Isomer Ratio of Ce^{137m} to Ce^{137g} Produced in Several Charged-Particle Reactions*

R. L. KIEFER† AND K. STREET, JR.
Lawrence Radiation Laboratory
and
Department of Chemistry, University of California, Berkeley, California

(Received 1 April 1968)

The Ce^{137m}-Ce^{137g} isomer ratio is studied to obtain information on the effect of angular momentum in the de-excitation of a compound nucleus. The ratio of the formation cross section of the high-spin isomer to that of the low-spin isomer is measured as a function of energy for reactions induced by He^3, He^4, Li^7, and C^{12}. The ratio is seen to increase with increasing projectile energy and is shown to correspond to an increase in the average angular momentum. The isomer-ratio results are compared with calculations using the Huizenga-Vandenbosch theory. Using the nuclear temperature based on the Fermi-gas model and the spin cutoff factor calculated with 0.5 of the rigid-body moment of inertia, reasonable agreement was obtained with experimental results in all four reactions studied.

I. INTRODUCTION

A METASTABLE state of a nucleus differs from the ground state by at least several $\hbar$ units of angular momentum but only by a small amount of energy. Together, they form an isomer pair. In reactions proceeding by a compound-nucleus mechanism, the angular momentum introduced by the projectile increases with bombarding energy. Thus, by investigating the energy dependence of the formation cross sections of a pair of isomers formed in a compound-nucleus reaction, one could obtain information about the effect of angular momentum in the de-excitation process.

A number of authors have studied the formation cross-section ratios of nuclear isomers.[1–14] A successful method of predicting isomer ratios has been formulated by Huizenga and Vandenbosch,[2] and modified by Need[15] and Dudey.[16] In only a few cases, however, have heavy ions ($A>4$) been used to initiate the reaction.[13,14] The heavy ion is capable of introducing many $\hbar$ units of angular momentum to the compound nucleus. If the compound nucleus de-excites to one of two nuclear isomers, the large value of the angular momentum should favor formation of the high-spin isomer.

This investigation was undertaken to compare the energy dependence of the isomer ratio formed in a heavy-ion reaction to that formed in a corresponding light-particle reaction. The isomer pair $Ce^{137g,137m}$ was chosen because the decay scheme appeared to be well characterized, the half-lives were convenient for investigation, and there were suitable stable isotopes available as target materials.

II. EXPERIMENTAL PROCEDURE

The isomer pair was produced in four different reactions all leading to Ce^{137} through the compound nucleus Ce^{140}*. The reactions were $Ba^{137}(He^3,3n)Ce^{137}$, $Ba^{136}(He^4,3n)Ce^{137}$, $Cs^{133}(Li^7,3n)Ce^{137}$, and Te^{128}-

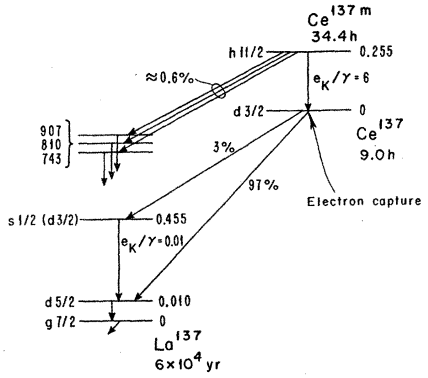

FIG. 1. Decay scheme of Ce^{137} taken from Ref. 17.

* Based on work performed under the auspices of the U. S. Atomic Energy Commission.

† Present address: Department of Chemistry, College of William and Mary, Williamsburg, Va.

[1] James Wing, Argonne National Laboratory Report ANL-6598, 1962 (unpublished). This is a tabulation of isomer ratio data up to 1962.

[2] J. R. Huizenga and R. Vandenbosch, Phys. Rev. **120**, 1305 (1960).

[3] R. Vandenbosch and J. R. Huizenga, Phys. Rev. **120**, 1313 (1960).

[4] A. J. Cox, Nucl. Phys. **49**, 577 (1963).

[5] J. L. Need and B. Lindner, Phys. Rev. **129**, 1298 (1963).

[6] R. Vandenbosch, L. Haskin, and J. C. Norman, Phys. Rev. **137**, B1134 (1965).

[7] C. Riley, K. Ueno, and B. Lindner, Phys. Rev. **135**, B1340 (1964).

