It is interesting to note that the values of β deduced from direct observation of the de-excitation γ cascade in Ba^{126} [16] and those deduced from lifetime measurements in Ba^{128} [17] (both even-even nuclei) indicate a deformation greater than that which can be inferred from the existence of, and energy relation between the isomeric states in Ba^{125} and Ba^{127}. However, it is not clear whether these earlier measurements or those reported here refer to the natural ground-state configuration or are merely indicative of the deformability in the excited states. It would appear that the deductions based on the relative spacing of the isomeric levels would more realistically approach the picture of the true ground-state configurations.

At this point, it is not at all clear as to the origin and detailed nature of what appears to be negative-parity excited states at 160 keV in Cs^{125} and 310 keV in Cs^{127}. The allowed nature of the β decay from the high-spin negative-parity precursors seems to require this spin assignment $(\frac{7}{2}-)$, however. And again, the absence of states at energies above a few hundred keV cannot be explained in the framework of existing information.

[16] J. Clarkson, doctoral dissertation, University of California, 1965 (unpublished); J. Clarkson, R. M. Diamond, F. S. Stephens, and I. Perlman, Nucl. Phys. A93, 272 (1967).

[17] P. J. Pan, Y. S. Horowitz, R. B. Moore, and R. Barton, Can. J. Phys. 44, 1029 (1966).

ACKNOWLEDGMENT

The authors thank the staff of the heavy-ion accelerator for their cooperation and help in various sample bombardments.

PHYSICAL REVIEW VOLUME 172, NUMBER 4 20 AUGUST 1968

Ternary Fission of U^{238} Induced by Intermediate-Energy Helium Ions*

R. H. Iyer† and J. W. Cobble
Department of Chemistry, Purdue University, Lafayette, Indiana 47907

(Received 6 March 1968)

Evidence is presented for the existence of ternary fission of a heavy excited nucleus into three fragments of comparable size from radiochemical studies on U^{238} bombarded with 20–120-MeV helium ions. The absolute formation cross sections for Na^{24}, Mg^{28}, Si^{31}, S^{38}, Ca^{47}, Mn^{56}, and Ni^{66} in the (39±1)-MeV He^4-induced fission of U^{238} clearly establish the transition region between binary and ternary processes, the crossover occurring around $A=47$. The corresponding data for Ta^{183} and Ta^{184} and upper limits for Au^{199}, Pb^{209}, and Pb^{212} also indicate the absence of any possible complimentary heavy fragments. The fission-product nature of the light ternary fragments was confirmed by the forward-to-backward recoil properties. The excitation function for the formation of Mg^{28} and the ratio $\sigma_{Mg^{28}}/\sigma_{total}$ drop very rapidly below an excitation energy of 20 MeV, making it highly unlikely that such light fragments can be observed at lower excitation energies. These conclusions are consistent with the previously reported lack of radiochemical evidence for the possible thermal-neutron-induced ternary fission of U^{235}, but are inconsistent with the claims to the contrary based on purely instrumental measurements. The mass-yield curve appears to be smooth in both the ternary and low-mass binary regions, and gives no indication of any "fine structure" in the ternary region.

The recoil properties of Mg^{28} and Sr^{89} studies in the 40–120-MeV energy range indicate close similarities. A mean recoil range of 12.2±0.4 mg/cm² in uranium was obtained for Mg^{28}, corresponding to an average kinetic energy of 45–50 MeV based on similar heavy-ion range-energy data.

I. INTRODUCTION

SPECULATIONS concerning the possibility that a heavy excited nucleus might divide into three fragments of comparable mass, i.e., ternary fission, were made by Present and Knipp[1] shortly after the discovery of nuclear fission by Hahn and Strassmann[2] in 1939. Since that time experimental evidence for ternary fission has been sought by a variety of techniques, particularly by examination of fission tracks in nuclear emulsions impregnated with fissionable nuclides. Emission of small masses such as tritons, He^4, and other light fragments with $Z\leqslant 3$ and $A<10$ are also considered by some authors[3] as a type of ternary-fission process. Much of the earlier work on these light "ternary"-fission products has been summarized previously.[4–7] However, very light fragments can also result from direct interaction not involving a cooperative nuclear phenomena such as is responsible for larger fragment fission.

The first reported evidence for ternary fission into three fragments of comparable mass was that of Tsien

* Supported by the U. S. Atomic Energy Commission.

† On leave from the Bhabha Atomic Research Center, Bombay 74, India.

[1] R. D. Present and J. K. Knipp, Phys. Rev. 57, 1188 (1940).

[2] O. Hahn and F. Strassmann, Naturwiss. 27, 11 (1939).

[3] E. K. Hyde, *Nuclear Properties of the Heavy Elements* (Prentice-Hall, Inc., Englewood Cliffs, N, J., 1964), Vol. III, p. 131.

[4] N. A. Perfilov, *Physics of Nuclear Fission* (Pergamon Press Ltd., London, 1958), p. 84.

