PHYSICAL REVIEW C VOLUME 1, NUMBER 6 JUNE 1970

Spontaneous Fission Isomerism in Uranium Isotopes *

K. L. Wolf, † R. Vandenbosch, P. A. Russo, M. K. Mehta, ‡ and C. R. Rudy
University of Washington, Seattle, Washington 98105
(Received 9 February 1970)

A search for spontaneous fission isomerism in uranium and neptunium isotopes has resulted in the identification of a new isomer in ^{238}U with a half-life of 195 ± 30 nsec. A measurement of the half-life of previously reported ^{236m}U gives a value of 105 ± 20 nsec. Excitation functions have been determined for the production of the spontaneous fission isomers of ^{236}U and ^{238}U by deuteron bombardment of ^{236}U and ^{238}U targets. Isomer ratios have also been estimated. The low values of the isomer ratios and the failure to observe delayed fission in lighter uranium isotopes and in neptunium isotopes suggest that γ decay may be the dominant mode of decay of shape isomers in this mass region.

I. INTRODUCTION

Spontaneous fission isomerism is now known to exist in a large number of americium[1] and plutonium[2-5] isotopes, but only one example of isomerism in the uranium isotopes has been established,[3] and less conclusive evidence for isomerism in one neptunium isotope has been reported.[3] The purpose of this investigation was to search for, identify, and characterize spontaneous fission isomers in uranium and neptunium isotopes which could be produced by 13- to 21-MeV deuteron bombardment of uranium isotopes.

II. EXPERIMENTAL PROCEDURES

200–500-μg/cm^2 targets of ^{234}U, ^{235}U, ^{236}U, and ^{238}U were bombarded with 13–21-MeV deuterons from the University of Washington 60-in. cyclotron and the University of Washington two-stage Tandem Van de Graaff accelerator. The energy of the cyclotron deuteron beam was varied by use of degrader foils. Detection of delayed fission and half-life determinations were made by a direct electronic measurement of the elapsed time between arrival of a burst of particles at the target and the time of fission decay. The time distribution of fission events is determined by using fast signals from a semiconductor fission detector, and a signal from the cyclotron oscillator or Van de Graaff beam chopper to start and stop, respectively – a time-to-amplitude converter. The timing of complementary fragments in a fission event was measured independently in order to eliminate effects due to pileup in the detectors and electronics. The experimental details are discussed elsewhere.[4,6]

Initial experiments were performed with time intervals which were dictated by the oscillator of the cyclotron which produces beam packets of approximately 2-nsec duration at 87-nsec intervals. The delayed fissions that were observed with ^{235}U, ^{236}U, and ^{238}U targets were so long-lived that half-life determinations were not possible. Excitation functions were taken with the 87-nsec intervals, but half-life determinations were made by use of an electrically driven beam chopper[7] which deflects two out of each three beam bursts, and thus increases the time between bursts to approximately 262 nsec. This time interval was satisfactory for 110-nsec ^{236m}U, but marginal for 200-nsec ^{238m}U. A longer time interval between beam bursts was desirable in order to make a more precise determination of the half-life and to check for longer-lived contributions from other isomers. Therefore, the Tandem Van de Graaff accelerator was used by chopping and bunching[8] the deuteron beam from the ion source into packets of 50-nsec width at 760-nsec intervals. The bombarding energy was chosen to be the maximum obtainable from this accelerator, which was between 17.8 and 18.0 MeV. A small background of prompt fission between the beam bursts was present in the Van de Graaff experiments owing to the presence of very weak components in the ion source beam of slightly different energies from that of the primary beam. These arise from the variety of positive atomic and molecular ions extracted prior to charge exchange to produce the H^- ions eventually accelerated. The 20° magnet between the ion source and the accelerator eliminates most, but not all, of these components. The energy differences resulted in the weak components having different flight times from that of the primary beam between the chopper and the target. The magnitude of the background between two successive beam pulses was determined using a ^{232}Th target, as ^{232}Th bombarded with 13–21-MeV deuterons at the cyclotron produced no isomers. The background was approximately 15% at early times and appreciably larger after several half-lives.

