The Emission of Light Charged Particles in the Slow Neutron Fission of Uranium

L. L. Green and D. L. Livesey

Phil. Trans. R. Soc. Lond. A 1948 **241**, 323-343
doi: 10.1098/rsta.1948.0022

THE EMISSION OF LIGHT CHARGED PARTICLES IN THE SLOW NEUTRON FISSION OF URANIUM

By L. L. GREEN and D. L. LIVESEY
Cavendish Laboratory, University of Cambridge

(*Communicated by N. Feather, F.R.S.—Received* 2 *February* 1948)

[Plate 11]

This paper describes the extension of the photographic plate technique to the recording of fission fragment tracks and the application of this method to the study of rare modes of fission in the case of the slow neutron fission of uranium. The emission of a short-range group of light particles in the fission of U^{235}, reported by Cassels, Dainty, Feather & Green, is confirmed and the angular and range distributions of these particles described. In addition, the emission of long-range light particles with ranges up to 45 cm. of air is described. The abundance of this mode of fission is 1 in 340 ± 40 fission events. The angular and range distributions of these long-range particles are given.

1. Introduction

The fission of heavy nuclei into two charged fragments (binary fission) has been investigated theoretically on the basis of the 'liquid-drop' nuclear model by Bohr & Wheeler (1939), who studied the effect on the stability of the liquid drop of surface deformations described by low-order spherical harmonics. Present & Knipp (1940) pointed out that, in some cases, the deformations may be of such a form that fission into three or more charged fragments occurs. Evidence for the emission of a third charged particle in fission was first obtained in America by Alvarez and by Farwell, Segré & Wiegand (1946). These workers employed coincidence-counting methods and detected the emission of long-range light particles in the fission of U^{235}, but their results did not become generally available until March 1947, when the experimental work described in this paper had been completed. The emission of short-range particles in fission was detected by Cassels, Dainty, Feather & Green (1947), who, using coincidence-counting methods, found that in the fission of the uranium isotope, U^{235}, by slow neutrons, light charged particles were emitted in addition to the two heavy fragments in approximately 4 % of fission events, supposing the emission to be isotropic. Some results were obtained which suggested that the 'secondary' charged particles were α-particles with an energy of approximately 1 MeV.

The counting experiments were carried out in the presence of a high background flux of fast neutrons and the statistical accuracy of the results was low; it was therefore decided that the experiments should be repeated by a different method which would enable individual fission processes to be examined in detail. For this purpose, a track-recording technique, such as the expansion chamber or the photographic plate method, is eminently suitable. An expansion chamber has been successfully used by Bøggild, Brostrøm & Lauritsen (1940) in recording the tracks of fission fragments, but this method suffers from the disadvantage that the fragments originate in thin uranium foils in such a way that the point of fission, which is of special interest in an investigation of multiple fission processes, is obscured. The photographic plate method is free from this defect, since it is possible to record in an emulsion the tracks of pairs of fission fragments, including the point where fission occurs. This method was first used by Lark-Horowitz & Miller (1941). Attempts made by Kinsey and others to investigate rare fission phenomena by this method, using half-tone plates of the standard type, were unsuccessful because of the difficulty of distinguishing fission tracks from those of the natural α-particles emitted by the uranium from the time of its incorporation in the emulsion to the time of development, and protons, knocked on by neutrons during the irradiation of the plates. However, the introduction by Demers (1946*a*) and by Ilford Ltd. of concentrated emulsions (Powell, Occhialini, Livesey & Chilton 1946) has led to considerable improvements in the photographic plate technique, and these have rendered possible the production and identification of fission tracks in large numbers (Green & Livesey 1946).

This paper describes an investigation, by the photographic plate method, of those modes of fission which give rise to the emission of three charged particles. Some preliminary results were reported to the Physical Society Conference held at Cambridge in July 1946. A brief account of the work has been published already (Green & Livesey 1947) and similar observations have been described by Demers (1946*b*) and by Tsien, Chastel, Ho & Vigneron (1946). A detailed comparison of all the experimental results available at present is included in the last section of this paper.

2. The production and identification of fission tracks in photographic emulsions

In order to record the tracks of fission fragments in the Ilford Nuclear Research plates, the emulsions were first impregnated with uranium by immersion in a solution of uranyl acetate in dilute acetic acid. The period of immersion depended on the thickness of the emulsions used, and varied between 1 hr. for 20 μ layers, and 12 hr. for 100 μ layers. This treatment produced a uniform distribution of the uranium salt throughout the thickness of the emulsion. The plates were dried in a current of air, then enclosed in light-tight boxes and irradiated with slow neutrons. The neutrons were produced by bombardment of a lithium hydroxide target with 4·5 MeV protons accelerated in the Cavendish Laboratory cyclotron. This produced neutrons with a maximum energy of 3 MeV, and these were slowed down by surrounding the cyclotron target chamber with paraffin blocks to a thickness of approximately 30 cm. The plates were placed inside the stack of paraffin blocks at a distance of 15 cm. from the target, and, with a beam current of 50 μA, irradiations lasting 1 hr. were carried out. At the end of this period the plates were processed.

In developing the plates, it was necessary to allow for the action of the acetic acid present in the emulsions in retarding the development process. This effect is similar to that caused by under-development, and it was found to be most pronounced in the deeper layers of the 100 μ emulsions. In order that these layers should be developed sufficiently, it was necessary to develop the plates for a considerable period in very dilute reagents. It is possible to remove the acetic acid before development by washing the plates, but this method gave less satisfactory results. The best results were obtained by developing the plates, immediately after exposure, for 40 to 50 min. in a solution containing I.D. 19 X-ray developer diluted to the extent of 1 part in 10 parts of water. The plates were then immersed for 5 min. in a potassium metabisulphite stop-bath, fixed for 2 hr. in a 30 % hypo solution, and were finally washed and dried.

During the period between the impregnation and the processing of the plates, appreciable numbers of α-particles were necessarily emitted by the uranium. In addition, the sensitive layers were traversed by protons scattered by fast neutrons, which are always present when slow neutrons are produced by the use of paraffin wax, and by protons emitted in the disintegration by slow neutrons of nitrogen present in the gelatin of the emulsion. The plates were also affected by γ-rays arising from various sources; these were of sufficient intensity to cause some general fogging. In order that fission tracks may be observed in the presence of such an intense background, it is necessary to employ emulsions which, while having the normal sensitivity to particles of high specific ionization, have a relatively lower sensitivity to particles of low specific ionization. In such emulsions the tracks of α-particles and fission fragments may be of normal strength while proton tracks are not recorded.

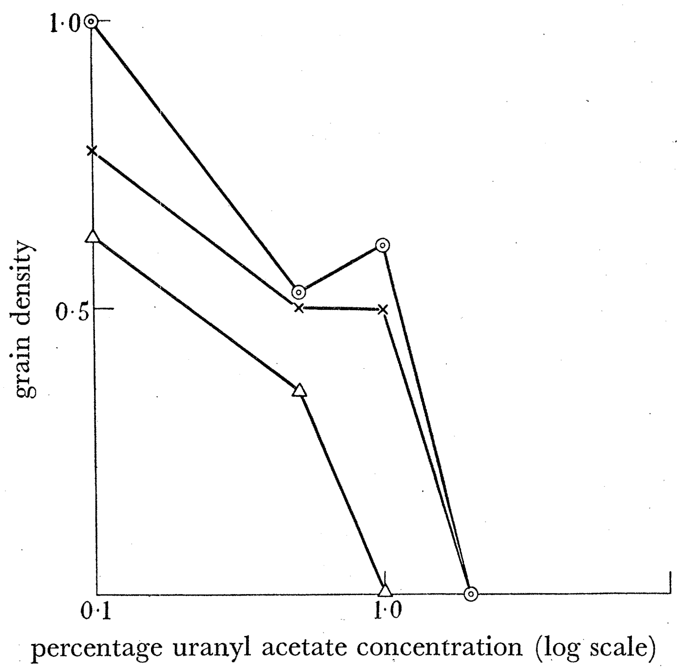


FIGURE 1. Desensitization by uranyl acetate.