[8] C. Riley and B. Lindner, Phys. Rev. **134**, B559 (1964).

[9] B. Keisch, Phys. Rev. **129**, 769 (1963).

[10] C. T. Bishop, H. K. Vonach, and J. R. Huizenga, Nucl. Phys. **60**, 241 (1964).

[11] C. T. Bishop, J. R. Huizenga, and J. P. Hummel, Phys. Rev. **135**, B401 (1964).

[12] T. Matsuo, J. M. Matuszek, N. D. Dudey, and T. T. Sugihara, Phys. Rev. **139**, B886 (1965).

[13] D. W. Seegmiller, Lawrence Radiation Laboratory Report UCRL-10850, 1964 (unpublished).

[14] V. V. Bredel, B. A. Gvozdev, and V. A. Fornichev, Zh. Eksperim. i Teor. Fiz. **45**, 904 (1963) [English transl.: Soviet Phys.—JETP **18**, 622 (1964)].

[15] J. L. Need, Phys. Rev. **129**, 1302 (1963).

[16] N. D. Dudey and T. T. Sugihara, Phys. Rev. **138**, B896 (1965).

$(C^{12},3n)Ce^{137}$. The decay scheme of Ce^{137} from Ref. 17 is shown in Fig. 1.

Targets enriched to 92.9% in Ba^{136} were made by successive application of the nitrate solution onto 1-mil Cu foils with heating between each application. In a similar manner, targets enriched to 81.9% in Ba^{137} were made with 0.5-mil Cu foils. Tellurium metal enriched to 96.4% in Te^{128} was electroplated onto 0.05-mil Cu foil from a very dilute nitric acid solution. Targets of natural cesium (100% Cs^{133}) were made by evaporating $CsNO_3$ under vacuum onto foils of 0.2-mil copper.

Eight target foils were stacked together for each irradiation. The beam was degraded by each successive foil as it passed through the stack. The beam energy at each foil was determined from the range-energy relationships of Williamson and Boujot.[18] The He^4 irradiations were performed at the Berkeley 60-in. cyclotron. The rest were performed at the Berkeley heavy-ion linear accelerator. In all cases, a copper target holder served as a Faraday cup to measure the beam current.

After irradiation, the cerium produced in the reaction was chemically separated from the target and backing material using the solvent extraction procedure of Glendenin *et al.*[19] The cerium was precipitated as the oxalate, dried, weighed as $Ce_2(C_2O_4)_3 \cdot 9H_2O$ and mounted on aluminum counting plates.

The relative and absolute amounts of the Ce^{137} isomers present were established by counting the 445-keV γ ray characteristic of the ground-state decay (Fig. 1). The detector was a 3-in.$\times$3-in. sodium iodide unit coupled to a 400-channel pulse-height analyzer. The decay curves resulting from the He^3 and He^4 bombardments were analyzed with the aid of a least-squares computer program to give the amount of each isomer present at the end of bombardment. The computer program treated each datum as having equal statistics. This was generally the case. However, in the samples irradiated near 50 MeV with C^{12} and Li^7, contribution from the 511-keV radiation from Ce^{135} was large and the counting statistics did not always remain constant. These data were analyzed by a graphical method similar to that proposed by Biller.[20] For consistency, all of the data obtained from the C^{12} and Li^7 irradiations were analyzed in this manner. A comparison of the two methods of analysis with samples produced at lower energies showed them to be compatible.

The activity of the metastable state was also determined by analysis of the 255-keV isomeric transition. However, in every case, it was about 20% larger than the same quantity determined using the 445-keV peak. It is possible that either the conversion coefficient of the 255-keV peak or the branching ratio to the 445-keV peak is in error. These decay scheme uncertainties cancel out when the isomer ratio is determined from the analysis of the 445-keV peak.

[17] G. T. Danby, J. S. Foster, and A. L. Thompson, Can. J. Phys. **36**, 1487 (1958).

[18] C. Williamson and J. P. Boujot, Centre d'Études Nucleaires de Saclay, 1962 (unpublished).

[19] L. E. Glendenin, K. F. Flynn, R. F. Buchanan, and E. P. Steinberg, Anal. Chem. **27**, 59 (1959).

[20] W. F. Biller, University of California Radiation Laboratory Report UCRL-2068, 1952 (unpublished).

