which originates in the pumping plant. The fact that the layer is much more transparent to electrons than a similar thickness of carbon shows that it contains a high proportion of atoms with a scattering power less than that of carbon, which would be the case if the layer consisted of hydrocarbon molecules.

The thickness of layer A is about 25 Å. and that of layer B about 10 Å. This suggests that layer B is monomolecular, since it is probable that the molecules would be about 10 Å. in diameter ; a monolayer can thus form in the system in 1 sec., but it appears that a dynamic equilibrium is reached within 20 min. when a layer two to three molecules thick has been deposited.

The direct examination of contaminant layers in this way could be useful in determining the efficiency of a vacuum system with regard to the amount of hydrocarbon vapour present.

It has been found that the technique described above can be used as a method for measuring the thickness of very thin evaporated films to a high degree of accuracy. It can be seen from Fig. 2 that the lines, which are very sharp, can be used as reference marks for measuring the thickness of the carbon deposit. For example, the distance between lines C and D is 7 mm. thick on the micrograph, representing a film thickness of 175 Å. This will equal the thickness of carbon deposited on a substrate placed horizontally beside the step, since the angle of evaporation in this case was 45° (see Fig. 1). By lowering the angle of deposition, the ratio of the horizontal to vertical thickness will be altered geometrically so that a greater thickness exists on the vertical step face than on the horizontal target. With a suitable angle a thickness 'magnification' of about three times could be attained without difficulty on the step face, so that a measurement of 60 Å. in the electron microscope to an accuracy of 9 Å. would give a target thickness of 20 Å. to within 3 Å.

A preliminary comparison between thickness measurements carried out on the same carbon film using an optical method and that described here gives a reasonable correlation :

Optical method	Electron microscope method
223 Å.	206 Å.
140 Å.	110 Å.
121 Å.	118 Å.

The optical method employed a shearing interferometer microscope[2], which is at least as accurate as the conventional Fizeau fringe technique[3].

It has been found that the electron microscope method is satisfactory for silicon monoxide and platinum/carbon[4] films ; aluminium films have also shown promising results.

I thank Miss P. Rush for carrying out the comparative measurements, and Dr. T. E. Allibone, director of the Laboratory, for permission to publish this communication.

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[1] Ennos, A. E., *Brit. J. App. Phys.*, **4**, 101 (1953).
[2] Dyson, J., *J. Opt. Soc. Amer.*, **47**, 557 (1957) (see pp. 561–62).
[3] Tolansky, S., "Multiple Beam Interferometry" (Clarendon Press, Oxford, 1948).
[4] Bradley, D. E., *Brit. J. App. Phys.*, **10**, 198 (1959).

Rare Modes in the Spontaneous Fission of Californium-252

CALIFORNIUM-252, a man-made trans-uranic element, having an α-particle half-life of 2·2 yr., undergoes spontaneous fission. Approximately one fission occurs per 38 α-particles, and the material is therefore well suited to an investigation of the possibility of rare modes of spontaneous fission.

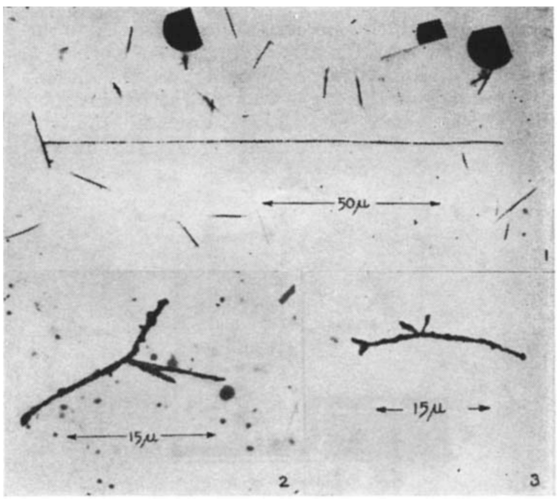
In slow-neutron fission[1,2], fast-neutron fission[3] and photofission[4,5], two rare fission modes have been established. In one the emitted particle is long-range and uniquely an α-particle ; in the other, short-range particles, apparently light elements, appear to be emitted in the process. There is also, in slow-neutron fission, some evidence for fission into three particles of approximately equal mass and kinetic energy, but this is not unambiguous[6].

Observation of the quaternary fission of uranium-235 was claimed in 1946 by a French group[7], but later work in this laboratory[8] showed that, if it occurred, the frequency was much less than that estimated from the French investigation.

In the present experiment, where minute traces of californium-252 were incorporated into Ilford K_0 and E_1 emulsions, the conditions of observation were more favourable than in any of the earlier nuclear emulsion studies. Since the plates required no irradiation, they contained only the californium-252 fission and α-tracks plus the usual slight background from inevitable radioactive inclusions. Random association of tracks yielding 'spurious' events was therefore greatly reduced and could be virtually eliminated by measurement of a range of distributions of tracks in the emulsion.

In a study of 50,000 binary events in these emulsions the following types of event have been recorded :

(1) 179 cases have been observed of ternary fission into two heavy fragments and a long-range α-particle ; the frequency is 1 per 280 (± 25) spontaneous binary fissions. The mode appears to be significantly more frequent than in the case of slow-neutron fission of uranium-235, uranium-233 or plutonium-239 (ref. 8). An example is shown in Fig. 1.



