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Tripartition in the Spontaneous-Fission Decay of Cf^{252}†

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The long-range alpha particles associated with the spontaneous-fission decay of californium-252 have been studied by means of nuclear-emulsion techniques. The alpha energy spectrum was found to peak at about 19 Mev with a half-width of 10 Mev, and the preferential angle of emission was found to be slightly less than 90 deg with respect to the light fission fragment. These results support the view that alpha emission occurs at the time of scission and that the direction is determined by the extent of electrostatic repulsion by the fragments. A trend toward a more nearly symmetric mass division in fission accompanied by alpha emission is indicated. The frequency of occurrence of the long-range alpha particles was observed to be 1 in 415±10% binary fissions.

Ternary events consisting of two heavy fragments and one light fragment of short range were observed, but the frequency with which these events occur was not measured. A parallel search was made for ternary-fission events in which fragmentation into comparable masses occurs.

INTRODUCTION

THE liquid-drop model[1] was used by Present[2,3] in 1941 to predict that fission into three charged fragments of comparable mass is dynamically possible. Subsequent experimental observations[4] have indicated the existence of multiple fission modes which may be grouped into four types as follows: (a) ternary fission in which the third fragment is a long-range alpha particle, (b) tripartition in which the third fragment is a short-range charged particle of small mass, (c) fission into three charged fragments of roughly equal mass, and (d) multiple fission in which fragmentation into four (quaternary fission) or more charged particles takes place.

The existence of type (a) events, first reported in the literature by Green and Livesey[5] and Tsien *et al.*[6] (who studied fission induced in U^{235} by thermal neutrons) is well established.

Type (b) events, tripartition with the fragment of small mass and short range, was studied by the same two groups and confirmed by the work of Cassels *et al.*[7] and by Allen and Dewan.[8] However, Marshall takes issue with these earlier results on the basis of possible errors in measuring these events and regards the events as arising from recoil interactions of binary-fission fragments.[9] Laboulaye *et al.*, using cloud chamber techniques, were unable to detect such events.[10]

Tsien *et al.* have reported a single case of tripartition into roughly equal fragments [type (c) events], and no frequency of occurrence was given.[11] Rosen and Hudson, using a triple ionization chamber and coincidence circuitry, observed a value of 6.7 ternary fissions per 10^6 binary fissions,[12] but because of the low frequency of occurrence, confirmation by other means has not been made.

Tsien *et al.* have reported cases of quadripartition into roughly equal masses as occurring with a frequency of 1 per 3000 binary fissions[13,14]; however, Titterton was unable to confirm these results.[15] The frequency (comparable to that for tripartition) reported by Tsien seems too high, and no arguments are given to preclude the possibility of double recoil by binary-fission fragments.

Even the more recent investigations have been concerned with *induced* fission (mainly in U^{235} with thermal neutrons) and, hence, with excited compound nuclei.[16–20] Therefore, it seemed worthwhile to study multiple-fission modes of *spontaneous* fission. The most convenient isotope available for this purpose is Cf^{252}, which has an alpha half-life of 2.2 yr and a spontaneous-fission half-life of 70 yr.[21] A study has been made, therefore, of the long-range alpha particles associated with spontaneous fission in Cf^{252} using nuclear-emulsion