△ 30 min. development. × 50 min. development. ⊙ 70 min. development.

Emulsions possessing the property of particle discrimination were prepared by applying the technique of desensitization, which has been described elsewhere (Powell *et al.* 1946). Preliminary tests were carried out with chromic acid as the desensitizing agent, but it was found that the uranyl salts used in impregnation of the plates accomplished the same purpose. This was shown by the results of a series of measurements on the average grain density of fast proton tracks produced in Ilford B1 emulsions which had previously been treated with solutions containing different amounts of uranyl acetate dissolved in 10 % acetic acid. The results are represented in figure 1, in which the average grain density of 2 MeV proton tracks is plotted as a function of the concentration of uranyl acetate for different periods of development. It will be seen that the proton tracks are inhibited in B1 plates treated with 2 % uranyl acetate solution, and that this inhibition limit does not depend markedly on the development time. At the same stage of desensitization the α-particle tracks produced by disintegration of uranium are still clearly visible. The tracks of the α-particles are not appreciably diminished in intensity until the concentration of uranyl acetate reaches 4 %.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

The discriminating property of desensitized emulsions may be illustrated by an experiment in which a series of B 1 plates was immersed in 1, 2 and 4 % solutions of uranyl acetate and was irradiated with slow neutrons under the same conditions. After development, the 1 % plates were found to contain α-particle tracks, the ranges of which corresponded to the energies of α-rays emitted by the isotopes U^{234} and U^{238}, and also a few weak proton tracks. The background of single developed grains was high, but it was much less than that produced in untreated B 1 plates. Some very dense tracks were observed. The mean range of these was 24·0 μ, that is, considerably greater than the maximum range of α-particles emitted by uranium. These longer tracks were caused by pairs of fission fragments. Figure 17, plate 11, shows a uranium α-particle track and a double-fission fragment track recorded in a 1 % plate and photographed in the same field of view of the microscope. It will be seen that there is an appreciable difference in grain density between the two tracks, and this feature, together with the longer range of the fission tracks, enables the observer immediately to identify the two types of track.

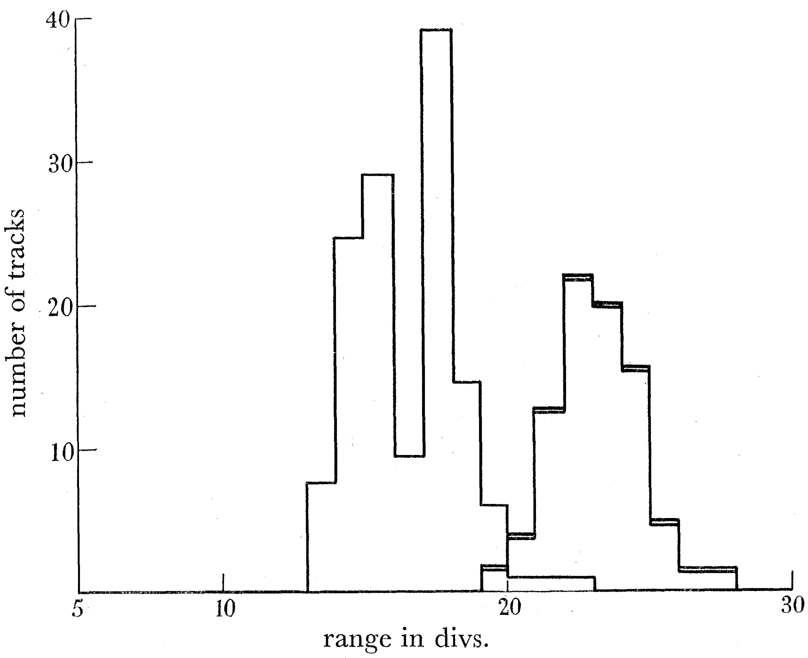


FIGURE 2. Range distribution of tracks in 2 % plate.

1 div. = 1·03 μ.
— α-particle group.
= fission fragment group.

The 2 % plates, representing a further stage in the desensitization of the B 1 emulsion, were found to contain fission and α-particle tracks only, the proton tracks being entirely inhibited. Figure 2 shows the distribution in range of tracks observed in one of these plates, the two histograms referring to dense (fission) tracks and light (α-particle) tracks respectively. The two groups were almost completely resolved, and partial resolution of the α-particle groups produced by the isotopes U^{234} and U^{238} was obtained. The mean range of the fission tracks in these plates was 23·5 μ.

The 4 % plates recorded α-particle tracks only very weakly, but the grain density of the fission tracks was not much less than that in the 2 % plates. Figure 18, plate 11, shows a fission track and two α-particle tracks photographed in the same field of view. The range distribution of the tracks is reproduced in figure 3, where it is seen that the α-particle group, though poorly defined, is still separate from the fission group. The mean range of fission tracks in the 4 % plates was found to be 23·0 μ.

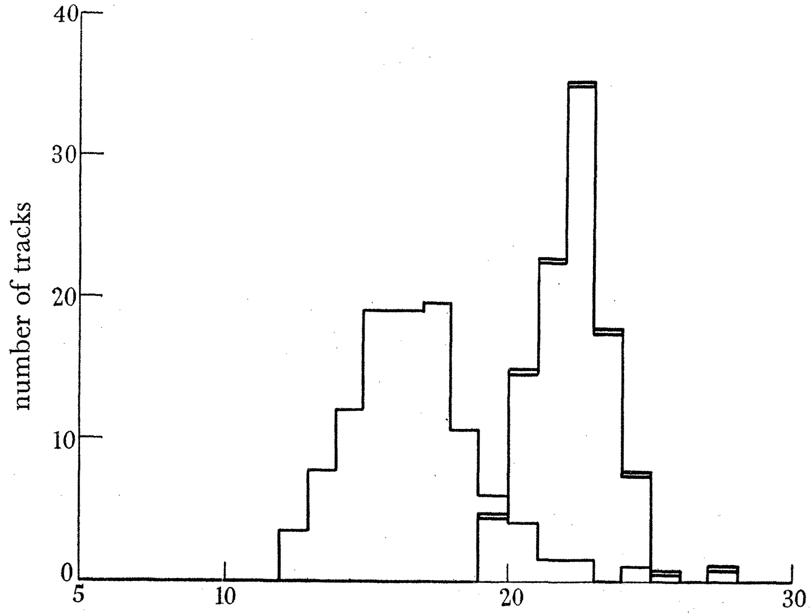


FIGURE 3. Range distribution of tracks in 4 % plate.
1 div. = 1·03μ.
– α-particle group.
= fission fragment group.

These results show that it is possible to obtain B 1 plates varying widely in sensitivity and discriminating power by employing different concentrations of uranyl acetate in the impregnating solution. Further tests were carried out on other types of emulsion, and fission tracks were produced and identified in C 1, C 2 and D 1 plates. The C 2 emulsion was found to be generally similar in properties to the B 1 emulsion, while the C 1 and D 1 types were progressively less sensitive. C 1 plates treated with 1 % uranyl acetate solution did not record proton tracks, and D 1 emulsions impregnated with uranium did not detect either protons or α-particles, although fission tracks were satisfactorily recorded.

