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P. B. Price, R. L. Fleischer, R. M. Walker and E. L. Hubbard

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P. B. Price, R. L. Fleischer, and R. M. Walker
General Electric Research Laboratory

E. L. Hubbard

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INTRODUCTION

On the basis of a charged liquid drop model,[1] one would expect nuclei with very large Z^2/A to undergo fission into three or more fragments. Figure 1, taken from Swiatecki's paper,[1] shows the difference in energy between a spherical drop and two, three, and four widely separated fragments as a function of Z^2/A. On this model ternary fission of both U and Cf are energetically favorable.

Fission into two heavy fragments and a long-range α-particle is well established. A number of attempts have been made to detect fission of a heavy element into three or more fragments of comparable mass. Rosen and Hudson,[2] using triple coincidence counters, measured a ternary/binary ratio of $\sim 7 \pm 3 \times 10^{-6}$ for thermal neutron fission of U^{235}, where only fragments of mass greater than ~40 were counted. Because of experimental difficulties, this result was never confirmed by other workers. Muga et al.[3] scanned ~120,000 spontaneous fissions from Cf^{252} loaded in a nuclear emulsion and found ~75 having three prongs of roughly equal length. An alternative interpretation for three-pronged tracks which was difficult to rule out was that binary fission occurred with one fragment then scattering from a Ag or Br nucleus in the emulsion.

Qualitative arguments by Swiatecki (private communication), using the liquid drop analogy, suggest that the probability of ternary fission should increase rapidly as Z^2/A increases above the value at which it first becomes detectable. We have produced two sets of compound nuclei, A + Pb and A + U, both with Z^2/A considerably larger than that of Cf^{252}, and have made a search for ternary fission, employing a

new detector which registers fission fragment tracks but not tracks of high energy A ions. Experiments are also underway, using this detector, to look for three-way splits in the spontaneous fission of Cf^{252}. In this paper we describe the experimental procedures and the observations of two- and three-pronged events. We then consider various possible explanations for three-pronged tracks and conclude that ternary fission is the only process we have thought of that can occur with observable frequency in the case of A on Pb.

EXPERIMENTAL PROCEDURE

Certain transparent non-conducting solids, such as mica and various glasses, will register tracks of heavy particles such as fission fragments but will not register tracks of light ions.[4,5] The tracks are actually trails of radiation-damaged material which can be "developed" to optically visible size by a preferential chemical etching technique.[6] The chemical reagent dissolves the material along the path of a heavy particle, provided its trajectory intersects a free surface of the track-registering solid.

Bombardments of these solid detectors with various particles having various energies have established that there exist critical rates of energy loss for track registration in the various solids.[7] Visible tracks are made only by ions whose dE/dx is greater than the critical value. Figure 2 shows the experimentally determined thresholds for three detectors—a phosphate glass, mica, and Lexan (a polycarbonate resin).

In a search for ternary fission, we have exploited the discriminating ability of these detectors. Samples of mica and glass were loaded with heavy elements and bombarded with 414 MeV argon ions from the Berkeley Heavy Ion Accelerator. Argon ions of this energy are below the threshold. Three sets of experiments were done to try out various ways of putting heavy elements into the detectors:

(1) Synthetic mica containing ~1 atom percent of Pb dispersed uniformly throughout its volume was obtained from the U. S. Bureau of Mines.

(2) A film of phosphate glass was blown from a melt containing ~1 atom percent U.

(3) A sandwich of mica - U - mica was prepared by partially cleaving a slab of natural mica, inserting ~10^{-3} ml of a solution containing 300 mg/ml of U, and allowing the partially cleaved layers to close.

These samples were exposed at an angle of 30° to ~$10^{11}/cm^2$ 414 MeV A ions, etched in hydrofluoric acid to produce tracks, and scanned at 500x by transmitted light.

In addition, a sandwich of mica - Cf^{252} - mica was prepared in the same way from a concentrated solution of $CfCl_3$. This sandwich was left overnight, then etched and scanned for spontaneous fission tracks from Cf^{252}.

RESULTS

Table I summarizes our observations of two-pronged and three-pronged events in the various detectors and includes the observations of previous workers for purposes of comparison.

Figure 3 shows unretouched micrographs of two- and three-pronged tracks in irradiated Pb-mica. Only a portion of each event is in focus. The argon beam enters the surface from the right. The straight tracks parallel to the beam represent some kind of contamination present in the beam. The argon ion tracks are invisible and are about 20,000 times as frequent as the visible tracks.

