3. The experimental values of ΔT_Y are obtained by direct subtraction of the T_Y values for ^{137g}Ce from those for ^{137m}Ce at the same bombarding energy. The results are shown in Fig. 6. Because of the very small difference in Q values for the formation of ^{137m}Ce and ^{137g}Ce , the experimental points are plotted at the average value of $E_{cm} + Q$. Within the rather large uncertainties introduced by the subtraction of T_Y values, the experimental points are in satisfactory agreement with the theoretical curve.

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TABLE I

Values of Quantities Derived from Angular Distributions

Isomer	Bombarding Energy (MeV)	$E_{\text{CM}} + Q$ (MeV)	$\langle \theta_L \rangle$ (deg)	$\langle \tan^2 \theta_L \rangle$	T_n (MeV)	T_Y (MeV)
m	43.6	17.4	11.3	.0574	$14.0 \pm .3$	$3.4 \pm .3$
g	43.6	17.7	12.4	.0673	$16.2 \pm .8$	$1.5 \pm .8$
m	41.3	15.2	10.8	.0522	$12.1 \pm .2$	$3.1 \pm .2$
g	41.3	15.5	11.7	.0603	$13.8 \pm .7$	$1.7 \pm .7$
m	39.2	13.2	10.0	.0437	$9.7 \pm .1$	$3.5 \pm .1$
g	39.2	13.5	10.8	.0503	$11.1 \pm .6$	$2.4 \pm .6$
m	39.1	13.1	10.8	.0534	$11.7 \pm .2$	$1.4 \pm .2$
g	39.1	13.4	11.0	.0542	$11.9 \pm .6$	$1.5 \pm .6$
m	39.0	12.9	10.1	.0446	$9.8 \pm .2$	$3.1 \pm .2$
g	39.0	13.2	10.5	.0466	$10.2 \pm .5$	$3.0 \pm .5$
m	36.1	10.2	9.8	.0421	$8.6 \pm .2$	$1.6 \pm .2$
g	36.1	10.5	10.1	.0442	$9.0 \pm .5$	$1.5 \pm .5$
m	33.3	7.5	9.5	.0390	$7.5 \pm .2$	$0.0 \pm .2$
g	33.3	7.8	10.1	.0434	$7.8 \pm .4$	$0.0 \pm .3$

m	32.5	20.5	16.0	.1284	$16.4 \pm .3$	$4.1 \pm .3$
g	32.5	20.8	16.9	.1440	$18.2 \pm .7$	$2.6 \pm .9$
m	28.0	16.1	15.4	.1187	$13.2 \pm .3$	$2.9 \pm .3$
g	28.0	16.4	16.2	.1336	$14.7 \pm .7$	$1.7 \pm .7$
m	27.8	15.9	15.1	.1164	$12.9 \pm .3$	$3.0 \pm .3$
g	27.8	16.2	15.3	.1174	$13.0 \pm .6$	$3.2 \pm .6$
m	27.6	15.8	15.5	.1202	$13.2 \pm .3$	$2.6 \pm .3$
g	27.6	16.1	16.1	.1315	$14.3 \pm .7$	$1.8 \pm .7$
m	24.3	12.5	15.7	.1213	$11.7 \pm .1$	$0.8 \pm .2$
g	24.3	12.8	15.9	.1242	$11.9 \pm .6$	$0.9 \pm .6$
m	20.3	8.6	15.0	.1059	$8.6 \pm .2$	$0.0 \pm .4$
g	20.3	8.9	15.2	.1082	$8.8 \pm .4$	$0.1 \pm .4$

TABLE II

Average Angular Momentum of Compound Nuclei^a
 Leading to ^{137m}Ce and ^{137g}Ce

Method of Calculation ^b	$\langle J_c^m \rangle$	$\langle J_c^g \rangle$
Method 1	13.5	2.4
Method 2	11.9	2.5
Method 3	10.0	2.5

a. The compound nucleus is formed in ^4He bombardment
 with an excitation energy of 42.8 MeV.

b. See text for explanation.

FIGURE CAPTIONS

- Fig. 1 Dependence of the angular distribution of ^{137m}Ce from the $(^4\text{He}, 3n)$ reaction on target thickness and collimator size. $\bigcirc$ = 39.0 MeV, 0.32 cm collimator, and $10.0 \mu\text{gm}/\text{cm}^2$ target; Δ - 39.2 MeV, 0.32 cm collimator and $5.5 \mu\text{gm}/\text{cm}^2$ target; $\times$ - 39.0 MeV, 0.16 cm collimator, and $12.0 \mu\text{gm}/\text{cm}^2$ target. The curves are normalized to the same area.
- Fig. 2 Average recoil angles of $^{137m,g}\text{Ce}$ from the $(^3\text{He}, 3n)$ and $(^4\text{He}, 3n)$ reactions. The circles refer to ^{137g}Ce and triangles to ^{137m}Ce .
- Fig. 3 Approximate values of the average angular momentum of compound nuclei leading to ^{137m}Ce and ^{137g}Ce . The circles refer to ^4He induced reactions and triangles to ^3He induced reactions. Open symbols show compound nuclei leading to ^{137m}Ce and shaded symbols those leading to ^{137g}Ce .
- Fig. 4 Comparison of T_n and T_γ for ^{137g}Ce from $(^4\text{He}, 3n)$ and $(^3\text{He}, 3n)$ reactions. Triangles represent ^3He and circles the ^4He reactions. Open symbols refer to T_n and shaded symbols to T_γ .
- Fig. 5 Comparison of T_n and T_γ for ^{137m}Ce from the $(^4\text{He}, 3n)$ and $(^3\text{He}, 3n)$ reactions. Symbols have same meaning as in Fig. 4. The widths of the lines drawn through the points correspond to the expected differences between ^4He and ^3He reactions.
- Fig. 6 Differences between T_γ for reactions leading to ^{137m}Ce and ^{137g}Ce . Symbols represent the experimental points and the solid line is from the theoretical calculation.

Fig 1

Fig 2

Fig 6