III. RESULTS AND DISCUSSION

The ratio of the formation cross section of Ce^{137m} to that of Ce^{137g} as a function of excitation energy for all of the reactions is shown in Fig. 2. The limits of error include the standard deviations of the counting rates and uncertainties in the background subtraction. All of the data are plotted together in Fig. 3. At a given excitation energy, the isomer ratio for each reaction is different. In Fig. 4, the same data are plotted as a function of the average angular momentum $\langle l \rangle$. At a given value of $\langle l \rangle$, where overlap of the data occurs, the isomer ratio measured in the various reactions are all close to a common value. Thus, in the region studied, the isomer ratio definitely varies with the average angular momentum in the compound nucleus. The values of $\langle l \rangle$ were calculated by using the parabolic approximation to the real part of the optical-model potential.[21]

The relative excitation functions for these reactions were measured. The approximate energy at the peak of the excitation function for each reaction is shown in Table I. The energy values represent the average between the excitation function peaks of the two isomers.

It was found that the value of the isomer ratio increased rapidly at energies corresponding to those above the peak of the excitation function. The angular momentum however does not rise as sharply in this region. Above the peak of the excitation function, competition occurs between the xn and $(x+1)n$ reactions. In levels of the compound nucleus with high angular angular momentum, γ-ray emission would probably compete with neutron emission even though the latter is energetically possible.[22] The xn reaction would then arise from the high angular momentum levels and the cross-section ratio of high-spin to low-spin isomers would increase sharply. This has been noted by other authors.[7,11] The effect is seen most clearly in the C^{12}-induced reaction [Fig. 2(d)], where the highest values

TABLE I. Approximate energy at the peak of the excitation function for each reaction.

Reaction	Excitation energy (MeV)	Projectile energy (MeV)
$Ba^{137}(He^3,3n)Ce^{137}$	40	25
$Ba^{136}(He^4,3n)Ce^{137}$	37	37
$Cs^{133}(Li^7,3n)Ce^{137}$	44	31
$Te^{128}(C^{12},3n)Ce^{137}$	46	50

[21] T. D. Thomas, Phys. Rev. **166**, 703 (1959).

[22] J. R. Grover, Phys. Rev. **123**, 267 (1961); J. R. Grover, *ibid.* **127**, 2142 (1962).