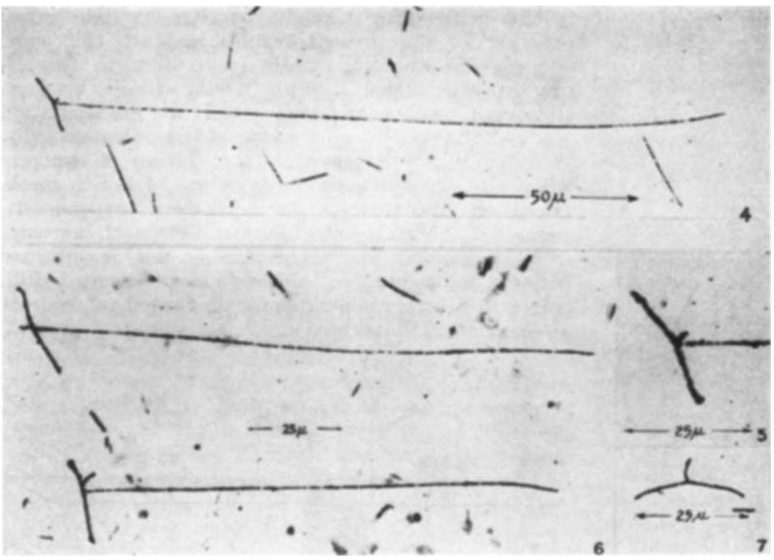
(1) Ternary fission of californium-252
(2) Probable quaternary fission of californium-252
(3) Projection of recoils in emulsion shortly after fission giving appearance similar to a quaternary fission

(2) Eight good examples and four additional possible cases of fission into four heavy fragments (Fig. 2) have been observed. Apart from quaternary fission, the only other possible explanation for these events is that they arise from recoils projected in the emulsion by the pair of fission fragments within $\frac{1}{2}\mu$ of the origin. Two experimental results oppose such an explanation. Although events of this general type do occur (as is indicated by Fig. 3 but where the 1μ spacing between the forks clearly eliminates the event as quaternary), their observed frequency is too low to explain the four-particle events. Moreover, if scattering accounted for such events, then in the earlier study of 600,000 uranium-235 binary fissions[9], some 100 such events should have been observed; in fact only 2 were recorded. We therefore believe the events to be true spontaneous quaternary fission, the frequency being roughly 1 per 5,000 binary fissions.

(3) Three events of the type Fig. 4 (and see enlargement of origin, Fig. 5) have been observed. Again these might be explained as an event of type 1 with a recoil occurring near the origin, and such spurious cases do occur as indicated in Fig. 6. However, during our earlier work where more than 10^6 binary fissions were examined, only one other example of such an event was observed. These events may, therefore, represent a mode of spontaneous quaternary fission involving emission of a long-range α-particle and a short-range light nucleus in addition to the two fission fragments.

(4) As in previous experiments, a considerable number of events are observed which could be interpreted as ternary fission, the third fragment being a short-range light nucleus. The frequency of occurrence of such tracks at the origin appears to be greater than would be expected from Rutherford scattering. In addition to these events, which occur at a rate of 1 per 80 ($\pm$ 20) binary fissions, others have been observed which could be ternary fission into three roughly equal parts. Analysis of the type employed by Catala *et al.*[6] indicates that many of these arise through fission fragments colliding with silver or bromine nuclei in the emulsion. However, it does not exclude all, and it could be that this mode of fission exists in californium-252.

The experiments are continuing, and will be reported in detail later. We wish to thank Prof. E. M. McMillan and Dr. S. G. Thompson, of the University of California, who provided the californium for these experiments.



(4) Possible fission into two heavy fragments, a long-range light particle and a short-range light nucleus
(5) Enlargement of origin of (4)
(6) Examples of a ternary event of type (1) with a recoil track close to origin, easily distinguishable from (4)
(7) Case of possible ternary fission into three roughly equal fragments

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[1] Titterton, E. W., *Nature*, **168**, 590 (1951). Demers, P., "Ionographie, Les Emulsions Nucléaires", 353 (Montreal University Press, 1958).
[2] Fulmer, C. B., and Cohen, B. C., *Phys. Rev.*, **108**, 370 (1957).
[3] Titterton, E. W., *Phys. Rev.*, **83**, 673 (1951).
[4] Titterton, E. W., and Brinkley, T. A., *Phil. Mag.*, **41**, 500 (1950).
[5] Titterton, E. W., and Goward, F. K., *Phys. Rev.*, **76**, 142 (1949).
[6] Catala, J., Casanova, J., and Dimingo, V., *Nature*, **184**, 1057 (1959).
[7] Tsien, S. T., Ho, Z. W., Chastel, R., and Vigneron, J., *J. Phys. Rad.*, **8**, 165 and 200 (1947).
[8] Allen, K. W., and Dewan, J. T., *Phys. Rev.*, **80**, 181 (1950).
[9] Titterton, E. W., *Nature*, **170**, 794 (1952).

The Second Virial Coefficient of Methane at Low Temperature

THE second virial coefficient of methane has been determined over a range of temperatures from 108° to 250° K. by means of an apparatus which is described elsewhere[1].

The principle of the experiments consists in measuring the difference in the expansion of volume of the gas under study and a reference gas. In our experiments we chose hydrogen as reference substance. The working pressure is less than one atmosphere; this allows us to neglect coefficients of higher order than the second in the expansion of the compressibility factor pV/nRT, where p is pressure V, is volume, R is the gas constant, T is absolute temperature and n is number of moles.

Methane was obtained by distillation at low temperature of commercial 'pure grade' gas. The melting of the solidified sample we used took place over a range of less than 1×10^{-3} °K. The vapour pressure at the triple point was 87·58 mm. mercury. The value of 87·60 $\pm$ 0·10 mm. mercury was found at the National Bureau of Standards of Washington by Armstrong, Brickwedde and Scott[2] with a carefully purified sample. The purity estimated from the range of freezing point was better than 99·9 per cent (ref. 3). The hydrogen used was 'research grade' gas of 99·99 per cent purity.

Values obtained for methane are listed in Table 1. In Fig. 1 these values are reproduced for compari-