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1 N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
2 R. D. Present and J. K. Knipp, Phys. Rev. **57**, 751 (1940).
3 R. D. Present, Phys. Rev. **59**, 466 (1941).
4 For a brief but thorough summary of the earlier work see K. W. Allen and J. T. Dewan, Phys. Rev. **80**, 181 (1950).
5 L. L. Green and D. L. Livesey, Nature **159**, 332 (1947).
6 S. T. Tsien, R. Chastel, Z. W. Ho, and L. Vigneron, Compt. rend. **223**, 986 (1946).
7 J. M. Cassels, J. Dainty, N. Feather, and L. L. Green, Proc. Roy. Soc. (London) **A191**, 428 (1947).
8 K. W. Allen and J. T. Dewan, Phys. Rev. **82**, 527 (1951).
9 L. Marshall, Phys. Rev. **75**, 1339 (1949).
10 H. Laboulaye, C. Tzara, and J. Olkowsky, J. phys. radium **15**, 470 (1954).
11 S. T. Tsien, Z. W. Ho, R. Chastel, and L. Vigneron, J. phys. radium **8**, 165 (1947).
12 L. Rosen and A. M. Hudson, Phys. Rev. **78**, 533 (1950).
13 Z. W. Ho, S. T. Tsien, L. Vigneron, and R. Chastel, Compt. rend. **223**, 1119 (1946).
14 S. T. Tsien, Z. W. Ho, R. Chastel, and L. Vigneron, Compt. rend. **224**, 272 (1947).
15 E. W. Titterton, Nature **170**, 794 (1952).
16 K. F. Flynn, L. E. Glendenin, and E. P. Steinberg, Phys. Rev. **101**, 1492 (1956).
17 C. B. Fulmer and B. L. Cohen, Phys. Rev. **108**, 370 (1957).
18 G. F. Denisenko, N. S. Ivanova, N. R. Novikova, N. A. Perfilov, E. I. Prokoffieva, and V. P. Shamov, Phys. Rev. **109**, 1779 (1958).
19 V. N. Dmitriev, L. V. Drapchinskii, K. A. Petrzhak, and Yu. F. Romanov, Doklady Akad. Nauk S.S.S.R. **127**, 531 (1959) [translation: Soviet Phys. Doklady **4**, 823 (1960)].
20 E. L. Albenesius, Phys. Rev. Letters **3**, 274 (1959).
21 D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958).

techniques. A parallel search was made for other types of tripartition in Cf^{252}.

EXPERIMENTAL

The following experimental procedure was used. A volume of 0.01 cc of a solution containing a small amount (10^4 fissions/min) of Cf^{252} was diluted to 5 cc with $0.6M$ sodium citrate solution (pH 5). Two cc of this solution was placed on a 1- by 3-in. Ilford KO emulsion (200 microns thick) fitted with a plastic rim to contain the liquid.

After 1 hr of contact, the excess solution was removed and the emulsion surface washed. The emulsion was dried 1 hr in a desiccator containing concentrated H_2SO_4 and equipped with a fan to provide adequate air circulation. After a 48-hr exposure, the emulsion was developed with a modified Brussels-type developer.[22]

Upon fixing and final washing and drying, the shrinkage factor was obtained for each plate by measuring the tracks produced by the 6.11-Mev alpha particles from the branching decay of Cf^{252}. The plates were then scanned systematically for unusual events by viewing the emulsions through a 10× eyepiece and either a 98× or 45× objective. The nature of the tracks of fission events accompanied by long-range alpha emission was such that detection and identification was easily made with a 45× objective. In the case of other three-pronged events, scanning was done with a total magnification of 980×. Since three-pronged events may result from scattering by binary-fission fragments, an initial selection was made on the basis of the location of the least dense track with respect to the base formed by the other two tracks. If tripartition into roughly equal masses occurs, the third track would be expected to originate near the middle of the track. Hence, only those events were analyzed in which the least dense track appeared to originate from the center 5 microns of the fission track. (Thus, a large number of events arising from scattering by binary-fission fragments near the end of their paths were eliminated.)

In general, these events do not lie entirely within the focal plane of the microscope, and hence for analysis, the projected lengths, projected angles, and the depths of the origin and ends of the tracks were measured. From these projected measurements and the shrinkage factor, the true angles and ranges were computed.

The frequency of occurrence for the long-range alpha events was determined as follows: A vaporized source of Cf^{252} (10^5 fissions/min) was covered tightly with aluminum foil (9.6 mg/cm^2) just sufficient to stop the 6.11-Mev alpha particles emitted by Cf^{252}. The more energetic long-range alpha particles were then counted

[22] This developer consists of a mixture of 35 g boric acid, 15 g sodium sulfite, 4.5 g Amidol, and 8 cc of 10% potassium bromide solution, dissolved and diluted to 1000 cc with water.