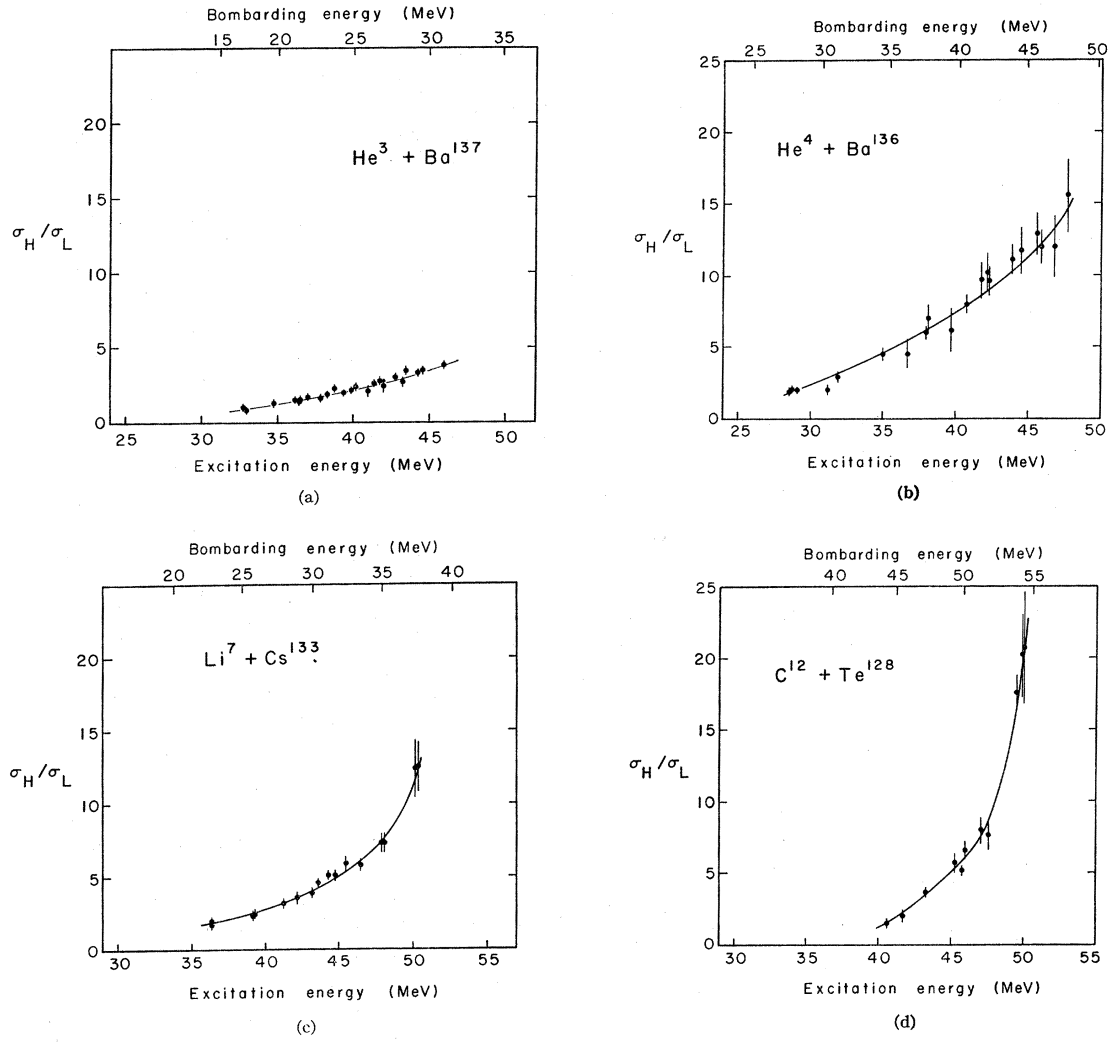

FIG. 2. Experimentally determined isomer ratios as a function of the excitation energy of the compound nucleus. The upper scale shows the laboratory energy of the projectile. The solid line connects the points.

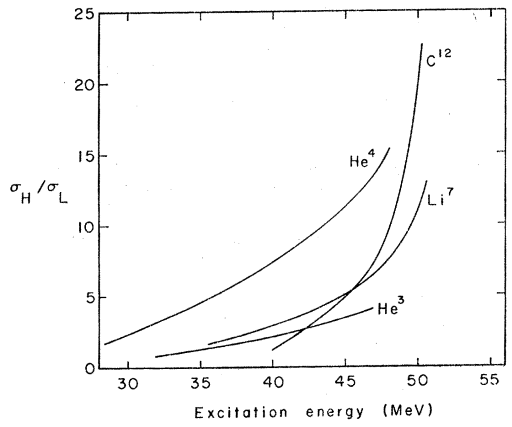

FIG. 3. Isomer ratios of the four reactions studied shown together as a function of the excitation energy.

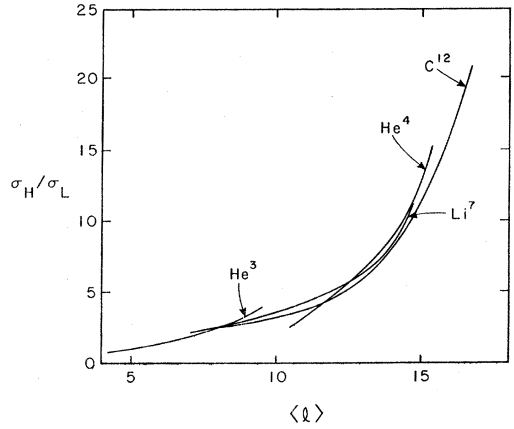

FIG. 4. Isomer ratios of the four reactions studied shown together as a function of the average angular momentum.

of angular momentum are reached. The He^3-induced reaction, which has little angular momentum, shows no sharp increase. At the peak of the excitation function, the effect of the competition between γ-ray emission and neutron emission on the isomer ratio should be minimized, since competing reactions are minimized. Figure 5 shows the isomer ratio at an energy corresponding to the peak of the excitation function for all four reactions as a function of $\langle l \rangle$. The relationship is nearly linear over the range of $\langle l \rangle$ covered by our experiments. Thus, when the γ-ray-neutron competition is minimized, the formation cross-section ratio Ce^{137m}/Ce^{136g} increases linearly with the average angular momentum given to the compound nucleus Ce^{140*}.