It was shown, in a separate experiment, in which plates surrounded by cadmium shields were irradiated with neutrons from the cyclotron source and paraffin moderator, that approximately 99 % of the fission tracks occurring in the plates were produced by neutrons in the cadmium absorption band. Since the isotope U^{238} is not known to be fissionable by slow neutrons, whereas fission of the isotope U^{235} is induced by neutrons of all energies, it is evident that less than 1 % of the fission tracks observed were due to the isotope U^{238}.

Observations of the details of the fission tracks were carried out with a microscope magnification of $\times$ 1500, obtained by using a 2 mm. oil-immersion objective in conjunction with eyepieces of power $\times$ 15. The general features of the tracks resemble those shown in cloudchamber photographs published by Bøggild *et al.* (1940). Many collisions occur between the moving fragments and the nuclei composing the material of the emulsion, and forked tracks, showing the nuclei recoiling after impact, are frequently observed. An example of a fission track with several branch tracks is shown in figure 19, plate 11. The recoil tracks were made the subject of a separate investigation, which is described in the next section.

The point of fission cannot be located exactly in the tracks produced in these plates because the specific ionization is not highly dependent on the mass of the ion at the beginning of its path, as may be seen by an inspection of the range-velocity relations published by Bohr (1941). Nevertheless, the point of origin of the fragments producing a track may be located within a limited distance if the data of Bøggild *et al.* are employed. According to these authors, the mean ranges of the two principal groups of fission fragments in air are 25 and 19 mm. respectively. Assuming the stopping power of the emulsions relative to air to be the same for both groups of fragments, the point of fission must occur, in practically all cases, in the central 5μ of each double track as observed in the plates.

The mean range of fission tracks in B 1 and C 2 emulsions, desensitized by 0·5 % uranyl acetate solutions, is $24{\cdot}0\mu$. The mean total range of fission fragments in air is 44 mm., hence the stopping power of these emulsions for heavy ions is 1840 approximately. In strongly desensitized plates, the mean range is reduced, and it is $23{\cdot}0\mu$ in an emulsion desensitized with a 4 % uranyl acetate solution. This effect may be explained on the basis of the range-velocity relation for heavy ions derived by Bohr (1941); this shows that the specific ionization decreases with decreasing velocity over the greater part of the trajectory before rising to a sharp maximum at very low velocities. This maximum, however, is concentrated within a very short distance, of the order of the mean diameter of the grains in a photographic plate, and its effect is therefore not normally evident in the Ilford plates. The general decrease of specific ionization with decreasing velocity does, on the other hand, account for the fact that, in strongly desensitized plates, the latter parts of the fragment tracks are not recorded, since the density of ionization is insufficient to render the grains developable. The observed mean range under these conditions is, therefore, less than the true mean range of the fragments.

3. The analysis of recoil tracks produced by nuclear collisions

The occurrence of a large number of nuclear recoil tracks along the path of a fission fragment is a consequence of the high charge and high mass (*vide* Bohr 1940) of the fragment. Such recoil tracks occurring in the central parts of double-fission tracks may be confused with the tracks of particles emitted at the instant of fission, and it is therefore necessary to make an analysis of the recoil tracks in order that they may be distinguished, on the basis either of their total abundance or of the deflexion of the fission fragment track.

The importance of multiple nuclear recoils in contributing to the stopping of fission fragments, especially at low velocities, has been emphasized by Bohr (1941). In the case of an individual collision, the principles of conservation of energy and momentum establish the following relation between the recoil energy (ϵ) and the fragment energy (E):

$$\epsilon = \frac{4Mm}{(M+m)^2} E \cos^2\theta,$$

where θ = angle between the original fragment direction and the direction of the recoil nucleus (see figure 4), M = mass of fission fragment, and m = mass of recoil nucleus.

Under the normal conditions of microscope observation, it is necessary to impose a lower limit on all range measurements, so that tracks of range shorter than the arbitrary limit are rejected. In this experiment, the limit was fixed at $1\cdot7\,\mu$, corresponding to a range of 3 mm. in air for α-particles and heavier particles. (In converting ranges in the Ilford concentrated emulsions to the corresponding ranges in air, the stopping-power data of Lattes, Fowler & Cüer (1947) are employed.) This restriction leads to the imposition of a lower limit on ϵ, for a given value of m, and an upper limit ($\theta_{max.}$) on θ when m and E are specified. The cross-section (σ) for the production of recoil tracks of range greater than $1\cdot7\,\mu$ can then be calculated, if the charge numbers (z, z') of the fission fragment and the recoil nucleus are known; the Rutherford scattering formula yields the relation

$$\sigma = \pi P^2_{max.},$$

$$P_{max.} = \frac{M+m}{m}\frac{zz'e^2}{2E}\tan\theta_{max.},$$

where σ = total collision cross-section and $P_{max.}$ = maximum impact parameter corresponding to recoil angle $\theta_{max.}$. The cross-section calculated from these formulae is in fact greater than the true cross-section because no allowance has been made for the screening of the nuclear charges by extranuclear electrons. Nevertheless, it may be used to estimate an upper limit for the probability of occurrence of nuclear recoil tracks longer than $1\cdot7\,\mu$.

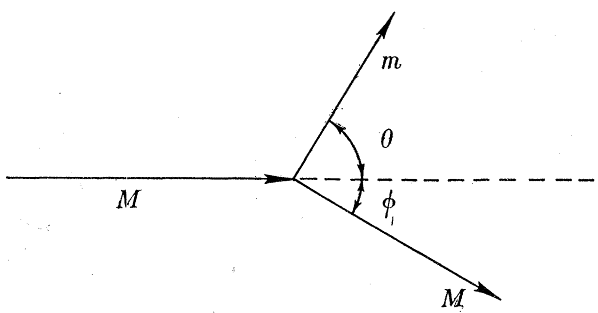


FIGURE 4. Recoil process diagram.

In the case of fission fragments traversing a photographic emulsion, the nuclear recoils may be divided into three groups, according to the mass of the nucleus struck; these groups are:

(i) Protons ($m = 1$).
(ii) Light recoils: carbon ($m = 12$), nitrogen ($m = 14$), oxygen ($m = 16$).
(iii) Heavy recoils: bromine ($m = 80$), silver ($m = 108$).

The proton recoils may be neglected, since they do not contribute appreciably to the stopping of the fragments and since proton tracks were suppressed in the emulsions used in this experiment.

In the majority of cases it is possible to distinguish the tracks of light recoils from those of heavy recoils by measuring the angle of deflexion (ϕ) of the main fission track at the point of collision. For the light nuclei, m is always less than M, so that there is a maximum angle of deflexion ($\phi_{max.}$) determined by the relation

$$\phi_{max.} = \sin^{-1}\left(\frac{m}{M}\right).$$

Taking the minimum value of M to be 80, the following values of $\phi_{max.}$ are obtained:

$$\text{carbon} \quad \phi_{max.} = 9^\circ, \qquad \text{nitrogen} \quad \phi_{max.} = 10^\circ, \qquad \text{oxygen} \quad \phi_{max.} = 12^\circ.$$

On the other hand, for heavy recoils there is a minimum angle of deflexion ($\phi_{min.}$) imposed by the condition that the recoil nucleus should have a range greater than $1{\cdot}7\,\mu$. The values of $\phi_{min.}$ for silver and bromine may be calculated from the range-velocity relation of Bohr (1941); assuming the maximum value of the fission fragment energy to be 100 MeV, the following figures are obtained:

$$\text{bromine} \quad \phi_{min.} = 8^\circ, \qquad \text{silver} \quad \phi_{min.} = 9^\circ.$$

It appears, therefore, that if ϕ exceeds 12° in any collision, the recoil nucleus is heavy, while if ϕ is less than 8° it is light. Light and heavy recoils may be differentiated in this way, and in practice ambiguity arises in less than 20 % of the total number of recoils observed.