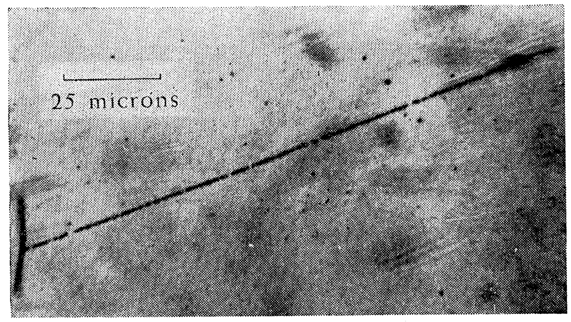

FIG. 1. Photomicrograph in emulsion of fission of Cf^{252} with emission of long-range alpha particle.

by using a standard ionization chamber of known geometry. A frequency of one long-range alpha particle (i.e., of energy greater than 10 Mev) per 415±10% binary fissions was found. This value agrees with that found by Nobles—one per 450 binary fissions[23]—and is comparable to the frequency observed in the fission of U^{235} induced by thermal neutrons.[24,25]

RESULTS AND DISCUSSION

A. Fission with Emission of Long-Range Alpha Particle

A photomicrograph of a typical fission event in which a long-range alpha particle is emitted is shown in Fig. 1. An energy spectrum for the long-range alpha particles was determined from the analysis of 203 events and is shown in Fig. 2. For comparison, a replot is shown of the energy spectrum of the long-range alpha particles associated with the slow-neutron-induced fission of U^{235}.[25] For fission of Cf^{252}, the most probable energy of the alpha particle is 19±1 Mev, some 4 Mev greater than that for the long-range alpha particles from the fission of U^{236*}; the maximum observed energy was 34 Mev as compared with a maximum of 29 Mev observed by Titterton for the alpha particles from U^{236*} fission.[25] It should be noted that the maximum observed energy from emulsion work[25] is 2 to 3 Mev higher than that observed by ionization-counter methods.[24,26] No explanation is immediately apparent in view of the fact that the widths at half maximum are the same for both techniques, thus eliminating as an explanation the possibility of excessive range straggling in emulsion measurements.

From an angular correlation of the long-range alpha particles with both the heavy and light fission fragments (Fig. 3), it appears that the most probable angle of emission is slightly less than 90 deg to the light frag-

[23] R. A. Nobles, Los Alamos Scientific Laboratory, Los Alamos, New Mexico (unpublished), (private communication, September, 1959).
[24] K. W. Allen and J. T. Dewan, Phys. Rev. **80**, 181 (1950).
[25] E. W. Titterton, Nature **168**, 590 (1951).
[26] C. B. Fulmer and B. L. Cohen, Phys. Rev. **108**, 370 (1957).

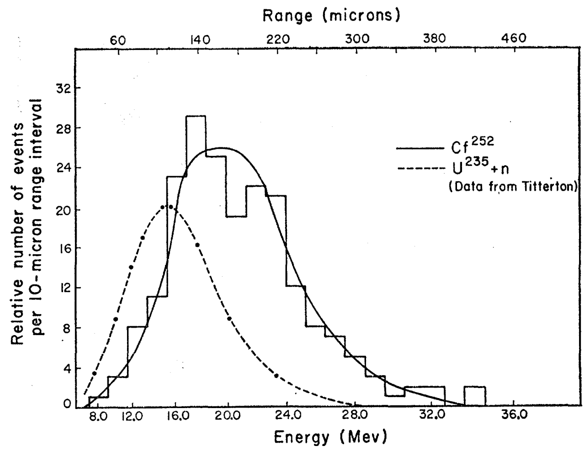

FIG. 2. Energy spectrum of long-range alpha particles associated with fission. Solid curve, Cf^{252}; dashed curve, $U^{235}+n$ (data from Titterton[25]).

ment. The existence of this maximum in Fig. 3 again supports the view (originally proposed by Tsien)[27] that alpha emission occurs at the time of scission of the two fission fragments and that the alpha particle is subsequently accelerated by the electrostatic fields in a direction that is most likely to be approximately perpendicular to the fission-fragment paths.