[5] W. J. Whitehouse, *Progress in Nuclear Physics* (Academic Press Inc., New York, 1952), Vol. II, p. 120.

et al.,[8,9] studied in nuclear emulsions loaded with U^{235} irradiated with thermal neutrons. Catala *et al.*[10] and Perfilov[4] have also reported observing such three-pronged tracks using emulsion techniques. However, other investigators,[11–14] using the same techniques, could not attribute observed tracks to be unambiguously due to fission. Rosen and Hudson,[15] using triple coincidence techniques and ionization chambers, reported the frequency of symmetrical ternary fission to be 6.7 ± 3.0 per 10^6 binary fissions in U^{235} irradiated with thermal neutrons. A systematic radiochemical search for ternary-fission products in the mass range 30–60 was first made in 1951, but no positive evidence could be obtained.[16] A careful survey[3–7] of the earlier work on the ternary fission of U^{235} with thermal neutrons easily reveals the lack of agreement of the existing data on this admittedly experimentally difficult subject. There is, of course, the inherent uncertainty in distinguishing between a track due to a true ternary-fission event and one due to a binary event with an accompanying heavy-ion recoil originating at approximately the same point. The triple coincidence measurements suffer from the defect that triple events can also be registered from collisions of binary-fission fragments with carbon, nitrogen, oxygen, fluorine, etc., atoms present in the target at the point of fission. There is also the possibility that a triple coincidence can be triggered by the occurrence of two binary events within the resolution time of the coincidence circuitry.[15]

Ternary fission in heavy elements has again attracted considerable attention recently both at very low excitation energies[17–19] and at very high excitation energies.[20,21] The most recent and rather extensive instrumental data[17–19] on the spontaneous and thermal-neutron-induced fission of heavy-element isotopes have indicated rather high ternary to binary ratios. However, the equally recent, extensive, and careful radiochemical search[7,22–24] for a number of possible ternary-fission products in the thermal-neutron-induced fission of U^{235} and the spontaneous fission of Cf^{252} in the mass range covered by the nuclides Be^7, Mg^{28}, S^{38}, Ar^{37}, Ar^{39}, Ar^{41}, Ar^{42}, Sc^{48}, Cr^{51}, Mn^{54}, Mn^{56}, Co^{56}, Co^{57}, Fe^{59}, Co^{58}, and Co^{60} has failed to support those claims, and suggests that ternary fission has not probably been observed in heavy elements at lower excitation energies ($\leqslant 6.5$ MeV). The reported cases of high-energy ternary fission involve heavy ions[20] and protons[21] of energies in the several-hundred-MeV range, but it is not completely certain that such events result from compound nuclear processes.

It seemed clear that studies in the intermediate-energy range would be of interest since direct-interaction events leading to heavier fragments are not very probable at such energies. It is in this energy range that we have discovered the existence of what appears to be a true compound-nuclear asymmetric ternary-fission process. A preliminary report of this work based on limited data has appeared elsewhere.[25] The present communication describes the results of a more extensive study of the ternary process in U^{238} excited with helium ions between 20–120 MeV and provides additional support to our original hypothesis.

II. EXPERIMENTAL

The experimental arrangements and details were the same as those reported earlier.[25] Briefly, bombardments of 1-mil natural uranium foils were carried out with helium ions on the cyclotrons at the Argonne National Laboratory and the Lawrence Radiation Laboratory at Berkeley. The total currents involved were 10–35 μAh at energies ranging from 20–120 MeV. Below 60 MeV, various excitation energies were obtained by using degrading foils in front of the target. The 65-, 80-, and 120-MeV beams were provided directly from the Berkeley 88-in. variable-energy cyclotron.

Target foils were placed between ultrapure (99.9999%) silver catcher foils which served to collect recoiling fission fragments in the forward and backward (to the beam) directions. The uranium foils had been analyzed[26] both spectroscopically and radiochemically for some 54 elements which might produce spallation products that could possibly interfere with the fission fragments. The silver catcher foils were made by evaporation of ultrapure (99.9999%) silver beads (obtained from the United Mineral and Chemical Corp., New York) onto Lexan

[6] P. Demers, *Ionographie Les Emulsions Nucleaires* (Montreal University Press, Ottawa, Canada, 1958).
[7] J. C. Roy, Can. J. Phys. **39**, 315 (1961).
[8] S. T. Tsien, Z. W. Ho, L. Vigneron, and R. Chastel, Nature **159**, 773 (1947).
[9] S. T. Tsien, Z. W. Ho, R. Chastel, and L. Vigneron, J. Phys. Radium **8**, 165 (1947).
[10] J. Catala, J. Casanova, and V. Domingo, Nature **184**, 1057 (1959).
[11] L. L. Green and D. L. Livesey, Nature **159**, 332 (1947).
[12] S. P. Dutta, Indian J. Phys. **27**, 547 (1953).
[13] P. Demers, Can. J. Phys. **31**, 78 (1953).
[14] E. W. Titterton, Nature **168**, 590 (1951).
[15] L. Rosen and A. M. Hudson, Phys. Rev. **78**, 533 (1950).
[16] P. P. Metcalf, J. A. Sieler, E. P. Steinberg, and L. Winsberg, in *Radiochemical Studies: Fission Products*, edited by C. D. Coryell, and N. Sugarman (McGraw-Hill Book Co., New York, 1951), Book I, papers 47–51.
[17] M. L. Muga, Phys. Rev. Letters **11**, 129 (1963).
[18] M. L. Muga, in *Symposium on the Physics and Chemistry of Fission, Salzburg, Austria* (International Atomic Energy Agency, Vienna, 1965).
[19] M. L. Muga, C. R. Rice, and W. A. Sedlacek, Phys. Rev. Letters **18**, 404 (1967); Phys. Rev. **161**, 1266 (1967).
[20] R. L. Fleisher, P. B. Price, R. M. Walker, and E. L. Hubbard, Phys. Rev. **143**, 943 (1966).
[21] R. Brandt, F. Carbonara, E. Cieslak, M. Dakowski, Ch. Gfeller, H. Piekarz, J. Piekarz, W. Reizler, R. Rinzivillo, E. Sassi, M. Sowinski, and J. Zakrzewski, CERN Report, Geneva, 1966 (unpublished).
[22] R. W. Stoenner and M. Hillman, Phys. Rev. **142**, 716 (1966).
[23] R. J. Prestwood and B. P. Bayhurst, Bull. Am. Chem. Soc. **151**, J15 (1966).
[24] W. E. Nervik, Phys. Rev. **119**, 1685 (1960).
[25] R. H. Iyer and J. W. Cobble, Phys. Rev. Letters **17**, 541 (1966).
[26] The analyses were performed at the Oak Ridge National Laboratory by P. F. Thomason, J. R. Sites, and W. Laing.