III. RESULTS

Excitation functions for the delayed fission resulting from ^{236}U and ^{238}U bombardments at the cyclotron were taken in approximately 2-MeV steps, and are illustrated in Fig. 1. Half-life determinations were made for ^{236}U and ^{238}U at 21 MeV using the cyclotron, and at 18 MeV using the Van de Graaff. The two determinations were consistent within their uncertainties, yielding final values of 105 ± 20 nsec for the ^{236}U target, and 195 ± 30 nsec for the ^{238}U target. The decay curves are shown in Fig. 2. No delayed fissions were observed for the ^{234}U target at 21 MeV. The results are summarized in the table. The evidence for the mass assignments are discussed below.

Lark *et al.*[3] have searched for fissioning isomers in several uranium and neptunium isotopes by bombarding uranium targets with 13-MeV protons and 11–13-MeV deuterons. The only fissioning isomer that was identified in this region was (130 ± 30)-nsec ^{236m}U which was produced by the ^{235}U(d, p) reaction. This assignment was confirmed by observing coincidences between protons and delayed fissions. An isomer with a half-life of 300 ± 100 nsec produced in deuteron bombardment of ^{238}U was tentatively identified as ^{239m}Np. Metag *et al.*[5] have also observed ^{236m}U, deducing a half-life of 70 nsec. We have observed delayed fission in deuteron bombardments of ^{235}U, ^{236}U, and ^{238}U targets. Bombardment of a ^{235}U target produced nearly equal isomer yields at deuteron energies of 16 and 21 MeV, as expected in a (d, p) reaction. Therefore, the delayed fission that was observed with a ^{235}U target is probably ^{236m}U, which is consistent with the identification of Lark *et al.* The excitation functions for ^{236}U and ^{238}U targets are quite different from the ^{235}U case. The delayed fission yields for ^{236}U and ^{238}U increase rapidly with deuteron bombarding energy above 16–17 MeV as shown in Fig. 1. The half-life of the isomer that was produced with ^{236}U target deuteron energies of 18 and 21 MeV was found to be 105 ± 20 nsec. This half-life is in agreement with the (130 ± 30)-nsec half-life of ^{236m}U reported by Lark *et al.*[3] This suggests that we have observed the decay of ^{236m}U which was produced by a (d, d') or (d, pn) reaction. The spallation cross section for a (d, d') or (d, pn) reaction cannot be measured, because the final nucleus is the same as the target. Measurements[9] of the proton energy spectrum for

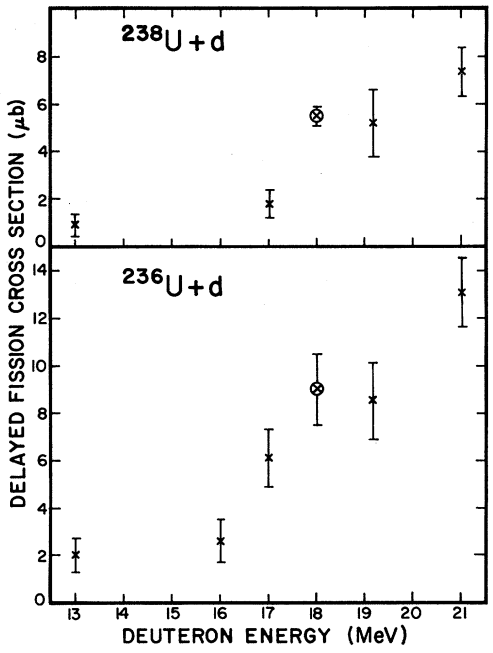

FIG. 1. Excitation functions for production of delayed fission for deuteron bombardment of ^{236}U and ^{238}U. The points at 18 MeV were determined in a different experiment using the Van de Graaff rather than the cyclotron.