According to Fig. 4, the average total length of fission-fragment tracks from alpha-emitting fission events appears to be 1 micron shorter than the average fission track length for binary fission. This is a decrease of 4% in total range (corresponding to a 6% decrease in energy). For the slow-neutron-induced fission of U^{235}, Marshall has reported a corresponding decrease of 6% in range.[9] Thus, as previously suggested, the long-range alpha particle apparently receives its energy at the expense of decrease in the resulting fission-fragment ranges.

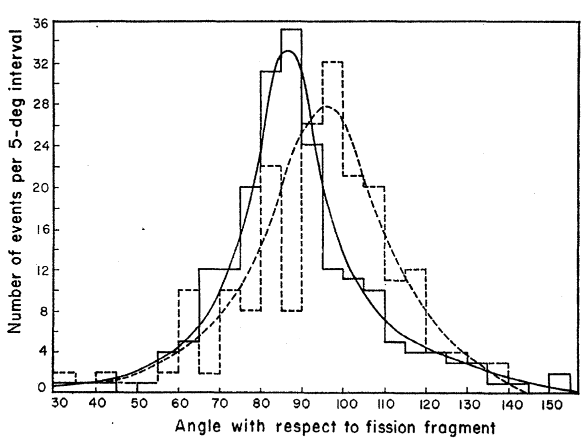

FIG. 3. Number of events per interval vs angle of alpha particle with respect to fission fragment for Cf^{252}. Solid curve, light fragment; dashed curve, heavy fragment.

[27] S. T. Tsien, Compt. rend. **224**, 1056 (1947).

Previous investigators have studied the effect of alpha-particle emission on the mass division in fission of U^{235} induced by thermal neutrons.[9,14,28] A replot of combined data by Marshall,[9] Tsien *et al.*,[14] and Wollan *et al.*[28] is given in Fig. 5, in which the number of events vs the ratio R is shown. Here we define $R=R_L/(R_L+R_H)$ where R_L is the range of the light fission fragment and R_H is the range of the heavy fission fragment. These workers have effectively compared the mean values of R (marked by arrows in Fig. 5) for fission with alpha emission and binary fission and have found only a small difference. They have attached no significance to the slight shift indicating a more nearly symmetric mass division when alpha particles are emitted in the fission process.

However, a similar effect is observed for the spontaneous fission of Cf^{252} (Fig. 6) in which a direct comparison can be made of the two types of fission.[29] Again, the difference in the mean values of R (as

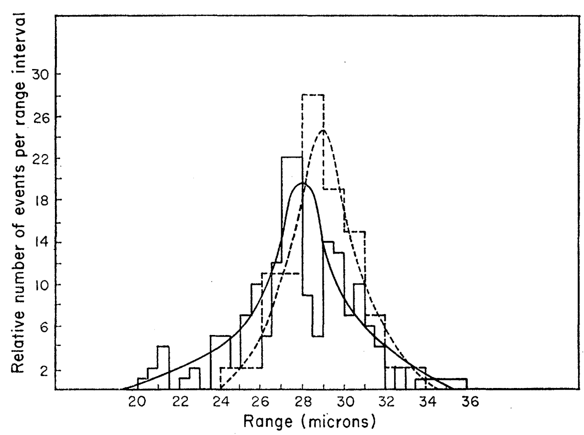

FIG. 4. Fission-fragment total track length for fission with alpha emission (solid curve) and ordinary fission (dashed curve) of Cf^{252}.

marked by arrows) for the two fission modes is not large. Although a trend toward a more symmetric mass division is indicated in the case of alpha emission, we do not feel that the data are sufficient to support a definite conclusion.