TABLE I. Fission-product cross sections for U^{238} excited by 39-MeV He^4 ions.

Fission product	Measured cross section (nb)	Total chain yield[a] (nb)	
Na^{24}	230±110	230	
Mg^{28}	200±50	200	
Si^{31}	60±30	60	
S^{38}	25± 6	25	
Ca^{47}	≤5	≤5	
Mn^{56}	400±100	400	(4)
Ni^{66}	(29± 6)×10^3	29×10^3	(5)
Ta^{183}	(1.5±0.8)×10^3	1.6×10^3	(4)
Ta^{184}	270±140	300	(4)
Au^{199}	≤5	≤5[b]	(3)
Pb^{209}	≤3	≤4[b]	(2)
Pb^{212}	≤4	≤13[b]	(2)

[a] These cross sections were calculated from the individual fission-product cross sections assuming the constant-charge-ratio hypothesis for independent yields to be valid; the number of neutrons estimated to be emitted in the fission process is given parenthetically (see text).
[b] These values were estimated from the measured limits as if the products has originated from a binary event.

polycarbonate resin strips (General Electric Co., New York) which were later dissovled off in methylene dichloride.

Standard chemical procedures using carrier techniques were used to separate the various fission products. After irradiation, the target and the catcher foils were removed and dissolved separately in the presence of added carriers. One of the problems encountered in this investigation was to find satisfactory radiochemical procedures capable of separating the trace amounts of radioactivity due to ternary-fission nuclides from gross amounts of binary and spallation products. The counting rates of the ternary species varied from about 1 to 100 counts/min (cpm) and, in general, required radiochemical decontamination factors of the order of 10^7. Details of the separation and purification schemes are described elsewhere[27] and summarized briefly in the Appendix. The activities were counted in suitable chemically stable precipitates in low-background standardized[28] (0.12–0.20 cpm) gas-flow window Geiger counters provided with anticoincidence guard counters. The decay was followed in many cases for four to five half-lives. The radioactive purity of the samples was established by constant specific-activity tests, by characteristic decay, and in the case of Mg^{28} by its γ-ray spectrum. The resolution of the decay curves was aided by a modified[29] Los Alamos PAKAG IBM 7094 computer program.[30] Sr^{89} was isolated from many of the targets to serve as an internal binary-fission standard.

Na^{24}, Mg^{28}, and Sr^{89} were isolated from both the silver catcher foils and the uranium targets. Mn^{56}, Ni^{66}, and Si^{31} were isolated only from the catcher foils mainly because some of these nuclides could be produced within the target by spallation reactions on impurities. One-mil aluminum catcher foils were used in the case of Si^{31} because it readily dissolves in strong base, which helps assure proper exchange of radioactive silicon species with the silicate carrier used. Analysis of blank silver foils (and aluminum foils in the case of Si^{31}) irradiated along with the target assembly indicated that except for Na^{24} and Si^{31} none of the other nuclides were produced in detectable amounts by nuclear reactions on impurities in the catcher foils under the conditions of the experiments. Appropriate blank corrections were applied to the Na^{24} and Si^{31} data.

In computing the absolute cross sections for the recoil products Si^{31}, Mn^{56}, and Ni^{66}, the assumption was made that the same fraction of the total activity recoiled out of the thick uranium target as was experimentally found for Mg^{28} and Sr^{89} (7% in both the forward and backward direction in each case).

Na^{24} can result from the uranium target by an (n,γ) reaction on trace amounts of sodium due to the general thermal-neutron flux around accelerators, and by He^4-induced reactions on aluminum and magnesium impurities in the uranium. A suitable correction can be determined by irradiations below the effective threshold for the production of fission Na^{24} (<20 MeV). Mg^{28} can result from spallation reactions induced on aluminum and magnesium impurities in the uranium target: $Al^{27}(\alpha,3p)Mg^{28}$, $Mg^{26}(\alpha,2p)Mg^{28}$. Corrections are only necessary to the lower-energy data (<25 MeV) and were made using the known cross sections[31] and an activation analysis of the uranium for aluminum and magnesium content.

III. EXPERIMENTAL RESULTS

Table I gives a summary of the experimental cross sections measured in this research for the fission of U^{238} induced by He^4 ions with an average bombarding energy of 39.0±1.0 MeV. The cross sections of all but Mn^{56}, Ta^{183}, and Ta^{184} are derived from more than one determination, and have been corrected for contributions from helium ions and secondary-neutron-induced spallation reactions on possible target and catcher impurities.[25,27] (These corrections were appreciable only in the case of Na^{24}, Si^{31}, and the lower-energy Mg^{28} data.) They were further corrected for recoil losses of 14%, the value experimentally observed for Mg^{28} and Sr^{89}. Column 3 of this table represents the corrected yields for the mass chain identified with each isotope. These total isobaric yields for binary fission were estimated on the basis of the constant-charge-ratio hypothesis which has proved both useful and accurate in this range of excitation energy.[32] The number of neutrons boiled off, $\bar{\nu}$, with each mass division, was estimated from mass-

[27] R. H. Iyer, Ph.D. thesis, Purdue University, 1967 (unpublished).
[28] F. Lisman, Ph.D. thesis, Purdue University, 1965 (unpublished).
[29] Clarence Menninga, Ph.D. thesis, Purdue University, 1966 (unpublished).
[30] Los Alamos PAKAG Program, LA-2367, La-2367 addenda, 1963 (unpublished).
[31] R. J. Carr and R. H. Lindsay, Phys. Rev. **118**, 1293 (1960).
[32] N. A. Wogman, J. A. Powers, and J. W. Cobble, Phys. Rev. **152**, 1088 (1966).