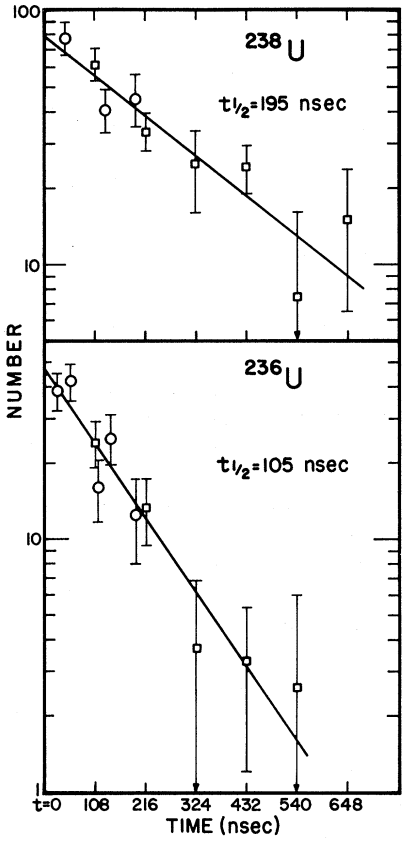

FIG. 2. Decay curves for ^{236m}U and ^{238m}U. The squares correspond to time bins of width equal to the distance between points, and were taken at 18 MeV using the Van de Graaff. The circles were obtained for deuteron energies between 19 and 21 MeV using the cyclotron, and have been normalized to the Van de Graaff data.

TABLE I. Summary of results.

Isomer	Reaction	Half-life (nsec)	Delayed fission cross section at 21 MeV (μb)	Isomer ratio
^{238}U	^{238}U(d,pn)	195 ± 30	7	5×10^{-5}
^{236}U	^{236}U(d,pn)	105 ± 20	13	9×10^{-5}
^{234}U (not observed)	^{234}U$+d$	if $t_{\frac{1}{2}} \geq 30$, $\sigma \leq 2$		
	^{232}Th$+\alpha$	if $t_{\frac{1}{2}} \geq 20$, $\sigma \leq 0.1$ if $t_{\frac{1}{2}} \geq 2$, $\sigma \leq 0.9$		

21-MeV deuteron bombardment of Au and Th show high yields of protons of low enough energy for the residual nucleus to emit a neutron and provide the estimated 2–3-MeV excitation energy of the isomeric state. Figure 3(a) shows the energy spectrum of protons at 90° for a ^{232}Th target. This spectrum is expected to be very similar to that for ^{236}U and ^{238}U, in view of the qualitative similarity[9] of the proton spectra for Au and Th targets. The vertical line indicates the proton energy below which there is sufficient energy to populate an isomeric state 2.5 MeV above the target-nucleus ground state. Some of these protons, however, are associated with Coulomb or nuclear dissociation of the deuteron rather than (d,p) stripping.[10,11] That stripping does occur, leaving considerable

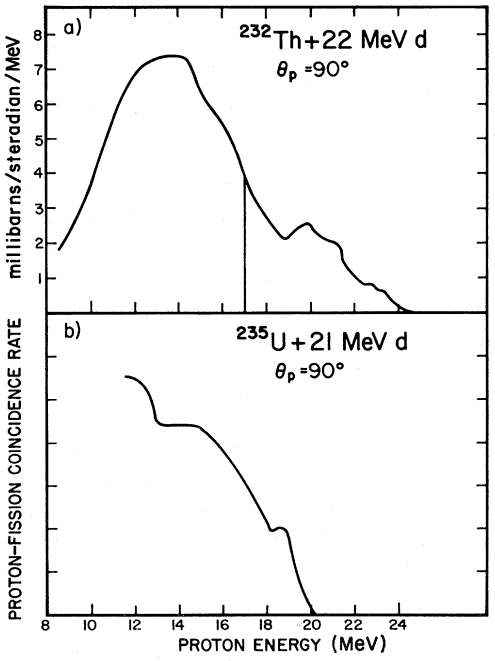

FIG. 3. (a) Proton energy spectrum for 22-MeV deuteron bombardment of ^{232}Th. Energy resolution of detector was approximately 0.5 MeV. (b) Relative coincidence rate for ^{235}U(d,pf) reaction with 21-MeV deuterons.