B. Tripartition with Emission of Short-Range Charged Particles of Small Mass

In the present study, no examples of lithium-8, boron-8, or beryllium-8 emission were observed among the 10^5 binary-fission events scanned. The special nature of the tracks of these events are such that they would have been easily detected.[30,31]

[28] E. O. Wollan, C. D. Moak, and R. B. Sawyer, Phys. Rev. **72**, 447 (1947).

[29] The curve for binary fission has been derived from range (in air)-mass data by J. A. Miskel and K. V. Marsh, Lawrence Radiation Laboratory (private communication, 1959) and is normalized to give equal areas under the curves.

[30] F. K. Goward, E. W. Titterton, and J. J. Wilkins, Nature **164**, 661 (1949).

[31] E. W. Titterton, Phys. Rev. **83**, 1076 (1951).

Tripartition with emission of a light charged particle of short range was observed (as reported by previous investigators in the case of U^{235} fission induced by slow neutrons)[4–7]; the frequency of these events was not studied. Examples of these events are shown in Fig. 7.

C. Ternary Fission into Comparable Masses

Of approximately 120 000 binary fission events scanned, 75 three-pronged events were considered as possible tripartition into roughly equal masses. Photomicrographs of typical events are shown in Fig. 8. Using an angular analysis described elsewhere,[32] four of these events could not be attributed to nuclear recoil collisions of a fission fragment with an emulsion nucleus. However, the short lengths (of the order of 10 microns) of the fission-fragment paths, the size of the individual grains (about 0.5 micron in diameter), small-angle scattering, and inherent errors in microscope measurements all contribute to an uncertainty in such an analysis. Confirmation of this type of tripartition is presently being investigated in this laboratory by use of solid-state detectors and triple-coincidence circuitry.

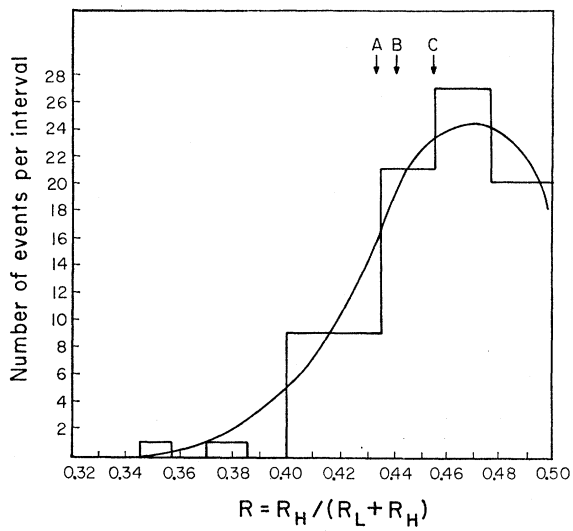

FIG. 5. Number of events vs R for fission fragments associated with long-range alpha-particle emission in fission of U^{235} induced by thermal neutrons. (A) Mean value of R for ordinary fission from J. K. Bøggild, K. H. Brostrom, and T. Lauritsen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **18**, No. 4, 1 (1940). (B) Mean value of R for ordinary fission from P. Demers, Phys. Rev. **70**, 974 (1946). (C) Mean value of R for fission accompanied by alpha emission.

SUMMARY

The main features of alpha-particle emission accompanying the spontaneous fission of Cf^{252} are similar to those of U^{235} induced by thermal neutrons. Fission with alpha particle (>10 Mev) emission occurs at the rate of 0.24% relative to binary fission. The energy spectrum of the long-range alpha particle was observed to peak at 19±1 Mev, with a width at half maximum of 10 Mev; the maximum observed energy was 34 Mev. The most probable angle of emission is slightly less than 90 deg relative to the lighter fission