TABLE II. Recoil data for Mg^{28} and Sr^{89} in uranium metal. E_{av} is the average bombarding energy, $\frac{1}{2}(E_{in}+E_{out})$, σ_{av} is the measured total cross section in the thick target corrected for recoil losses, F_{av} and B_{av} are the measured fractions of total activities recoiled in the forward and backward directions, respectively, F_{corr} and B_{corr} are the corresponding quantities corrected for difference in σ at the front and back faces of the target foil due to beam energy degradation, and W is the thickness of the uranium target foil in mg/cm².

E_{av} MeV	σ^{av}	F_{av} (%)	B_{av} (%)	F_{corr} (%)	B_{corr} (%)	$(F/B)_{corr}$	$\eta=\frac{(F/B)^{1/2}-1}{(F/B)^{1/2}+1}$	$R_{FB}\approx 2W(F+B)/1+\eta^2$
				Mg^{28}				
39.0	190 nb	6.90	7.00	7.71	6.34	1.22	0.0498	12.0
62.5	1450	6.90	6.78	7.40	6.15	1.20	0.0459	11.7
78.5	3000	6.70	7.02	7.18	6.60	1.09	0.0219	11.8
118.5	9400	7.55	7.60	7.77	7.45	1.04	0.0099	13.1
								mean 12.2±0.4 mg/cm²
				Sr^{89}				
39.0	13.6 mb	7.70	7.00	8.22	6.58	1.25	0.0530	12.7
78.5	42.0	6.84	6.13	···	···	1.09	0.0219	11.3
118.5	38.0	8.25	7.48	···	···	1.10	0.0244	13.5
								mean 12.5±0.8 mg/cm²

energy–balance considerations.[33] In this estimation, the energy available for neutron emission is assumed to be the difference between the total mass energy released in the process and the sum of the total kinetic energy E_k, and that carried off by the γ rays E_γ. The mass energy released for each mass division was calculated from the mass tables of Wing and Varley[34] and Hillman.[35] The most probable charges of the light and heavy masses $Z_P{}^L$ and $Z_P{}^H$, respectively, were estimated from the constant-charge-ratio hypothesis. E_γ was fixed at 7.5 MeV[3] and the neutron kinetic energy at 1.2 MeV. The kinetic energy of two possible *binary* products was estimated as $E_k=Z_P{}^L Z_P{}^H e^2/D$, where D was taken as 18 F. The resulting calculations, taking into account the neutron binding energies of the fragments,[34] clearly indicate that $\bar{\nu}$ must decrease as the mass division becomes more highly asymmetric. Further, these estimates are in fair agreement with the values obtained by reflection of the binary mass-yield curve[36] in the very light and the very heavy mass regions, where the independent yield corrections are not significant by any hypothesis. The mass yields for Na^{24}, Mg^{28}, Si^{31}, S^{38}, and Ca^{47} were assumed to be approximated by the yield of the isotope measured. The mass-yield data are summarized in Fig. 1.

The excitation functions of Sr^{89} and Mg^{28} were obtained in the 20–120-MeV energy range to obtain information on the energy dependence and possibly the threshold for the ternary process. Na^{24} and S^{38} cross sections from He^4 studies as well as the Mg^{28} data from the He^3-ion-induced fission of U^{238} indicate approximately the same trend, although those measurements were not extended beyond 40 MeV. The excitation-function data (from Table II and Ref. 25) are summarized in Fig. 2.

[33] H. C. Britt, H. W. Wegner, and J. C. Gursky, Phys. Rev. 129, 2239 (1963).
[34] J. Wing and J. D. Varley, Argonne National Laboratory Report No. ANL-6886, 1960 (unpublished).
[35] M. Hillmann, Brookhaven National Laboratory Report No. BNL-846 (T-333), 1964 (unpublished).
[36] L. J. Colby, M. L. Shoaf, and J. W. Cobble, Phys. Rev. 121, 1415 (1961).

The errors quoted for the individual cross-section values are estimated from various uncertainties in the measurements. These errors are largely due to corrections for contributions from spallation impurities, estimation of the counter efficiencies, resolution of decay curves, determination of chemical yields, and counting statistics.