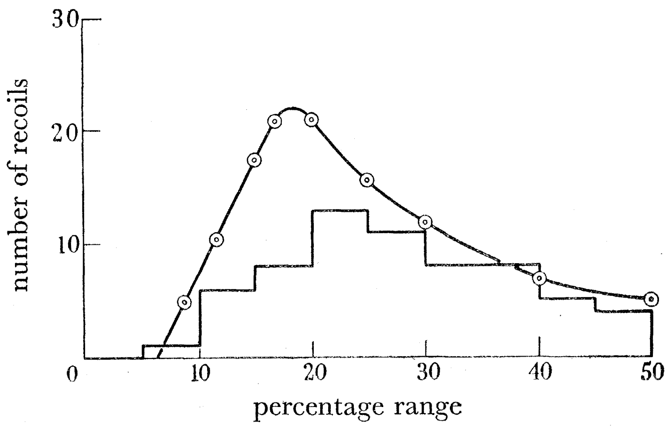


FIGURE 5*a*. ⊙ Calculated abundance of light recoils. — Histogram: observed abundance of light recoils.

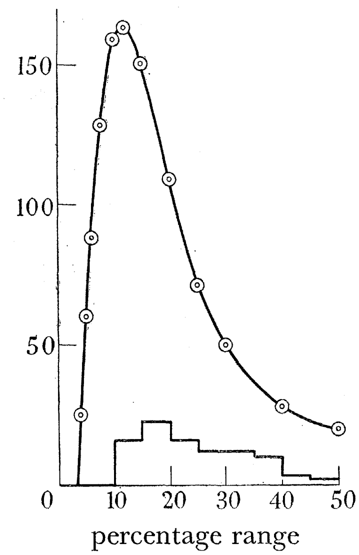


FIGURE 5*b*. ⊙ Calculated abundance of heavy recoils. — Histogram: observed abundance of heavy recoils.

An experiment was carried out to determine the distribution of the abundance of light and heavy recoils along the length of the fission tracks. 2400 fission tracks, lying in the focal plane of the microscope objective, were examined in plates which were fully sensitive to light recoils, but only partially sensitive to α-particles. This precaution was taken to avoid confusion between recoil tracks and the tracks of light particles, of mass similar to that of an α-particle, originating at the point of fission. The results of the observations are reproduced in histogram form in figure 5, where the number of recoil tracks is plotted as a function of the distance between the point of collision and the nearer end of the fission track, expressed as a percentage of the total range. In preparing this diagram, those recoil tracks which could not definitely be allocated to either the light or the heavy group (vide supra) were divided among the two groups in the ratio of the estimated abundance.

Figure 5 also shows curves representing the theoretical estimates of the abundance of recoil tracks, based on the Rutherford formula and on the known composition of the emulsion material, namely, 3·4 g./c.c. of silver bromide and 0·54 g./c.c. of gelatin. In calculating these curves, the range-velocity relations for light recoils were taken from the data of Knipp & Teller (1941), and the following figures were adopted for the mean mass, charge, and initial energy of the fission fragments:

$$\overline{M} = 120, \quad \overline{z} = 45, \quad \overline{E} = 80\,\text{MeV}.$$

In general, the theoretical curves lie above the experimental values, in accordance with the fact that the theory does not take into account screening effects. The existence of maxima in the graphs is due to the lower limit of acceptance of particle ranges; as is well known, if this limit is reduced to zero, the formulae for the collision cross-sections become divergent.

The total abundance of recoils of the two types was also determined. In 2400 fission tracks observed, 83 heavy recoils were found, corresponding to an abundance of 1 in (29 ± 3) tracks. Forty-three light recoils were found, corresponding to an abundance of 1 in (56 ± 5) tracks. Similar observations were carried out in a plate which was fully sensitive to α-particles and all heavier charged particles, but, in this investigation, the central $5\,\mu$ of each track were excluded, in order to eliminate any light particles emitted in the fission process. 620 fission tracks were examined, and 21 heavy recoils were found, that is, 1 in (30 ± 6) tracks. Thirteen light recoils were found, corresponding to an abundance of 1 in (48 ± 12) tracks. The agreement between these figures and those obtained in the more extensive investigations shows that the observers did not omit appreciable numbers of light recoils in the less sensitive plates.

4. On the fission of uranium into three charged particles

A. *Emission of light nuclei*

Cassels *et al.* (1947) reported that in approximately 4 % of fission events, a short-range secondary charged particle was emitted, and that this was probably an α-particle. Such particles should be recorded in photographic plates as short tracks originating near the centres of the double fission fragment tracks. It is not possible to observe the particles in B1 or C2 emulsions of normal sensitivity because of the heavy background caused by the irradiation of the plates with neutrons. Although this background may be largely eliminated by means of the desensitization technique, this process necessarily involves the inhibition of some, at least, of the proton tracks present, and therefore of protons which may be emitted in the fission process. However, the results of the counting experiments indicated that the particles were probably α-particles, and the tracks of these are not inhibited by moderate desensitization. In view of these facts, it was decided to search for particles, heavier than protons, emitted in the fission process. Accordingly, the investigation was carried out in plates which were fully sensitive to α-particles and only partially sensitive to protons; these conditions were realized in C2 plates impregnated with 0·5 and 1 % uranyl acetate solutions.

The preliminary work was carried out using plates with emulsion coatings $20\,\mu$ thick. One of the observations made with these plates was that, in a small proportion of fission events, very long tracks appeared to originate near the point of fission. The range distribution of these high-energy particles could not be measured accurately because most of them escaped from the sensitive layer. The abundance was estimated to be approximately one in 300 fission events, and the maximum range observed was $230\,\mu$, equivalent to 43 cm. of air.

These particles were certainly not the same as those investigated by Cassels *et al.*, whose coincidence-counting arrangement was not sufficiently sensitive to detect long-range particles of such low abundance.

Because of the low abundance and considerable range of these particles, it was not possible to obtain comprehensive data about them by the methods employed in investigating the short-range particles. The experimental work was therefore divided into two parts, with an arbitrary division determined by the particle range. This division was fixed at 5μ, equivalent to 8 mm. of air. Particles of range shorter than this were studied in plates with 20μ coatings, and the long-range particles were investigated separately in 100μ plates.

Long-range particles

In order to investigate the phenomenon of the emission of long-range light particles in fission, fission tracks were prepared in C 2 emulsions 100μ thick. The plates, which contained approximately 40 tracks/sq.mm., were examined systematically by two observers, working independently and using a microscope system with an overall magnification of $\times 1000$. The observers recorded details of the range of each long-range particle detected, the angle between its track and the tracks of the fission fragments, and the distances of the point of emission from the two ends of the main track. A typical example of a long-range particle track is shown in figure 20, plate 11.

The total number of fission tracks counted, in four plates of similar sensitivity, was 20,000, and 58 cases of long-range particle emission were observed. The abundance of particles with ranges exceeding 5μ is therefore 1 in (340 ± 40) fission events. No definite correlation between the range of the particle and the angle of emission was established, and the grain density of the particle tracks was approximately equal to that of α-particles of the same range. A rough estimate of the mass of the light particles may be obtained by measuring the small angle of deflexion of the fission track at the point of emission and by applying relations based on the principle of conservation of momentum. In such calculations it is necessary to assume the stopping power of the emulsion for fission fragments in order to derive the particle velocities from their ranges, and no allowance can be made for the momentum of secondary neutrons emitted in fission. The estimated mass depends sensitively on the magnitude of the angle of deflexion, which is usually less than 5°, and which is difficult to measure accurately because of the pronounced small-angle nuclear scattering of the fission tracks. For these reasons the method is not capable of high accuracy. Table 1 includes the measured angles of deflexion in seven typical cases and the calculated mass numbers. The mean value of the mass number is 3·6, and, considering the errors in the individual determinations, it is considered that all the results are consistent with the assumption of a single mass number of 4.