A total of 11 three-pronged tracks was seen in Pb-mica and their angles measured. Of these, two were eliminated on kinematic grounds as being possibly due to scattering, leaving 9 tracks which appear to represent genuine ternary fissions. 1860 two-pronged tracks were counted and about 600 straight tracks at a large angle to the beam direction were observed. Some of these may represent binary fissions which were initiated so close to the surface that only one track could be seen. It

Table I.

Nucleus	Z^2/A	Binary	Ternary	Binary/Ternary	Method
$n+U^{235}$	35.8	--	--	$1.4 \pm 0.6 \times 10^5$	Ref 2
Cf^{252}	38.2	~120,000	<75	$\geq 10^4$	Ref 3
Cf^{252}	38.2	3200	0	$>3 \times 10^3$	mica sandwich
A+Pb	40.4	1860	9	$\sim 2 \times 10^2$	synthetic Pb mica
A+U	43.5	250	11	≥ 25	U glass
A+U	43.5	75	0	>75	mica sandwich

is also expected that a few percent of the binary fission events were missed because neither fragment intersected a surface of the mica.

In U-loaded glass the trajectories of fission fragments are poorly known because the etchant produces cone-shaped tracks rather than cyclindrical tracks. The 11 three-pronged events could not, therefore, be subjected to an accurate angular analysis. They represent an upper limit to the number of ternary fissions.

Our failure to see any three-pronged events in the mica-U sandwich suggests that some or all of the events in the U glass may not have been ternary fissions. The sandwich technique appears to be most reliable and further exposures are being made to improve the statistics.

Observations of spontaneous fission tracks in the mica-Cf sandwich are in the early stages and will serve to provide a check on the argon ion results. This technique has two advantages over the loaded emulsion technique: (1) Scattering of heavy particles is greatly reduced over scattering in emulsion because of the much smaller average atomic number of atoms in the mica than Ag and Br atoms. (2) The origin of each fission event is defined by a short gap ~1μ long in each track resulting from the finite separation of the two halves of the mica sandwich. Of about 3200 tracks scanned, only four were forked as a result of scattering by an ion in

the mica sufficiently heavy to register a track. These forks were all several microns away from the origins of the fission events.

DISCUSSION

We now consider various possible sources of the two- and three-pronged events, restricting the discussion to A + Pb, for which most data have been obtained. The velocity of the compound nucleus is high enough so that binary fission tracks should have a V-shaped appearance. From the observed density of V-shaped tracks, we calculate a cross-section of $\sim 3 \times 10^{-24} cm^2$, which strongly suggests that they arise from binary fission. The included angles and orientations of several V-tracks have been measured. All of these events lie in planes containing the incident beam and all but one have the proper angles to be caused by fission of a compound nucleus into two equal fragments.

In considering the three-pronged events, only the angles of the tracks are reliable. The track lengths provide only lower limits to fragment ranges, because the etching does not always extend to the end of the ranges.

If a binary fragment is scattered off a heavy atom in the mica, both the fragment and the struck atom may leave a track. In order to be mistaken for a ternary fission, this event must take place within ~0.5 to 1μ of the vertex of the "V". In none of the 3200 spontaneous fission tracks of Cf^{252} was a fragment scattered closer than ~3μ from the fissioning nucleus, and in none of the 1860 V-tracks was a fork observed along either arm of the V. The observed ratio of 1 three-pronged event for every 200 binary fissions is too high for the three-pronged events to have been caused by scattering of a binary fission fragment.

Binary fission can be produced by an α transfer process. However, the scattered part of the projectile would not lose enough energy to make a visible track until it had traveled some distance from the fission, and a three-pronged event would not be seen.

A heavy background particle might scatter from a heavy constituent of the mica and leave a three-pronged event. A scattering event of this type must be coplanar in order to conserve momentum. For seven of the three-pronged events, the angle between one of the tracks and the plane of the other two was larger than the error in the measurement of this angle. Two other three-pronged events were nearly coplanar but could not be a scattering, because all three prongs were within an angle of less than 180°.

Events that appear to have 3 prongs might be produced by the superposition of the end of a background track on the vertex of a binary fission. Since none of the tracks in the three-pronged events observed is parallel to the beam, only background tracks that are not parallel to the beam need be considered. Background tracks not parallel to the beam can be produced by recoil particles from the collision of the high energy argon ions with heavy constituents of the mica. The 414 MeV argon ions cannot lose enough energy in a scattering collision to become detectable. The number of background tracks observed that are not parallel to the beam is low enough so that the probability of one being superimposed on a binary fission is much less than the observed frequency of three-pronged events.