IV. DISCUSSION

It is instructive to examine some reasonable possibilities other than ternary fission which might account for the observed light fragments, such as (a) spallation

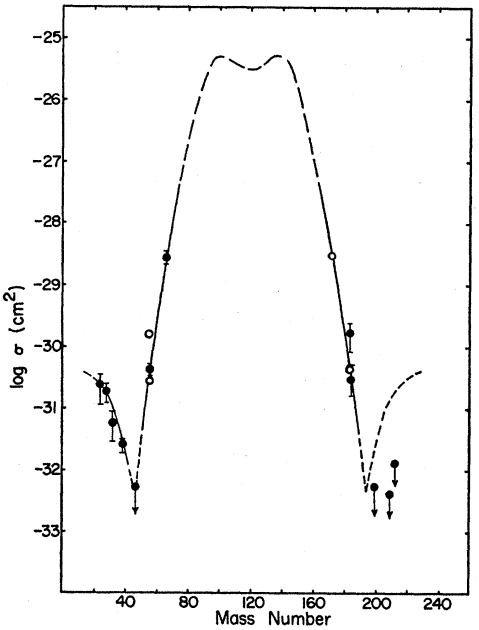

FIG. 1. Fission mass-yield curve for U^{238} excited by 39-MeV helium ions. Data are from Table I. The upper dotted curve was taken from the work of Colby, Shoaf, and Cobble (Ref. 36). The dotted curve to the lower right of the mass distribution represents the hypothetical heavy yields expected if the light-mass yields had originated from binary events. Open circles represent reflected points.

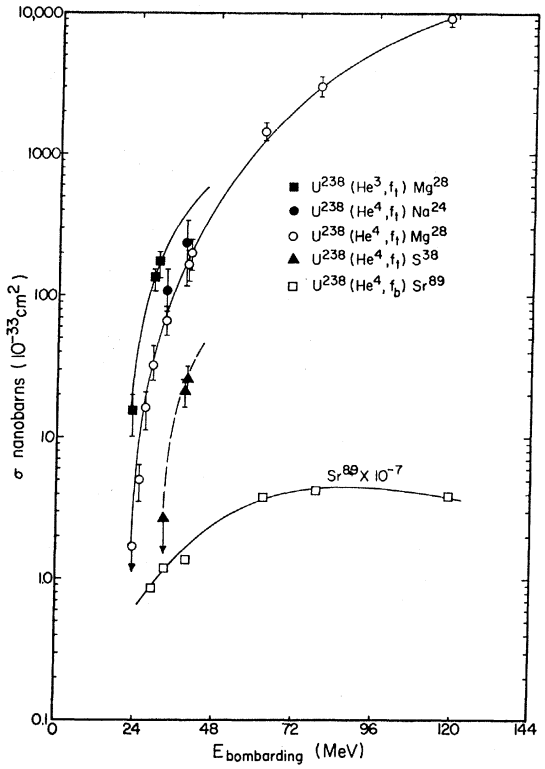

FIG. 2. Excitation functions for some fission products formed in the helium-ion-induced fission of U^{238}. Data are from Table II and Ref. 25. The cross sections of Na^{24} are 106±53 and 230±110 nb for 33- and 39-MeV He^4 ions, respectively.

reactions on impurities, (b) fragmentation, (c) two successive binary fissions, and (d) highly asymmetric binary fission.

As pointed out previously, neither the Mg^{28} or S^{38} data, which are most extensive, could have resulted entirely from He^4- or secondary-neutron-induced reactions on target impurities. However, most important is the fact that *neither* of these two isotopes can be produced by He^3-ion-induced reactions similar to the corresponding He^4-induced reactions: $Al^{27}(\alpha,3p)Mg^{28}$, $Mg^{26}(\alpha,2p)$-Mg^{28}, $Cl^{37}(\alpha,3p)S^{38}$, $S^{36}(\alpha,2p)S^{38}$. Further, secondary-neutron-induced reactions on impurities were studied separately and found to make only negligible contributions to the observed activities. These facts, together with the observation that Mg^{28} and S^{38} were produced in comparable amounts in both He^3- and He^4-induced fission[25] rule out impurities as being responsible for the observed activities.

The crucial forward-backward recoil experiments were carried out specifically to discriminate against nonfission nuclear reactions. These results are given in Table II. Columns 3 and 4 of this table summarize the measured fractions of the total Mg^{28} and Sr^{89} activities which completely recoiled out of the 1-mil-thick uranium target in the forward and backward directions. Since the incident He^4 ion beam is somewhat degraded in passing through the target foil, particularly at the lower bombarding energies, corrections must be applied to these data since the excitation functions are quite energy-dependent (particularly for Mg^{28}). F_{corr} and B_{corr} are the corresponding quantities so corrected for the difference in energies (and hence cross sections) at the front and back surfaces of the target foil. Since the present measurements could not be made with very thin targets, the necessary cross sections at different energies were obtained from the "thick-target" excitation functions. Mn^{56} and Ni^{66} had very similar recoil properties.

The F/B ratios in column 7 are comparable for both Mg^{28} and Sr^{89} and are near unity. This angular dependence would not be expected for spallation reactions at these energies; an F/B ratio considerably higher would be expected for such reactions which show strong forward peaking. (The slightly decreasing trend in the F/B ratio is indicative of the increasing contributions of direct particle emissions and hence decreasing momentum transfer to the residual nucleus.[37–39]) In two separate experiments, it was shown that about 30% of the total Mg^{28} recoiled in the forward direction completely penetrated a silver foil 1.5–2.5 mg/cm² thick.

From the recoil data an average forward-backward range of 12.2±0.4 mg/cm² in uranium can be estimated[37] for the Mg^{28} fragment. This corresponds[40] to an average kinetic energy of 45–50 MeV by interpolation of the heavy-ion range [41,42] for Ne^{20} and Ar^{40}, similar to a method used by Crespo, Alexander, and Hyde[43] for Mg^{28} produced by fragmentation (direct-interaction) reactions at >300 MeV. This recoil energy is not consistent with spallation products at the excitation energies used in this research.

Fragmentation is, of course, an unlikely process in this energy region. It has been shown[43,44] that such direct-interaction processes do not begin until the bombarding energies reach ~100 MeV. Further, the ejected fragments would show strong forward peaking, giving a F/B ratio considerably higher than unity.