excitation energy in the residual nucleus, can be demonstrated by observing fission from the residual nucleus in coincidence with protons. Measurements performed at this laboratory and reported elsewhere[12] indicate that the (d,pn) reaction, leaving the residual nucleus highly excited, has a sizable cross section. Figure 3(b) is a plot of the proton fission fragment coincidence rate versus proton energy for ^{235}U bombarded with 21-MeV deuterons. The vanishing of the coincidence rate for proton energies above 20 MeV is a result of insufficient excitation energy for the residual nucleus to fission. The rise in the coincidence rate at low proton energies ($E_p \sim 12.5$ MeV) is associated with the onset of the (d,pnf) reaction, allowing an additional contribution from fission following evaporation of a neutron. We therefore conclude that the (d,pn) reaction provides a reasonable mechanism for depositing sufficient excitation to produce fission isomers of the target nucleus.

The excitation function for isomer production can also be understood in terms of the (d,pn) reaction mechanism. The amount of excitation energy which can be deposited in the residual nucleus by the (d,p) reaction is limited by the low-energy proton cutoff due to the Coulomb barrier. At deuteron energies below 15–16 MeV, relatively few protons are emitted[13] at low enough energies to deposit the required 8–9-MeV (neutron binding energy plus isomer excitation energy) in the residual nucleus following the (d,p) reaction. As the deuteron energy is increased, the probability for populating highly excited states increases rapidly; thus there is a rapid increase in the isomer cross section as the deuteron energy is raised from 16 to 21 MeV.

The relative amounts of ^{236m}U produced by the (d,pn) and the (d,d') reaction is not known. The (d,pn) reaction is probably the principal contribution to isomer formation. It has been observed[14,15] that direct reactions such as (α,α'), (d,p), or (d,t) produce fissioning isomers in low yield compared to compound nuclear reactions such as

(α, xn) and (d, xn) reactions. Thus we expect that inelastic deuteron scattering produces low fission isomer yields. Neutron evaporation following the d, p stripping reaction probably populates energy levels that are similar to the levels populated in pure compound nuclear reactions, leading to large isomer yields. Evidence for this differentiation can be obtained by comparing the yield of ^{236m}U in the ^{235}U(d, p) reaction at 13 MeV, 1.3 μb,[3] with the yield of ^{236m}U in the ^{236}U(d, pn) reaction at 21 MeV, 13 μb. This point is pursued more quantitatively in a later section where estimates of isomer ratios are made.

The assignment of the 195-nsec activity to ^{238m}U is based primarily on the similarity of the two excitation functions in Fig. 1, and also on the negative results of Lark *et al.*[3] for producing an isomer in ^{237}Np by the ^{238}U$(p, 2n)$ reaction. A ^{238}U$(d, 3n)$ ^{237m}Np excitation function might be expected to be similar to that observed for ^{238m}U.

Small delayed-fission cross sections have been observed at a deuteron energy of 13 MeV for both ^{236}U and ^{238}U. We have not made half-life measurements at this low deuteron energy, so some of the observed activity may be due to other reactions. However, Lark *et al.*[3] report that 11- and 13-MeV deuteron bombardment of ^{238}U results in an isomer with a half-life of 300 ± 100 nsec. Since this half-life value is, within its uncertainty, the same as the half-life determined for ^{238m}U, the tentative assignment of the activity to ^{239m}Np by Lark *et al.* may be incorrect.[16] It is not understood, however, why ^{236m}U was not observed by Lark *et al.* with bombardment of a ^{236}U target with 13-MeV deuterons.

Since the ^{236m}U and ^{238m}U were made in high yield by deuteron bombardment of ^{236}U and ^{238}U targets, respectively, a ^{234}U target was bombarded with 21-MeV deuterons in an attempt to produce ^{234m}U. No delayed fission was observed, however, and an upper limit of about 2 μb can be set if the half-life of ^{234m}U is greater than 30 nsec. This negative result is in agreement with the results obtained for the ^{232}Th$(\alpha, 2n)^{234}$U reaction.[6,17] No delayed activity was observed with 26- and 27-MeV helium-ion bombardment of ^{232}Th. The upper limit[17] on the cross section of ^{234m}U is approximately 0.1 μb for a half-life greater than 20 nsec. From spallation cross-section systematics,[18] the ^{232}Th$(\alpha, 2n)$ cross section is estimated to be about 100 mb. Thus, the isomer ratio of ^{234m}U is less than 10^{-6} for a half-life greater than 20 nsec if there are no competing modes of decay.