TABLE 1. ESTIMATED MASS NUMBERS OF LONG-RANGE LIGHT PARTICLES

range of light particle in μ	angle of emission (°)	angle of deflexion (°)	velocity light particle $\times 10^{-8}$ (cm./sec.)	velocity fragment $\times 10^{-8}$ (cm./sec.)	estimated mass
228	63	4	34	12	3·8
32	78	5	18	10	6·9
150	88	7	30	9	5·1
163	67	1	28	9·5	1·0
146	73	4	29	9·5	3·4
62	57	2	22	9·5	2·5
120	80	3	27	9·5	2·6

The fraction of particles of a given range ending outside the sensitive layer is a function of the range; thus the observed range distribution differs from the true distribution. The true distribution is obtained by applying an escape correction to the experimental data. It is proved in the Appendix that, provided that the fission tracks are randomly oriented and are uniformly distributed throughout the thickness of the emulsion, the fraction (f) of tracks of range (R) ending inside an emulsion of thickness $2d$ is

$$f = 1 - \frac{R}{4d} \quad \text{if} \quad R < 2d,$$

and

$$f = \frac{d}{R} \quad \text{if} \quad R > 2d.$$

The reliability of these correction formulae was checked by comparing the observed number of long-range particles crossing the boundary planes of the emulsion with the number calculated from the above formulae. The experimental figure was 25 tracks out of a total of 58 long-range particles detected, and the calculated figure was 29, which is in sufficiently close agreement with the first figure. The range distribution of long-range particles, corrected for escape, is shown in figure 6. There is a continuous distribution between 5 and 180 μ, and the maximum range measured was 296 μ, equivalent to 55 cm. of air.

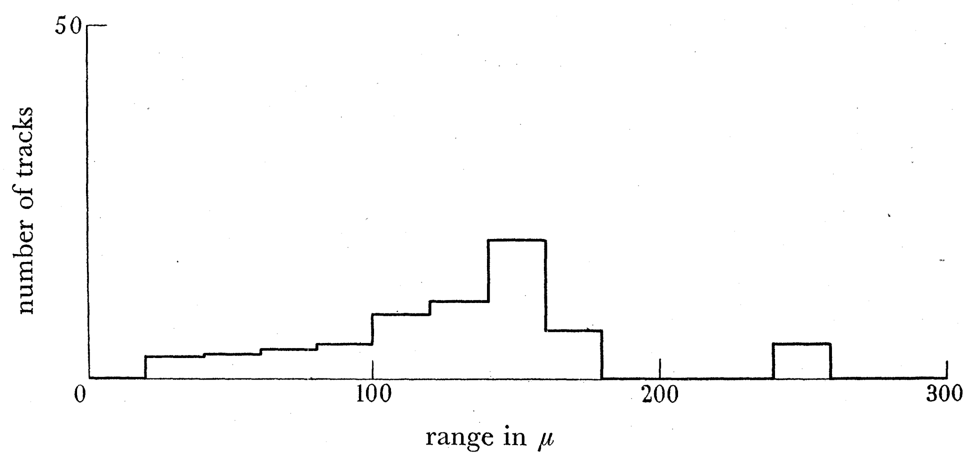


FIGURE 6. Range distribution of long-range light particles.

If it is assumed that the light particle is emitted at the instant of fission in each case, its track accurately defines the point of fission, and it is therefore possible to measure the range of the individual fragments. The range distribution of the individual fragment tracks is shown in figure 7, which expresses the results of observations on 24 fission events in which the angles of dip of the fission tracks were less than 10°. Two distinct groups are present. This indicates that the mass of the compound nucleus is divided asymmetrically in this mode of fission, and the group of longer range may be identified with the group of lighter fragments. The mean range of the lighter fragments is 12·5 μ, and that of the heavier fragments in 9·8 μ. These figures may be used in conjunction with the range-velocity data of Bohr (1941) to calculate the average mass for each group, applying the principle of conservation of momentum to the process. In these calculations, the momenta of the light charged particles and of secondary neutrons are neglected. If M_1, M_2 are the average masses for the two fragment groups, we have

$$M_1 + M_2 = 236, \quad M_1 V_1 = M_2 V_2,$$

where $$V_1 = 12{\cdot}9 \times 10^8 \text{ cm./sec.}, \quad V_2 = 9{\cdot}9 \times 10^8 \text{ cm./sec.},$$

whence $$M_1 = 96 \pm 10, \quad M_2 = 140 \pm 10.$$

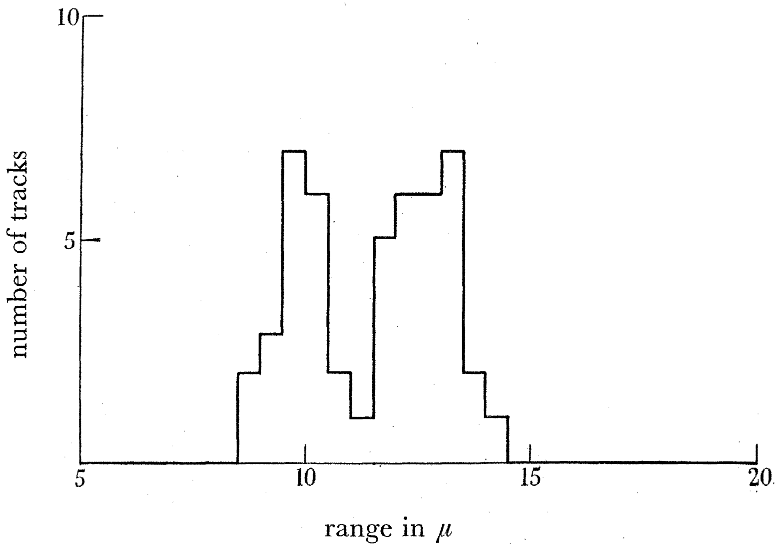


FIGURE 7. Range distribution of individual fission fragments for events in which a long-range light particle is emitted.

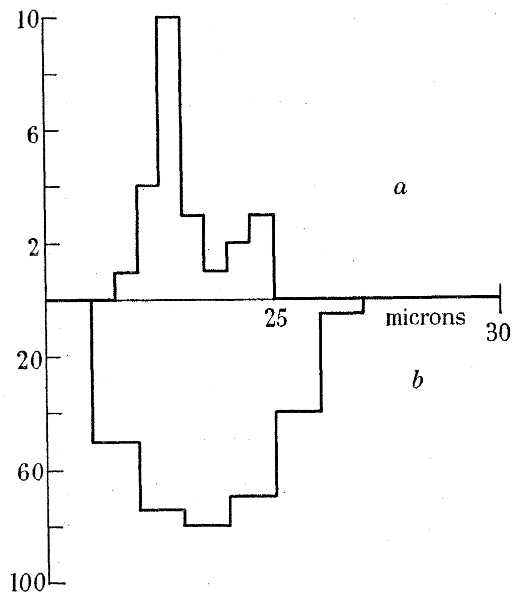


FIGURE 8. Combined range of fission fragments for events in which a long-range light particle is emitted. *a*, fission fragments associated with long-range particles; *b*, all fission fragments.