A third possibility is that the observed fragments may be resulting from two successive binary fissions. Assuming an initial binary split of the Pu^{242*} compound nucleus with initial excitation energy of 35 MeV to give, e.g., $_{12}Mg^{28}$ and $_{82}Pb^{214}$, it can be shown that the heavy $_{82}Pb^{214}$ fragment carries too little excitation energy to undergo further fission.

Finally, there is a possibility that the observed light

[37] N. T. Porile and N. Sugarman, Phys. Rev. **107**, 1410 (1957).
[38] N. Sugarman, M. Campas, and K. Weilgoz, Phys. Rev. **101**, 388 (1956).
[39] N. Sugarman, Helmut Münzel, J. A. Panontin, Karoline Wielgoz, M. V. Ramaniah, Gerald Langane, and Emilio Lopez-Menchero, Phys. Rev. **143**, 952 (1966).
[40] S. Mukherjee and L. Yaffe, Can. J. Chem. **43**, 232 (1965).
[41] E. L. Hubbard, University of California Lawrence Radiation Laboratory Report No. UCRL-9050, 1960 (unpublished).
[42] L. C. Northcliffe, Phys. Rev. **120**, 1744 (1960).
[43] V. P. Crespo, J. M. Alexander, and E. K. Hyde, Phys. Rev. **131**, 1765 (1963).
[44] J. M. Miller and J. Hudis, Ann. Rev. Nucl. Sci. **9**, 159 (1959).

nuclides could be considered as some kind of highly asymmetric *binary*-fission fragments. The mass-yield curve (Fig. 1) covering the mass range from 24 to 212 for U^{238} excited with 39-MeV helium ions indicates that this is not very probable. It can be seen from this figure that there is a clear separation between the light fragments and the light wing of the binary-fission curve, occurring in the mass region around $A=47$. On both sides of this mass region, the fission yields increase. It is important to note that the observed heavy mass-yield ($A>200$) limits are 1–2 orders of magnitude smaller than those expected if the observed light mass isotopes resulted from a highly asymmetric binary split. Unfortunately, the number of radioactive species which can be reliably measured in the heavy-mass region are quite limited, but we have never been able to detect any radioactivity in those which could be measured. The absence of such heavy products is, of course, expected for a true ternary event.

Actually, such a highly asymmetric binary fission ($A_L<47$ and $A_H>199$) is unlikely from energetic arguments, since the kinetic energy of such a light fragment from a binary event can be estimated to be considerably higher than the measured value of 45–50 MeV, based upon range-energy relationships derived from heavy ions.

It should also be noted that in the range of mass numbers from 95 to 47 on the light side of the binary region and from 145 to 199 on the corresponding heavy side, the mass yields decrease continuously and smoothly. Closely associated with this *decrease* in yields is a corresponding *increase* in nuclear deformation leading to asymmetric binary fission. If the mass division of $A_L<47$ were to be attributed to binary fission, then still greater deformations would be required. Consequently, to explain even the mass-yield data for $A_L<47$ requires a sudden reversal in the normal trend of decreasing yields with greater fission asymmetry ratios (A_H/A_L) and larger nuclear deformations. Such behavior would not be reasonable. While it is conceivable that some combination of shell effects might have given rise to a highly asymmetric binary fission (e.g., 82-proton shell), the observed low limit of Ca^{47} (20-proton shell) indicates that such shell effects do not seem to be important, at least at these energies.

Radiochemical evidence for ternary fission can be obtained only by searching for light fragments far removed from the binary-fission region. Theoretical predictions of Present and Knipp[1] and Swiatecki[45] do not rule out the possibility of ternary fission. On the basis of these facts (both experimental and theoretical), the rise in fission yields below $A\sim47$ is most simply understood as indicating ternary fission, which gives a statistical distribution of light fission masses, clearly separated from the binary region.

[45] W. J. Swiatecki, in *Second United Nations Geneva Conference on Peaceful Uses of Atomic Energy* (Pergamon Press Ltd., London, 1960), Vol. 15, p. 651.

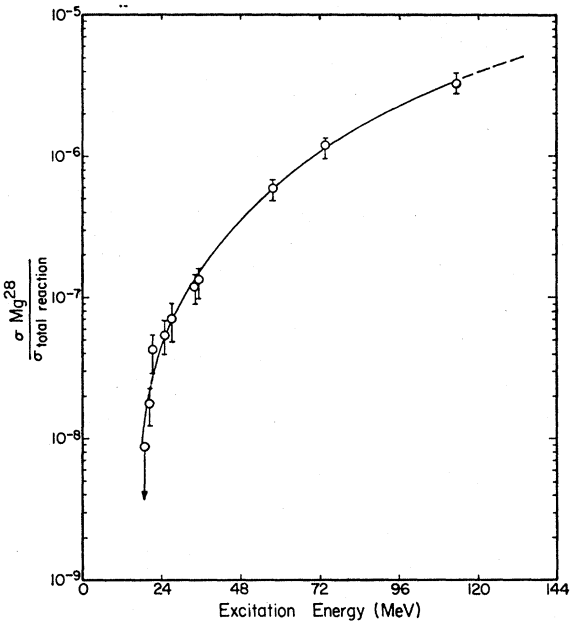

FIG. 3. Fraction of Mg^{28} formed in the irradiation of U^{238} with He^4 ions. Total cross sections are from Ref. 36 and S. S. Kapoor, H. Baba, and S. G. Thompson [Phys. Rev. **149**, 965 (1966)].