An indirect estimate of the isomer ratio for ^{236}U and ^{238}U at 21 MeV can be made. The isomer ratio is defined as the ratio of the delayed-fission cross section to the ground-state cross section, where the latter is usually determined from the appropriate spallation cross section. The latter can be estimated by integrating the proton spectra[9] over angle and over the energy region, which would correspond to emission of one, but not two, neutrons from the residual nucleus following (d, p) stripping. It is assumed that the Th results are valid for U, and that half of the observed protons are due to Coulomb breakup and cannot contribute to the excitation of the target nucleus. This latter estimate is believed to be good to about a factor of two. Isomer ratios at $E_d = 21$ MeV of 9×10^{-5} for ^{236m}U and 5×10^{-5} for ^{238m}U are obtained. For comparison, we have also estimated an isomer ratio of 1.3×10^{-5} for producing ^{236m}U by the ^{235}U(d, p) reaction at 13 MeV, using the 1.3-μb isomer cross section of Lark *et al.*[3] and a (d, p) stripping cross section of 100 mb interpolated from results on neighboring targets.[19,20]

IV. DISCUSSION

Fission isomerism is a common occurrence in the plutonium and americium isotopes, and can be successfully interpreted in terms of double-humped fission barriers.[3,4,21,22] It has been known for a long time that nuclei in their ground state with unpaired nucleons exhibit longer lifetimes than neighboring even-even nuclei. This effect has also been observed for fission isomers.[4] This odd-nucleon effect can be qualitatively explained in terms of a "specialization energy"[6,23,24] which increases the height of the fission barrier due to conservation of spin and parity of the nucleus.

In contrast to the plutonium and americium isotopes, ^{236m}U and ^{238m}U are the only isomers that have been identified in the uranium and neptunium isotopes (with the possible exception of ^{239m}Np as discussed in the last section). But the two even-even uranium isomers have half-lives that are longer than any even-even plutonium isomers that have been studied at this time. With this apparent higher stability of uranium isomers compared to plutonium isomers, it is surprising that fission isomerism is so rare in the uranium and neptunium isotopes. Due to the specialization energy effect, one would expect that many uranium and neptunium isomers with unpaired nucleons would have half-lives that are greater than the even-even uranium isomers. The lack of known cases of fission isomerism may, in part, be because of the detection techniques that have been used, such as the recoil technique,[3] which is most sensitive in the 10–500-nsec half-life range. Thus a more careful study in the microsecond to millisecond half-life range is necessary. Present studies,[3,25] including

this work, are sufficient to establish that long-lived isomers are not produced in high yield. Some other mode of decay may be responsible for deexcitation of the isomers, making the fission branch so small that it has escaped detection. A particularly likely mode of decay is γ emission, which occurs when the isomeric nucleus tunnels through the inner (first) barrier and decays by γ emission to the stable ground state. Because of the slow nature of γ emission, the penetrability of the inner barrier must be many orders of magnitude higher than the penetrability of the outer (second) barrier for γ emission to compete with fission. If the principal mode of decay for shape isomers in the uranium is γ emission, the delayed fission observed from ^{236m}U and ^{238m}U may be small fission branches of the isomers, with γ decay controlling the half-lives. The isomer ratios for producing ^{236m}U and ^{238m}U via the (d, pn) reaction, although considerably higher than for the (d, p) reaction, are still much smaller than observed for plutonium isomers.[1] Thus the observed fission branch may not represent the total production of the isomer.

A greater tendency for the uranium isomers to decay by γ emission than by fission is also indicated by the trends in the relative heights of the first and second barriers in recent theoretical calculations[26] of the potential energy curves for U and Pu isotopes. The calculations predict that the first barrier height relative to the second barrier height is 1 MeV less for U isotopes than for plutonium isotopes. This result has only qualitative significance, as these calculations are expected to underestimate this difference owing to the increasingly inadequate parameterization of the nuclear shape for deformations appropriate to the second barrier as Z^2/A decreases. The predicted decrease in the absolute magnitude of the height of the inner barrier for uranium compared to plutonium is not expected to be much affected by this difficulty.