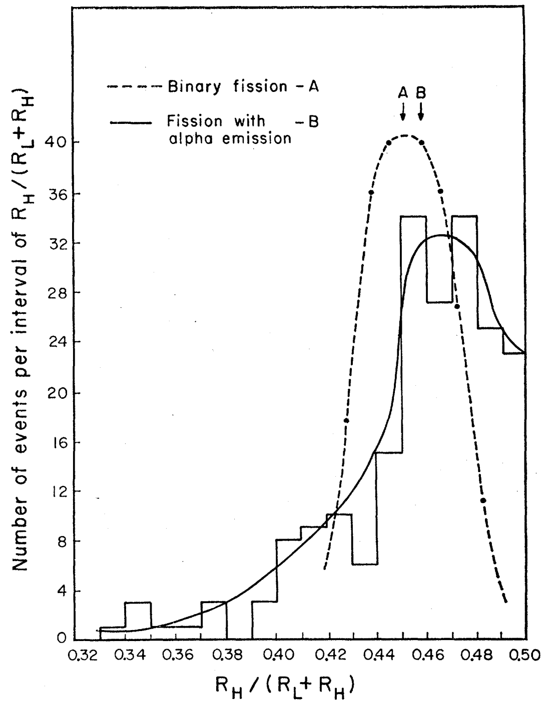

FIG. 6. Number of events vs R for fission fragments associated with long-range alpha-particle emission in Cf^{252}. Dashed curve, binary fission of Cf^{252}; A denotes mean value of R. Solid curve, fission accompanied by alpha emission; B denotes mean value of R.

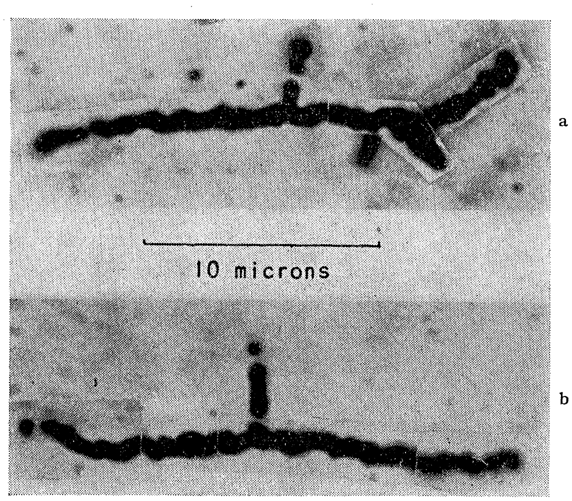

FIG. 7. Example of fission of Cf^{252} with emission of light charged particle of short range.

[32] N. A. Perfilov, in *Physics of Nuclear Fission* (Pergamon Press, New York, 1958), Chap. 7, p. 84.

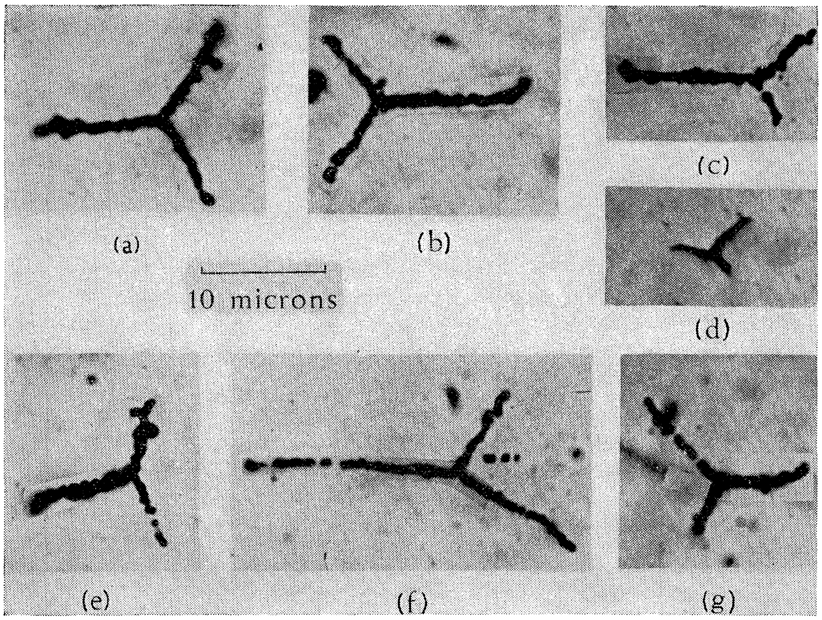

FIG. 8. Examples of three-pronged events.

fragment. A trend toward more symmetric mass division in fission associated with alpha-particle emission is indicated.