At the present time it is not known whether the ternary yields "peak" around $A\approx24$ or continue to rise and eventually merge with the very light mass species observed by others.[46,47] This question cannot be answered radiochemically for Pu^{242*} since there are no suitable isotopes below $A=24$ to measure. However, by ternary-fission studies on much lighter and heavier compound nuclei, the ternary *peak*, if one exists, can probably be resolved.

Preliminary studies[48] in our laboratories on the ternary yields of Mg^{28} in thorium and uranium isotopes indicate that the $\sigma_{Mg^{28}}/\sigma_{\text{binary}}$ fission ratio is not as sensitive a function of Z and A of the compound nucleus as it is of the excitation energy. It is this strong energy dependence (see Figs. 2 and 3), as we previously reported,[25] which led us to believe that the type of ternary fission observed in our experiments could not take place either in the spontaneous or the thermal-neutron-induced fission of other closely related heavey-element isotopes. It is instructive to review just what type of ternary fission is being proposed by others[19] from purely instrumental measurements in these cases. In the first place it is a highly selective type of fission that appears only to populate stable species over the mass range defined by the following isotopes which have been sought by others but not detected radiochemically[7,22,23]: Be^7, Mg^{28}, S^{38}, Ar^{37}, Ar^{39}, Ar^{41}, Ar^{42}, Sc^{48}, Cr^{51}, Mn^{54}, Mn^{56}, Co^{56}, Fe^{59}, Co^{57}, Co^{58}, and Co^{60}. Further, the reported instrumental

[46] S. L. Whetstone and T. D. Thomas, Phys. Rev. **154**, 1174 (1967).

[47] S. W. Cosper, J. Cerney, and R. C. Gatti, Phys. Rev. **154**, 1193 (1967).

[48] T. C. Roginski (private communication).

"ternary"-fission data[19] give yields, as pointed out previously, which are approximately five orders of magnitude higher than would be predicted from our own Na^{24}, Mg^{28}, S^{38}, and Ca^{47} yields. The present research clearly suggests a statistical (in the sense that the yields vary smoothly throughout the mass range involved) type of ternary-fission process accompanying a similar binary process, and it seems highly unlikely that the ternary process should behave so differently from the binary process on going to somewhat lower energies.

The data in Fig. 3 may be used to estimate a threshold energy for the ternary process, assuming Mg^{28} to be one of the major ternary products. The ratio of $\sigma_{Mg}/\sigma_{total}$ eliminates the effect of the Coulomb barrier for the entering α particle, and suggests that a minimum excitation energy of ~12–14 MeV is necessary to produce a ternary split. This value can be compared to the 6.3-MeV threshold energy[3] for binary fission for the same compound nucleus Pu^{242*}.

At present, efforts are underway to obtain some information on the angular distribution of the ternary Mg^{28} fragments, with a view to elucidating a mechanism of the ternary-fission process. Simple calculations based on conservation of kinetic energy and linear momentum indicate that the ternary Mg^{28} would be ejected at about 100° with respect to the binary fragments. Calculations similar to those reported by Whetstone and Thomas[46] for He, Li, and Be fragments from the fission of Cf^{252}, based on a model suggested by Halpern,[49] indicate rather high and unrealistic values for the energies required to release these massive fragments into the region between two binary fragments. Nevertheless, these calculated release energies show an interesting correlation between the mass number and the yields of the ternary fragments: The required energies increase regularly with increasing mass number and indicate a corresponding decrease in yield from Na^{24} to S^{38}. This trend tends to indicate the importance of the energy available for ternary fission, as pointed out by Halpern.[49]

ACKNOWLEDGMENTS

The authors wish to express their thanks to Dr. Bernard Harvey and Mrs. Ruth Mary Larrimer of the Lawrence Radiation Laboratory, Berkeley, for making available the 88-in. cyclotron irradiation facilities, and to Milan Oselka of the Argonne National Laboratory for his excellent cooperation and assistance with the Argonne cyclotron irradiations.

APPENDIX: RADIOCHEMICAL PROCEDURES

Uranium foils were dissolved in 8–10 *M* HCl and the silver foils in concentrated HNO_3 in the presence of added carriers. In most cases, the initial step was removal of the uranium (~200 mg) by means of a Dowex-1, *x*-8 anion column. Silver was precipitated out as AgCl. An initial separation of sulfur, gold, and tantalum was made directly from the uranium target solution; sulfur was precipitated as $BaSO_4$, gold was extracted as the chloro-complex into ethyl acetate, and tantalum was extracted as the fluoro-complex into diisopropyl ketone.

Sodium

Two sets of palladium, copper, nickel, cadmium, and tellurium sulfide precipitations were followed by ferric hydroxide scavengings on the solution containing sodium which was then evaporated to dryness to sublime away ammonium salts. The resulting NaCl was dissolved and precipitated using butanol saturated with HCl gas, in the presence of magnesium, rubidium, and cesium hold-back carriers. Three sets of ferric hydroxide and calcium, barium, and strontium carbonate scavengings were followed by two more sulfide and ferric hydroxide scavengings. NaCl was reprecipitated and converted into $NaClO_4$ along with potassium, rubidium, and cesium. $NaClO_4$ was preferentially dissolved in butanol, and the sodium was reprecipitated as NaCl. Final traces of potassium, rubidium, and cesium were removed by using a Dowex-50 column and eluting the sodium with 0.5 *M* HCl.