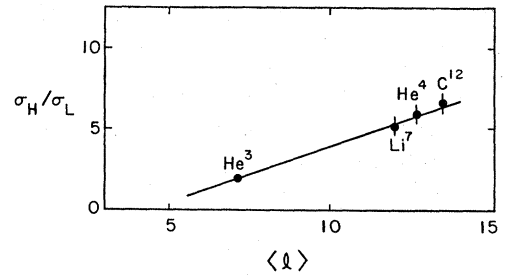

FIG. 5. Isomer ratio at an energy corresponding to the peak of the excitation function for each reaction studied as a function of the average angular momentum.

IV. CALCULATIONS

Calculations were performed using the formalism of Huizenga and Vandenbosch, in which the spin distribution is calculated for the excited compound nucleus and for each step in the de-excitation.[2] The results are shown in Fig. 6. The last γ ray is assumed to populate either the metastable or ground state, depending upon which transition has the smaller spin change. In the case of Ce^{137}, emissions of the last γ ray from a $J=\frac{7}{2}$ state should populate the $\frac{11}{2}-$ metastable and the $\frac{3}{2}+$ ground

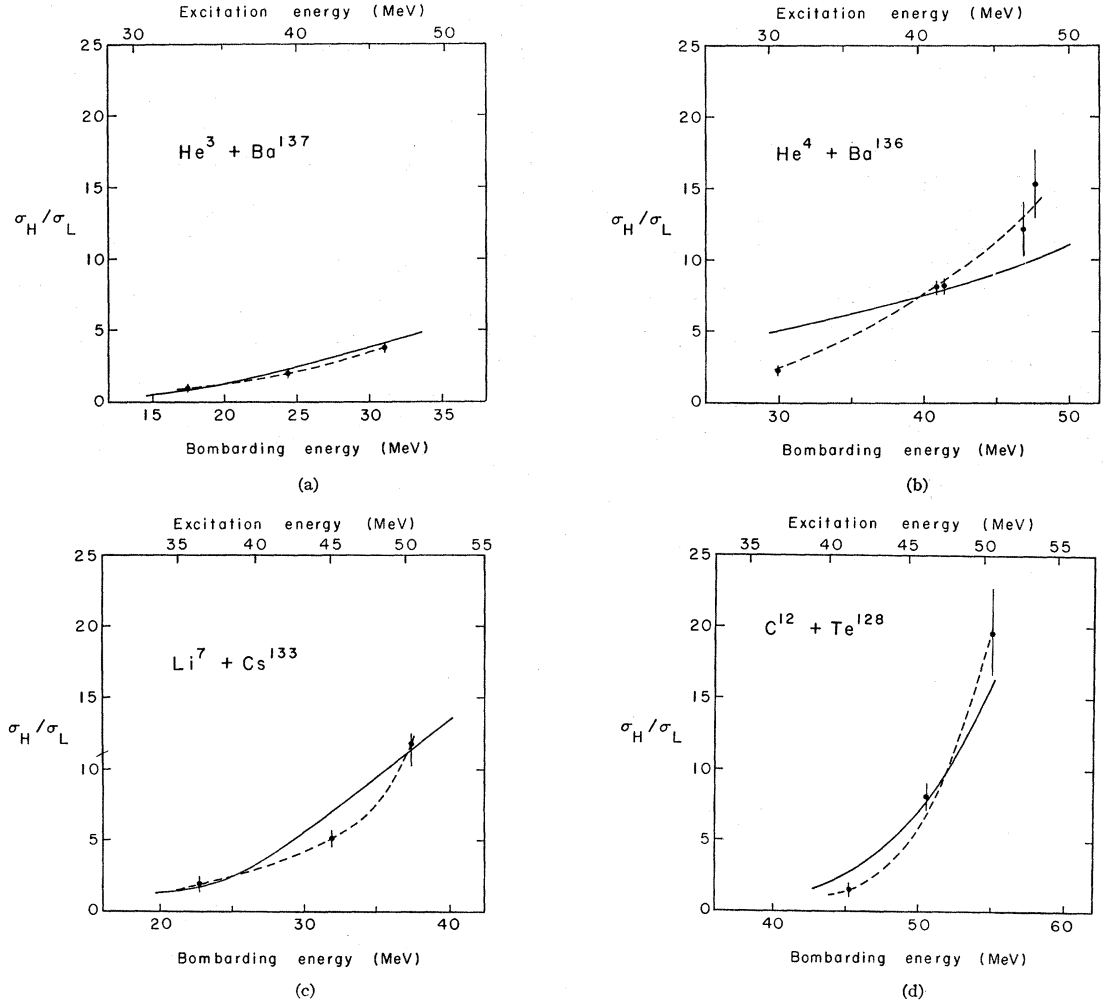

FIG. 6. Comparison of the experimentally determined isomer ratios with calculations based on the Huizenga-Vandenbosch theory. The experimental values are represented by the dashed lines. The upper scale shows the excitation energy of the compound nucleus.