V. SUMMARY

The delayed fission that has been observed in deuteron bombardments of ^{235}U, ^{236}U, and ^{238}U targets has been attributed to two isomers, ^{236m}U and ^{238m}U. Identification of the fissioning isomers has been made with several pieces of indirect evidence, including half-life measurements, excitation functions, proton energy spectra, and (d, pf) coincidence measurements. The isomer which was produced by bombardment of ^{236}U with 18 and 21-MeV deuterons has a half-life that is in agreement with the previously measured half-life of ^{236m}U. This half-life agreement, along with the absence of ^{237m}U and neptunium isomers that could be produced by ^{236}U(d, xn) reactions, as determined by Lark *et al.*,[3] lead to the conclusion that ^{236m}U is being produced by the ^{236}U(d, pn) reaction. The similarity of the excitation functions for both ^{236}U and ^{238}U lead to the identification of ^{238m}U with a half-life of 195 ± 30 nsec. The only other assignment [^{237}Np produced by a $(d, 3n)$ reaction] which might be compatible with this excitation function is excluded by the absence of delayed fission in the ^{238}U$(p, 2n)$ reaction.[3] The failure to produce delayed fission by reactions resulting from deuteron bombardment of ^{234}U is in agreement with the results of Lark. The cross-section limits that have been set for the ^{232}Th$(\alpha, 2n)$ reaction indicate that if ^{234m}U has a half-life of the order of 20 nsec or longer, the isomer ratio is less than typically observed. It is suggested that the infrequent occurrence of fission isomerism in this mass region may be due to γ competition as well as due to the detection techniques, which are not sensitive enough in the μsec–msec half-life range.

Note added in proof: We have learned that R. Repnow *et al.*[27] have observed ^{238m}U, reporting a half-life of 110 nsec. They have also come to similar conclusions concerning the mechanism of its production.

---

*Research supported in part by the U. S. Atomic Energy Commission.

†Now at Argonne National Laboratory, Argonne, Illinois.

‡On leave from Bhabha Atomic Research Centre, Trombay, Bombay-85, India.

[1]G. N. Flerov *et al.*, in Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, Austria, 1965 (IAEA, Vienna, Austria, 1965), Vol. I, p. 307.

[2]K. L. Wolf and R. Vandenbosch, Bull. Am. Phys. Soc. 13, 1407 (1968).

[3]N. Lark, G. Sletten, J. Pedersen, and S. Bjørnholm, Nucl. Phys. 139, 481 (1969).

[4]R. Vandenbosch and K. L. Wolf, in Proceedings of Second Symposium on Physics and Chemistry of Fission, Vienna, Austria, 1969 (IAEA, Vienna, Austria, 1969), paper No. SM-122/110.

[5]V. Metag, R. Repnow, P. von Brentano, and J. D. Fox, Z. Physik 226, 1 (1969); in Proceedings of Second Symposium on Physics and Chemistry of Fission, Vienna, Austria, 1969 (IAEA, Vienna, Austria, 1969), paper No. SM-122/29.