The possibility that two of the tracks in the three-pronged events could be made by a binary fission can be ruled out in most cases by requiring conservation of momentum in the binary fission. In binary fission both fragments cannot come off on the same side of the beam trajectory and the minimum angle between binary fission fragments is 132°.

Superposition of a single track on some other type of event such as a scattering of a detectable particle would be even less probable.

We thus conclude that ternary fission of an excited A + Pb compound nucleus occurs with a higher frequency than has been reported previously for other nuclei. We expect that heavier compound nuclei should fission into three fragments even more frequently and we are presently employing the sandwich technique with uranium and plutonium.

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FOOTNOTES AND REFERENCES

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FIGURE CAPTIONS

Fig. 1. The energy released in the division of a drop into n equal parts.

Fig. 2. Critical rates of energy loss, dE/dx, for track registration in various solid detectors. The threshold for Lexan is still uncertain and may be somewhat lower than indicated.

Fig. 3. Events which are believed to represent ternary and binary fission of A + Pb compound nuclei in a synthetic mica containing ~1% Pb. The straight tracks define the direction of the incident beam. About 10^6 argon ions have passed through each of the fields of view.

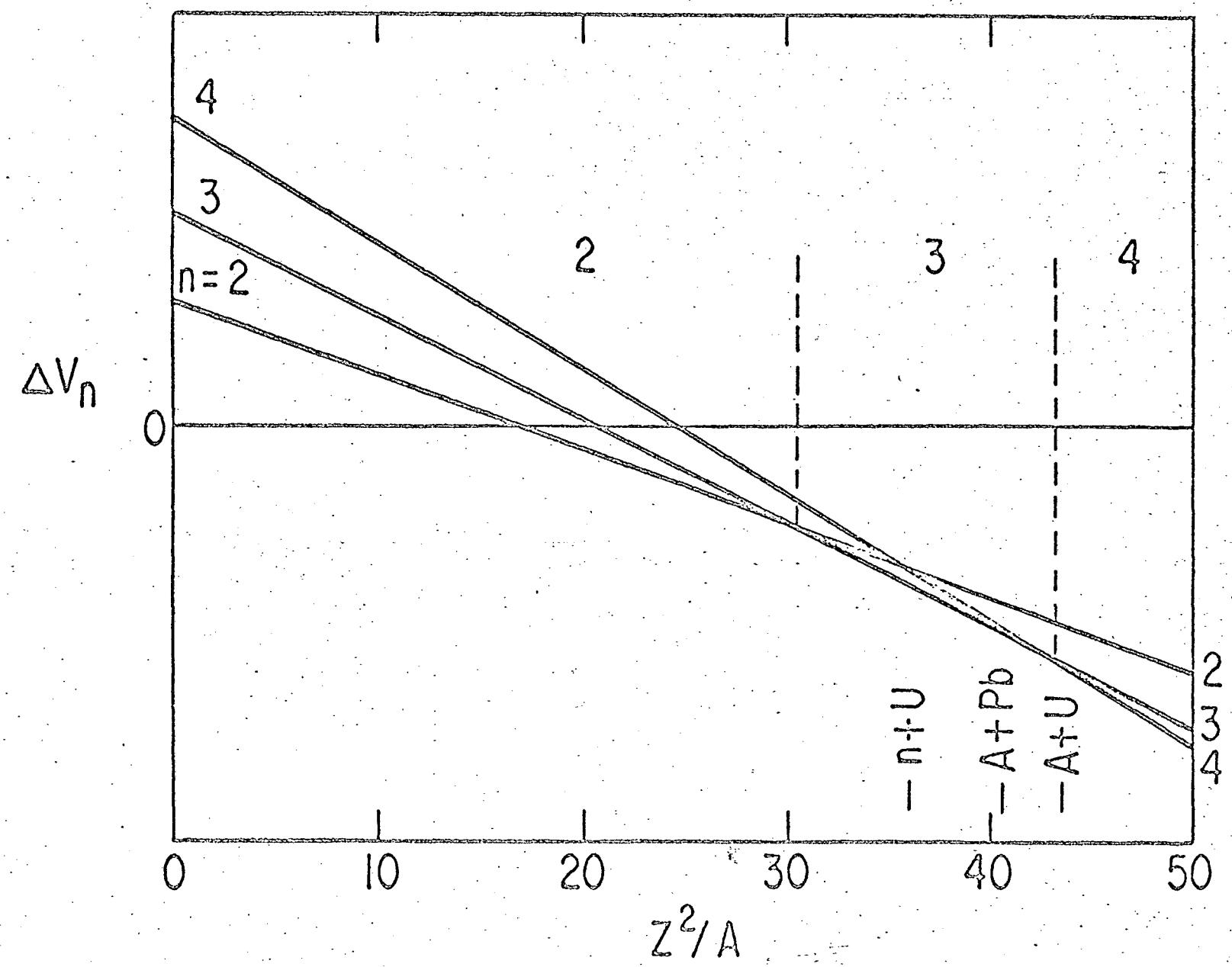

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Fig. 1

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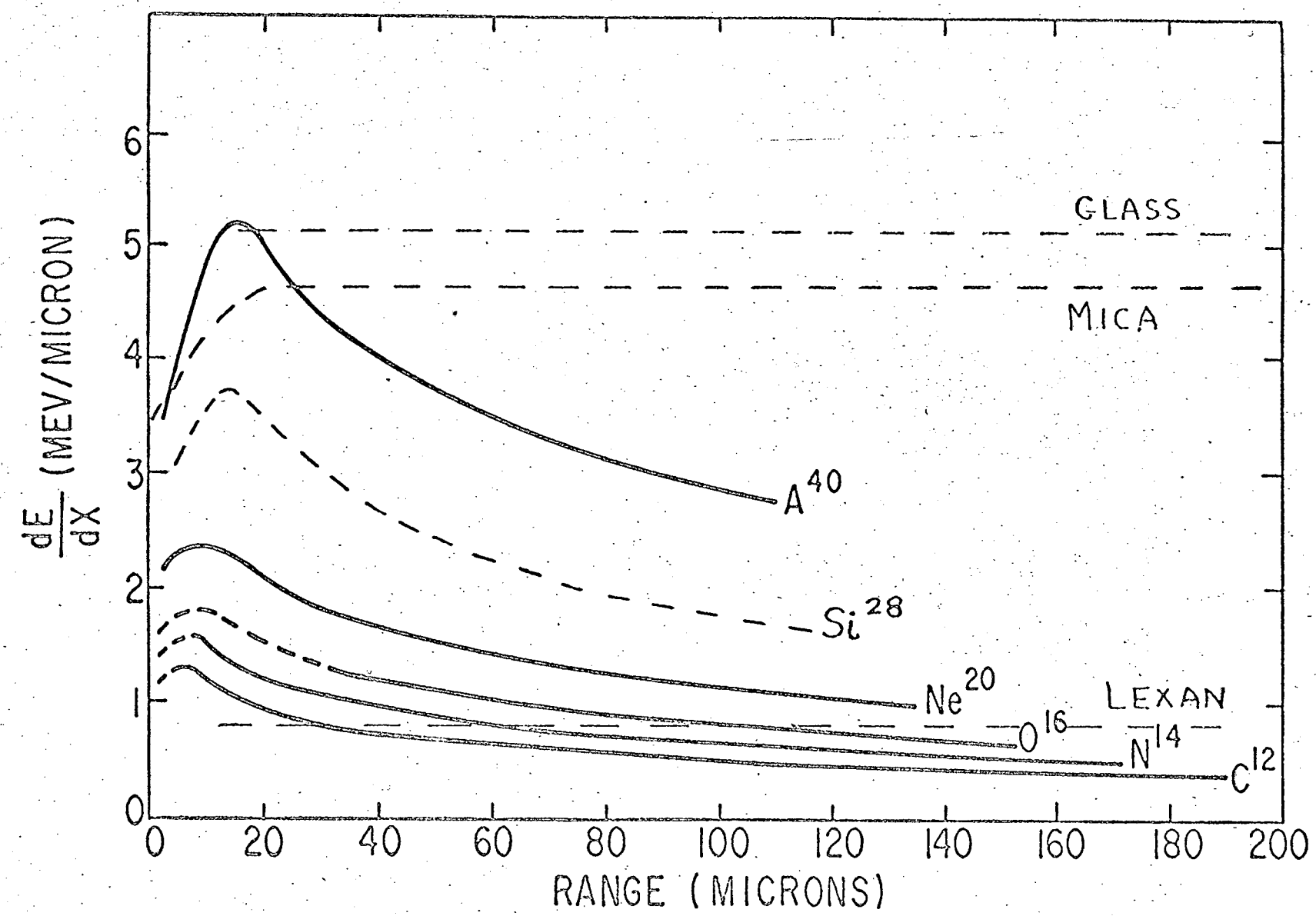

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Fig. 2

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