Fission events were observed in which a third particle of short range and small mass was emitted; the frequency was not measured. Some evidence was obtained for the existence of ternary fission into roughly equal masses.

ACKNOWLEDGMENTS

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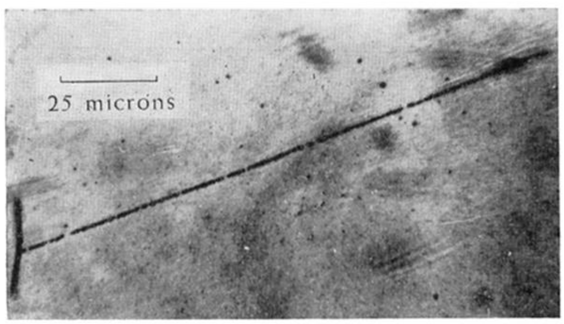

FIG. 1. Photomicrograph in emulsion of fission of Cf^{252} with emission of long-range alpha particle.

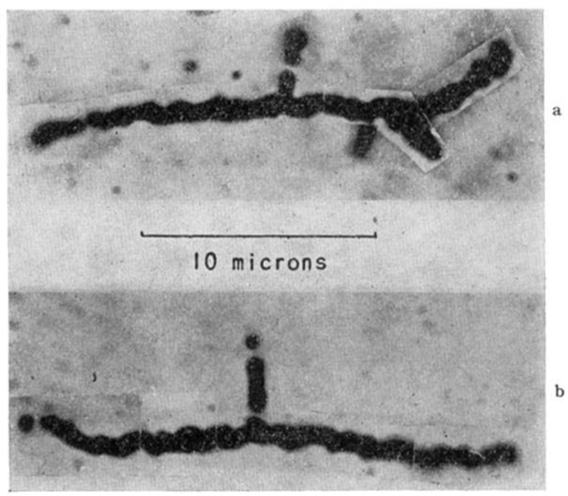

FIG. 7. Example of fission of Cf^{252} with emission of light charged particle of short range.

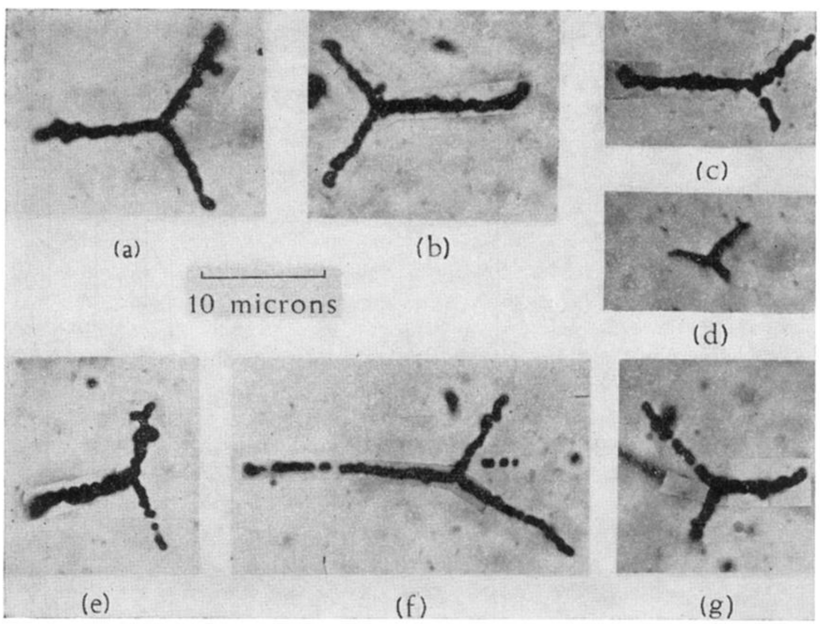

FIG. 8. Examples of three-pronged events.