Magnesium

Magnesium was initially separated as $Mg(OH)_2$ and was dissolved in dilute HCl followed by three sets of ferric hydroxide, copper, palladium, and nickel sulfides scavengings. $MgNH_4PO_4 \cdot 6H_2O$ was then precipitated and dissolved in concentrated HNO_3, and any barium and strontium activity accompanying the magnesium was removed by 3–4 $Ba\text{-}Sr(NO_3)_2$ scavengings. Phosphate was removed as zirconyl phosphate. More ferric hydroxide and sulfide scavengings were performed and $MgNH_4PO_4 \cdot 6H_2O$ reprecipitated. It was dissolved in 0.1 *M* HCl and passed through a Dowex-50 column. Magnesium was eluted with 3 *M* HCl and finally passed through a Dowex-1 column and precipitated as $MgNH_4PO_4 \cdot 6H_2O$.

Silicon

The aluminum catcher and blank foils were dissolved in concentrated NaOH to which sodium silicate carrier was added.[50] After carrying out three ferric hydroxide scavengings, $SiO_2 \cdot xH_2O$ was precipitated by adding concentrated H_2SO_4. The precipitate was redissolved in alkali, followed by a ferric hydroxide scavenging and reprecipitation of $SiO_2 \cdot xH_2O$. Silicon was distilled as SiF_4 and trapped in sodium hydroxide and finally reprecipitated as $SiO_2 \cdot xH_2O$.

[49] I. Halpern, in *Proceedings of the Symposium on the Physics and Chemistry of Fission, 1965* (International Atomic Energy Agency, Vienna, 1965).

[50] W. W. Meinke, Radio Chemistry Monograph Series, National Academy of Sciences—National Research Council, 1960 (unpublished).

Sulfur

Sulfur carrier was added both as sulfate and sulfide.[50] The sulfide was oxidized to sulfate with bromine water and initially precipitated as $BaSO_4$. This was metathesized with a sodium carbonate solution. The resulting sodium sulfate solution containing excess sodium carbonate was neutralized with concentrated HCl, and most of the sodium and choride was removed as NaCl by cooling the solution in ice. Sulfate was precipitated as benzidine sulfate by adding a 2% solution of benzidine hydrochloride. The benzidine sulfate precipitate was dissolved in a 6 *M* NaOH solution and was followed by a ferric hydroxide scavenging; the free benzidine was extracted into isopropyl ether. $BaSO_4$ was reprecipitated after neutralizing the NaOH solution containing the sulfate with dilute HCl. The whole cycle of operations was repeated 3–4 times.

Calcium

The Los Alamos procedure[51] was used. The whole cycle of operations was repeated 3–4 times and additional barium and strontium nitrate scavengings were done until the barium-strontium fractions showed no activity. The resulting calcium samples showed <1 cpm (at 40 MeV).

Manganese

Manganese was initially separated as MnO_2 and was purified by both copper sulfide and fast ferric hydroxide scavengings.[50] It was then converted into the carbonate with a sodium carbonate solution and reprecipitated as MnO_2 by oxidizing with potassium chlorate in the presence of hold-back carriers for nickel, arsenic, strontium, barium, rare earths, zirconium, palladium, and molybdenum. The whole cycle of operations was repeated 3–4 times.

Nickel

The Los Alamos procedure[51] was used. Nickel dimethyl glyoxime was precipitated in the presence of citrate and cobalt hold-back carrier and was extracted into chloroform. Nickel was purified by three sets of copper and palladium sulfide and ferric hydroxide scavenging steps and extraction of the dimethylglyoxime complex into chloroform.

[51] Collected Radiochemical Procedures, edited by J. Kleinberg, La-1721, 2nd Edition (1963) and the Supplement, 1966 (unpublished).

Tantalum

Tantalum was separated directly from the uranium target solution by converting the added tantalum carrier into the fluoro-complex with HF and extracting the fluoro-complex into di-isopropyl ketone.[50] It was back extracted into water and precipitated as $Ta_2O_5 \cdot xH_2O$ after a ferric hydroxide scavenging. This cycle of extraction of fluoro-complex, ferric hydroxide scavenging, and precipitation of $Ta_2O_5 \cdot xH_2O$ was repeated 3–4 times. The last three extractions were carried out in the presence of zirconium and niobium hold-back carriers.

Gold

Gold was extracted directly from the uranium target solution as the chloride complex (in 4 *M* HCl) into ethyl acetate. Gold metal was precipitated by dissolving magnesium powder in the presence of hold-back carriers for copper, nickel, palladium, cadmium, tellurium, arsenic, antimony, zirconium, niobium, barium, strontium, cesium, and the rare earths. Gold chloride was reextracted into ethyl acetate in the presence of all of the above hold-back carriers and reprecipitated as the metal. This cycle was repeated 3–4 times. One fast ferric hydroxide scavenging was done from a slightly alkaline (NaOH) solution of gold chloride. Finally, the gold was dissolved and fumed with a HNO_3-$HClO_4$ mixture in the presence of osmium, rhenium, and ruthenium carriers. Gold was finally precipitated as the metal and showed ≤ 0.5 cpm.

Lead

After initial separation as the sulfide, lead was converted into nitrate followed by a few ferric hydroxide scavengings from sodium hydroxide solution.[50,51] It was reprecipitated as the sulfide, oxidized to sulfate, dissolved in ammonium acetate, and the sulfate removed as $BaSO_4$. Lead sulfide was reprecipitated, dissolved in HCl, and a copper and palladium sulfide scavenging was carried out. The whole cycle of operation was repeated 3–4 times and lead was finally precipitated as $PbCrO_4$. The resulting sample showed the presence of some undetermined contaminant, but not any 10.6-h or 3.3-h lead isotopes.

Strontium

The Los Alamos procedure[51] was used without any further modification.