The distribution of the total range of the fission tracks associated with long-range particle tracks is shown in figure 8, together with the range distribution of all types of fission tracks. The mean range of all types of fission tracks in these plates is $24{\cdot}0\,\mu$ [this figure exceeds the sum of the mean ranges of the individual fragments ($9{\cdot}8$ and $12{\cdot}5\,\mu$) because the number of tracks in the two groups are not equal, owing to the inclusion in the longer group of a small number of cases of symmetrical division, whereas that of 24 tracks, associated with the emission of light particles of range exceeding $5\,\mu$, is $23{\cdot}2\,\mu$]. If the selection is further restricted to cases in which the range of the emitted particle exceeds $50\,\mu$, the mean range of 13 fission tracks is $22{\cdot}9\,\mu$. This evidence indicates that there is a small diminution in the fission fragment ranges when a long-range particle is emitted. It is probable, therefore, that the total energy release in the modes of fission here investigated is not appreciably greater than the average energy released in binary fission.

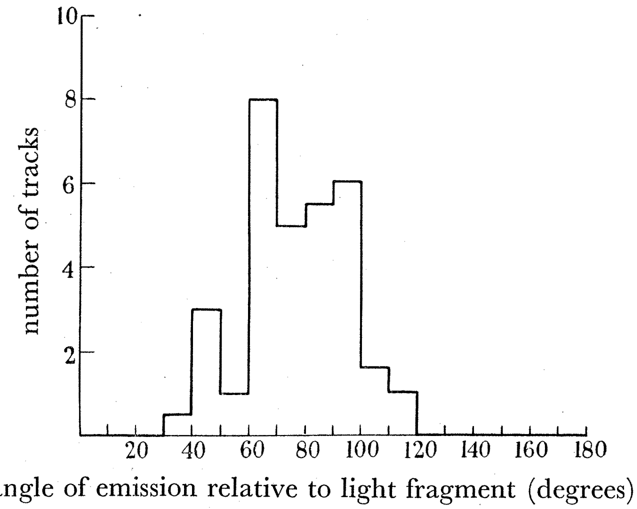


FIGURE 9. Angular distribution of long-range light particles.

The angular distribution of emission of the long-range particles was obtained by plotting the number of tracks as a function of the angle between the particle track and the longer of the two fragment tracks, that is, the track of the lighter fragment. Figure 9 represents the results of observations on 31 tracks; it shows that the particles are emitted in directions nearly perpendicular to the initial line of separation of the fission fragments, and that the most probable direction of emission makes an acute angle with the direction of motion of the lighter fragment. The mean angle of emission, relative to this direction, is approximately 77°.

Short-range particles

In searching for the tracks of short-range light particles emitted in the fission process, a difficulty is encountered which was not present in the investigation of long-range particles. The tracks of nuclear recoils are frequently longer than the lower limit of detection ($1{\cdot}7\,\mu$), and these may be confused with the tracks of particles emitted at the instant of fission.

The tracks of heavy nuclei are readily distinguished and eliminated by observations on the angle of deflexion of the main fission track at the point of collision; the minimum angle of deflexion caused by a heavy recoil is 8°, whereas the maximum due to the projection of short-range α-particles is less than 5°. However, the light recoils cannot be identified in this way, though their abundance may be deduced from the experiments described in § 3. These experiments show that the abundance of light recoils of all types located in those parts of the fission track which contain the point of fission, that is, in the central $5\,\mu$ of the track, is 9 recoils in 2420 fission events, or 1 in (270 ± 60) fission events. This is in agreement with the theoretical estimate based on the Rutherford formula. Unless, therefore, the abundance of short-range light particles emitted in fission is appreciably greater than this figure, reliable information concerning the emission of such particles cannot be obtained by the photographic plate method. On the other hand, the method is clearly applicable if the results of Cassels *et al.* regarding the abundance of short-range light particles are approximately correct.

Experimental results on the occurrence of short tracks originating in the central $5\,\mu$ of the double-fission tracks were obtained by two observers, who examined a large number of fission tracks with a microscope giving an overall magnification of $\times 1500$. In order to obtain accurate determinations of the angles of deflexion of the main fission tracks, the investigation was restricted to those fission tracks which lay entirely within the focal plane of the microscope objective. (The depth of focus was approximately $0{\cdot}5\,\mu$.) The $20\,\mu$ plates used were fully sensitive to α-particles and partially sensitive to protons; they contained approximately 25 fission tracks/sq.mm. lying in the focal plane. Fission tracks with subsidiary tracks originating near the point of fission were recorded and the number satisfying the following conditions was determined:

(i) the point of emission of the short-range particle lay within the central $5\,\mu$ of the fission track;

(ii) the angle of deflexion of the fragment tracks was less than 5°;

(iii) the range of the secondary track was between $1{\cdot}7$ and $5{\cdot}0\,\mu$.

It should be noted that the condition (ii) is effective in eliminating some proportion of the light recoil tracks, the total abundance of which, satisfying conditions (i) and (iii) is 1 in (270 ± 60) fission events, as previously quoted.

Out of 5000 fission tracks examined, the number with subsidiary tracks satisfying the above three conditions was 48, so that the abundance was 1 in (104 ± 10) fission events. This figure is significantly higher than the abundance of light recoils. Moreover, some of the short tracks observed had ranges of the order of $5\,\mu$, and were very nearly perpendicular to the directions of the fragments. Calculations showed that these could not have been produced by light recoils. A typical example of such a track is shown in figure 21, plate 11. This evidence, taken in conjunction with the positive result of the coincidence-counting experiments, establishes the existence of short-range light particles emitted in fission.

The abundance of short-range particles quoted above is subject to errors from two sources. First, it is probable that a number of light recoils was included in the count: since the abundance of light recoils of the correct type is less than 1 in (270 ± 60) events, the error from this cause is unlikely to have been greater than 30 %. Secondly, there is an error in the opposite sense caused by the difficulty of detecting short tracks emitted from the fission track in the direction of viewing. Such tracks are concealed by the grains composing the fission track. This error has not been estimated, but it should not exceed that due to the inclusion of light recoils. Taking these factors into account, the abundance of short-range particles is quoted as 1 in (100 ± 30) fission events.

The distribution in range of the short-range particles is shown in figure 10. The average range of these particles is $2{\cdot}8\,\mu$, equivalent to 5 mm. of air. Again there is no obvious correlation between the ranges of the particles and the angles of emission. On the assumption that the point of emission in each case defines the point of fission, the range distribution of individual fragments was deduced as before, and is shown in figure 11. In this graph two groups of fragments are not resolved, but this may be due in part to the inclusion of a small number of recoil tracks in the group investigated. The angular distribution of emission of the particles is shown in figure 12, in which the number of tracks is plotted as a function of

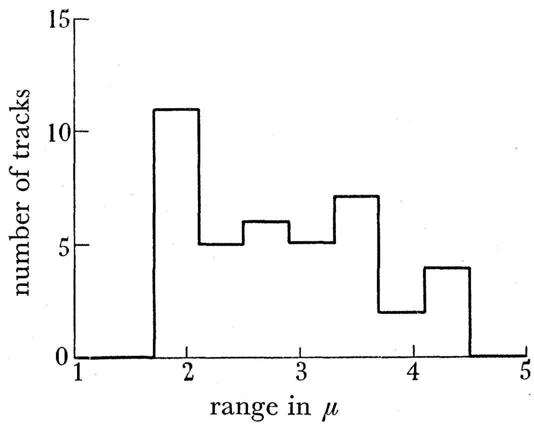


FIGURE 10. Range distribution of short-range particles.