states with equal probability. However, a study of the energy levels of Ce^{139}, which also has an $\frac{11}{2}^-$ metastable state and a $\frac{3}{2}^+$ ground state, revealed that all of the low-lying levels with the exception of the metastable state had positive parity.[23] By analogy, it was assumed that the lowest-lying $J=\frac{7}{2}$ state in Ce^{137} had positive parity and decayed entirely to the ground state by an $E2$ transition, since it would occur faster than an $M2$ transition to the metastable state.

In all of the calculations, transmission coefficients for outgoing neutrons were taken from Feld *et al.*[24] and those for the incoming projectile were calculated using the parabolic approximation.[21]

It was assumed that the energy carried off by the emitted neutrons was the average kinetic energy. This approximation has been found to be quite reasonable.[2] The values of the average energy were taken from the data of Simonoff and Alexander.[25] The results obtained using these data were consistent with similar calculations using the mean kinetic energy given by evaporation theory, $2(E^*/a)^{1/2}$, where E^* is the excitation energy and a is the level density parameter taken as $A/8$.

The average number of γ rays in the cascade was taken to be[26]

$$\bar{N}_\gamma=[1/(l+1)](aE^*)^{1/2}.$$

Results obtained using this relationship were similar to those obtained using the average value of 1.2 MeV/γ found experimentally by Mollenauer.[27] The value of the level density parameter a was taken as $A/8$. All of the γ rays were assumed to be dipole.

The most sensitive parameter in the calculation is the spin cutoff factor σ. It has been shown that for nucleons moving independently in an infinite square-well potential,[28,29]

$$\sigma^2=\mathcal{J}_r t/\hbar^2,$$

where $\mathcal{J}_r$ is the rigid-body moment of inertia and is equal to $\frac{2}{5}M_nR^2A$, with M_n the nucleon mass and R the nuclear radius taken as $1.2A^{1/3}$ F. The thermodynamic temperature t is calculated from the equation of state of the Fermi-gas model:

$$E^*=at^2-t.$$

The best results were obtained when σ was calculated using 0.5 of the rigid-body moment of inertia. These are the results shown in Fig. 6. Calculations using a σ based on a simple pairing model[30] and on a superconducting model[31] were also performed but were not found to duplicate the experimental results as well. Vonach *et al.*[32] have done a more thorough interpretation of isomer ratios using a Fermi-gas model and a superconductor model. They conclude that the agreement between calculations and experimental results is no better using the superconductor model than when the Fermi-gas model is used.

ACKNOWLEDGMENTS

We are indebted to Dr. Robert Vandenbosch for making the isomer-ratio program available to us. The assistance of the Health Chemistry Group and the accelerator crews is gratefully acknowledged.

[23] R. H. Fulmer, A. L. McCarthy, and B. L. Cohen, Phys. Rev. **128**, 1302 (1962).
[24] B. T. Feld, H. Feshbach, M. L. Goldberger, H. Goldstein, and V. F. Weisskopf, Atomic Energy Commission Report NYO-636, 1951 (unpublished).
[25] G. N. Simonoff and J. M. Alexander, Phys. Rev. **133**, B93 (1964).
[26] N. M. Strutinski, L. V. Groshev, and M. K. Akimova, Nucl. Phys. **16**, 657 (1960).
[27] J. F. Mollenauer, Phys. Rev. **127**, 867 (1962).
[28] H. A. Bethe, Rev. Mod. Phys. **9**, 84 (1937).
[29] C. Bloch, Phys. Rev. **93**, 1094 (1954).
[30] D. W. Lang and K. J. LeCouteur, Nucl. Phys. **14**, 21 (1959).
[31] D. W. Lang, Nucl. Phys. **42**, 353 (1963).
[32] H. K. Vonach, R. Vandenbosch, and J. R. Huizenga, Nucl. Phys. **60**, 70 (1964).