[6]K. L. Wolf, Ph.D. thesis, University of Washington,

1969 (unpublished).
[7]H. Fauska, R. E. Karns, and D. M. Drake, Nucl. Inst. Methods 72, 233 (1969).
[8]H. Fauska, N. G. Ward, J. Lilley, and C. F. Williamson, Nucl. Inst. Methods 63, 93 (1968).
[9]R. Vandenbosch, to be published.
[10]F. Udo and L. A. Ch. Koerts, Phys. Letters 3, 181 (1963).
[11]E. C. May, B. L. Cohen, and T. M. O'Keefe, Phys. Rev. 164, 1253 (1967).
[12]R. Vandenbosch, K. L. Wolf, J. Unik, C. Stephan, and J. R. Huizenga, Phys. Rev. Letters 19, 517 (1967).
[13]F. A. Aschenbrenner, Phys. Rev. 98, 657 (1955).
[14]S. Bjørnholm, J. Borggreen, Yu. P. Gangrsky, and G. Sletten, Yadern. Fiz. 8, 459 (1968) [transl.: Soviet J. Nucl. Phys. 8, 267 (1969)].
[15]It has been observed in this laboratory (see Ref. 6) that the isomer yield from the ^{236}U(α, α')^{236m}U reaction is less than 1 μb for 42-MeV helium-ion bombardments.
[16]Recent measurements at Copenhagen have led to the reassignment of this activity to ^{238m}U (S. M. Polikanov and G. Sletten, private communication).
[17]K. L. Wolf and J. P. Unik, private communication.
[18]R. Vandenbosch and G. T. Seaborg, Phys. Rev. 110, 507 (1958).
[19]R. Vandenbosch, Ph.D.thesis, University of California Radiation Laboratory Report No. UCRL-3858, 1957 (unpublished).
[20]R. M. Lessler, Ph.D. thesis, University of California Radiation Laboratory Report No. UCRL 8439, 1958 (unpublished).
[21]V. M. Strutinsky, Nucl. Phys. A95, 420 (1967).
[22]S. G. Nilsson, C. F. Tsang, A. Sobiczewski, Z. Szymanski, S. Wycech, C. Gustafson, I. Lamm, P. Möller, and B. Nilsson, Nucl. Phys. A131, 1 (1969).
[23]J. O. Newton, Progr. Nucl. Phys. 4, 234 (1955).
[24]J. A. Wheeler, Niels Bohr and the Development of Physics, edited by W. Pauli, with the assistance of L. Rosenfeld and V. Weisskopf (Pergamon Press, London, 1955).
[25]D. S. Brenner, L. Westgaard, and S. Bjørnholm, Nucl. Phys. 89, 267 (1966).
[26]C. F. Tsang and S. G. Nilsson, Nucl. Phys. 140, 275 (1970).
[27]R. Repnow, V. Metag, J. D. Fox, and P. von Brentano, to be published.

PHYSICAL REVIEW C VOLUME 1, NUMBER 6 JUNE 1970

Emission of Particles of $3 \leq Z \leq 8$ in the Fission of Cf^{252}

Yaacov Gazit and Eran Nardi
Israel Atomic Energy Commission, Soreq Research Center, Yavne, Israel
and Weizmann Institute of Science, Rehovot, Israel

and

Seymour Katcoff*
Weizmann Institute of Science, Rehovot, Israel
(Received 13 February 1970)

The energy spectra and yields of particles of $3 \leq Z \leq 8$ emitted in the spontaneous fission of Cf^{252} were measured. The experiment was carried out using a particle telescope consisting of two dE/dx gas-filled proportional counters and a solid-state E counter. The cutoff energies were relatively low since essentially no absorber foil was placed between source and detector. The yields per 10^3 fission α particles are: Li, 2.9 ± 0.5; Be, 6.5 ± 0.7; B, 0.35 ± 0.10; C, 4.3 ± 0.9; N, $\geq 0.66 \pm 0.15$; O, $\geq 0.05 \pm 0.01$. Trajectory calculations were carried out for Li, Be, and C, and the initial conditions which give best agreement with the experimental results were obtained.

I. INTRODUCTION

Most of the recent studies dealing with the process of α-particle-accompanied fission have been carried out with the main intention of obtaining information on the initial conditions of the fissioning nucleus at the point of scission.[1,2] The study of light-particle emission in fission, in general, contains other features of interest in addition to the one cited above. Particle emission in fission can, for example, furnish information on the dynamics of saddle-point–to–scission-point motion, and perhaps also on the clustering probabilities of the various light nuclei within the fissioning nucleus. We should also note that trajectory calculations performed for the H and He isotopes emitted in fission show that the initial conditions at scission vary somewhat from particle to particle.[3] This result might be of physical significance with regard to the light-particle emission mechanism. Further studies on light-particle fission are essential in obtaining a more complete description of this process, and for this reason the study of the heavier particles, i.e., of $Z > 2$, was under-