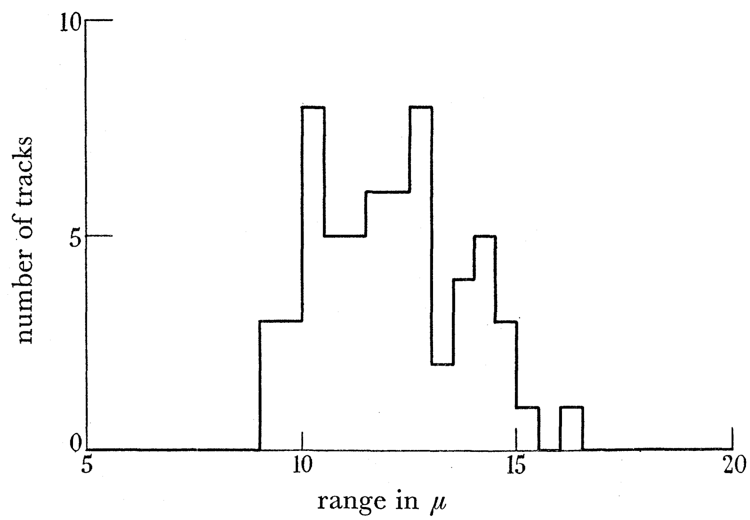


FIGURE 11. Range distribution of individual fission fragment ranges for events in which short-range particles are emitted.

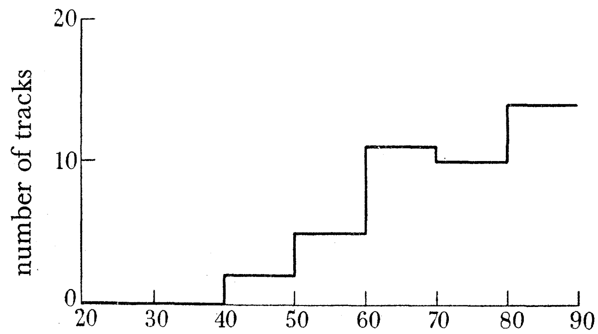


FIGURE 12. Angular distribution of short-range particles.

the angle between the particle track and the initial directions of the fragments. The results show once more that there is a preferential emission of particles in directions nearly perpendicular to those of the fragments.

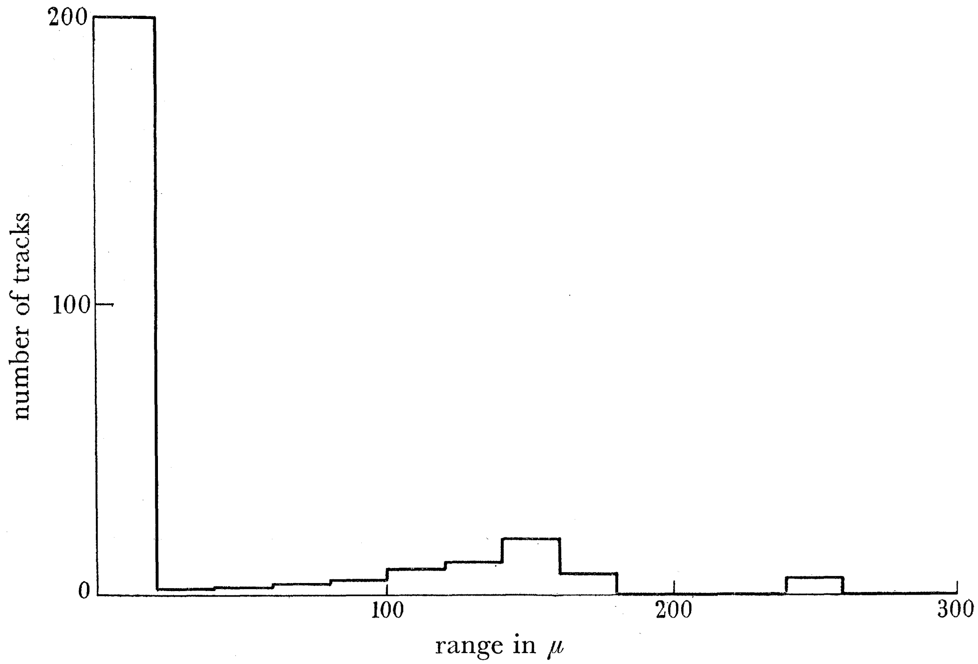


FIGURE 13. Combined range distribution of light particles.

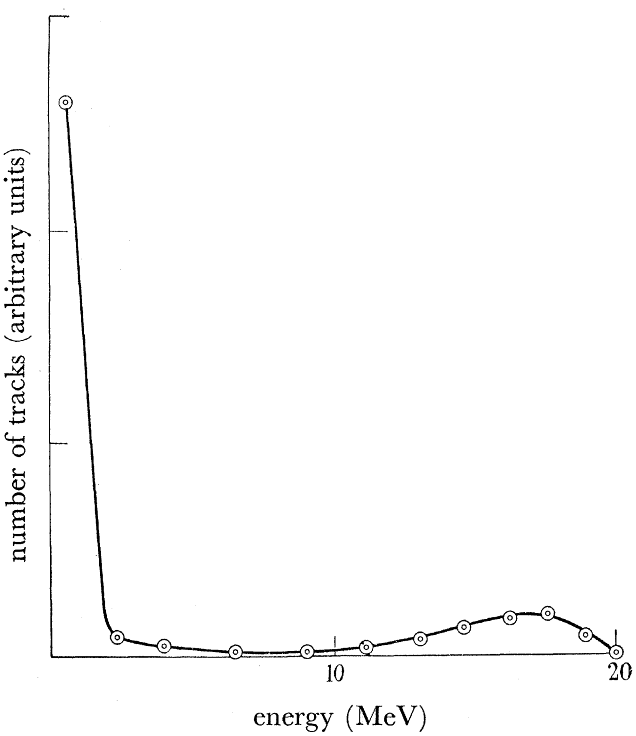


FIGURE 14. Energy distribution of light particles.

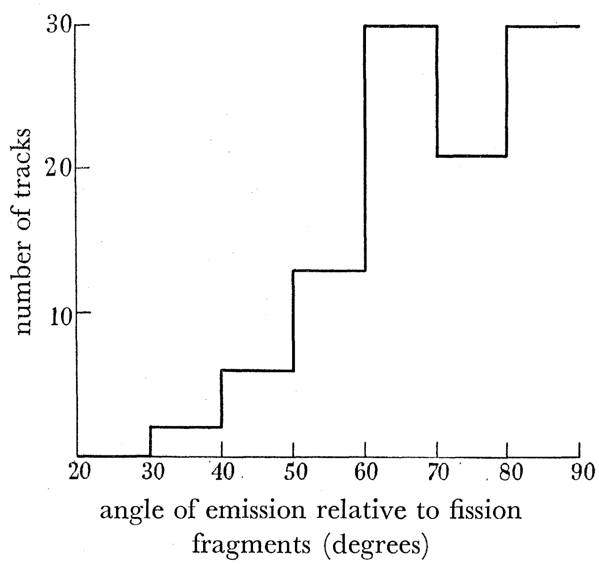


FIGURE 15. Combined angular distribution of light particles.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Combined results

The distinction, hitherto drawn, between long-range and short-range particles emitted in fission is entirely arbitrary. The combined range distribution of all the particles observed is shown in figure 13. If it is assumed that all the light nuclei emitted are α-particles, the energy distribution may be deduced and this is given in figure 14. It appears that there is a continuous distribution of energies between a minimum corresponding to the lower limit of detection, namely, $1{\cdot}7\,\mu$, and a maximum in excess of 20 MeV. There is some evidence for a broad peak in the distribution near 17 MeV, in addition to the peak caused by short-range particles.

The total abundance of light nuclei emitted in fission under these conditions is 1 in (80 ± 30) fission events. The combined angular distribution is shown in figure 15, in which the number of tracks is plotted as a function of the angle between the direction of the light particle and that of the fission fragments.

B. *The division of uranium into three heavy fragments*

The modes of ternary fission discussed in the previous sections involve the emission of a light nucleus in addition to the two heavy fragments. This process is roughly analogous to the emission of secondary neutrons, assuming that these neutrons are emitted instantaneously. It is possible that fission of the compound nucleus may also give rise to three fragments of comparable mass, as has been suggested by Present & Knipp (1940).

If such a mode of ternary fission occurs, the triple tracks produced in photographic plates are in many cases entirely similar to those caused by heavy nuclear recoils, and the two types of track are not always distinguishable by the application of rules based on the principle of conservation of momentum. The plates used in the experimental investigation of recoil tracks (§ 3) were fully sensitive to light recoils, and would therefore have recorded tracks of heavy fragments produced in ternary fission. In this investigation, such tracks would have been included in the count of heavy recoils; the fact that the observed abundance of heavy recoils was fairly low (1 in 29 fission events) proves that fission into three heavy fragments is not of frequent occurrence.

An independent investigation of a large number of fission tracks was undertaken in a search for triple tracks which could be ascribed definitely to ternary fission as distinct from heavy recoils. No such track was found in 5000 fission events examined in a plate which was fully sensitive to heavy ions but insensitive to α-particles. The abundance of these tracks must therefore be very low. Since this experiment was completed, Tsien, Ho, Vigneron & Chastel (1947) have reported a single case of ternary fission into heavy fragments, but these authors have not determined the abundance of such events.

C. *Discussion*

Before discussion of the results of various experiments on the fission of uranium into three charged particles is possible, it is necessary to ascertain which of the isotopes of uranium gives rise to the phenomenon. It was shown in § 2 that the isotope U^{238} was responsible for less than 1 % of the total number of fission tracks observed in the photographic plates used. In these plates, long-range particles were observed with an abundance of 1 in (340 ± 40) fission events. This figure may be compared directly with the recently reported results of Farwell, Segrè & Wiegand (1947), who, using a foil of U^{235} irradiated with slow neutrons, detected long-range particles with an abundance of 1 in (250 ± 50) fission events. The agreement between the two sets of results can only be explained by attributing the emission of the long-range particles detected in the plates to the isotope U^{235}.

The experimental results concerning short-range particles may be compared with those obtained in the coincidence-counting experiments of Cassels *et al.* These authors estimated the abundance to be 1 in 25 fission events, but this was derived on the assumption that the particles were emitted isotropically. If the estimate is corrected, using the angular distribution shown in figure 12, the abundance is reduced to 1 in 90 fission events, in agreement with the photographic plate results. This confirms the previous deduction that the light particles detected in the photographic plates were caused by the isotope U^{235}, since the uranium samples used in the counting experiments were enriched in the U^{235} isotope. The range and specific ionization of the particles detected in the coincidence-counting experiments correspond to those of a 1 MeV α-particle, and this interpretation is also consistent with the observations described above.

An independent investigation of the fission process by the photographic plate method has been made by Demers (1946*b*). This author employed a multiple layer technique to obtain paired fission tracks with the point of fission located in a thin uranium layer. By this means he was able to detect the emission of the long-range light particles and to show that it occurs within 2×10^{-14} sec. of the instant of fission. He assumed that the particles observed were α-particles.

Finally, the emission of long-range particles has been studied by the photographic plate method by Tsien *et al.* (1946), who confirmed the principal results described in this paper. The range distribution and the abundance ratio deduced by those authors also agree with those described above. Tsien *et al.* have made estimates of the masses of the light particles, based on the measured angles of deflexion of the fission fragment tracks, as described in a previous section. They state that some of the particles have masses differing significantly from that of the α-particle, and they quote two cases of emission of particles with mass numbers equal to 9 ± 2. However, it is to be emphasized that the results of these calculations are subject to large errors, for several reasons previously specified. On this point the results of this paper do not agree with the deductions of Tsien *et al.*, since they indicate that in all cases the estimated mass is consistent with a single mass number of 4.

If the results of all the experiments are considered, the following facts concerning the emission of light nuclei in the fission of U^{235} by slow neutrons may be regarded as established:

(i) A light nucleus is emitted in approximately 1 % of fission events, and in one-quarter of these cases the range exceeds that of the fission fragments.

(ii) The range of the light nucleus takes all values up to a limit exceeding 55 cm. air equivalent.

(iii) The specific ionization of the particle is similar to that of the α-particle.

(iv) The average mass number is very nearly 4.

(v) The interval of time elapsing between the fission process and the emission of light nucleus is frequently less than 2×10^{-14} sec.

(vi) The angular distribution of emission directions has a maximum in a direction nearly perpendicular to that of the initial motion of the fission fragments. In cases where long-range particles are emitted, the most probable direction of emission makes an acute angle with the direction of projection of the lighter fission fragment.

It is clear that the latter effect can only be explained as the result of electrical forces operating between the three particles when the light particle is situated centrally between the other two particles. In other words, the emission of the light nucleus is effectively simultaneous with the division of the compound nucleus.

The phenomenon of light particle emission may be interpreted in two ways. First, the compound nucleus may split directly into three fragments, with the lightest fragment situated centrally, as suggested by Tsien (1947). In consequence of this hypothesis, Tsien postulates that the light particle does not have a unique mass, and that the mass number varies between 1 and 9, at least. It is necessary to assume that the probability of formation of the light nucleus decreases with increasing mass number, in order to explain the low abundance of events in which the compound nucleus splits into three heavy fragments. If this interpretation is correct, it is probably to be expected that some of the light nuclei emitted will have a large neutron excess, and will therefore be radioactive; for example, the isotopes H^3 and He^6 should be produced. A simple test of the theory would thus be provided by a search for radioactive effects associated with the long-range particles emitted.

An alternative interpretation of the phenomenon, put forward by Feather (1947), is based on the assumption that all the light nuclei emitted are α-particles. It is pointed out that the nuclei occurring in the fission fragment region ($z = 35$ to $z = 60$ approximately) are most unstable with respect to α-particle emission when $z = 40$ and $z = 60$ approximately. For example, it is known that the samarium isotope Sm^{148} emits α-particles of energy 2 MeV. If, therefore, one of the primary fragments formed in fission has a nuclear charge close to 40 or 60, it may immediately emit an α-particle, provided that the potential barrier is sufficiently deformed to cause a reduction of the half-life of the process to a value less than 10^{-14} sec. A further necessary condition is that the α-particle should be emitted in a direction nearly opposite to the direction of motion of the parent fragment, in order to explain the observed angular distribution. One feature of this hypothesis is that it explains the pronounced asymmetry of division of the compound nucleus which occurs when long-range particles are emitted. It also relates the abundance of light particles to that of a specific range of primary fragments, namely those with $z = 40$ and $z = 60$ approximately, and this may in principle be determined by other means (Plutonium Project Report 1946).

In conclusion, it may be noted that the emission of light nuclei in fission should not be confined to the slow neutron induced fission of U^{235}, but should occur also in fission of other elements. Farwell *et al.* (1947) have in fact already reported the emission of long-range particles in the fission of plutonium Pu^{239} by slow neutrons, and further experiments to detect the effect in fission of U^{238}, Th^{232}, etc., may be expected.

We are indebted to Professor N. Feather for much helpful advice and criticism throughout this work, and to Dr W. E. Burcham and Mr J. Dainty for valuable discussions. We wish to thank the Department of Scientific and Industrial Research for financial assistance.

Appendix

It is required to calculate the fraction (f) of particles of range R which is contained within a photographic emulsion of thickness $2d$. It is assumed that the sources of the particles are uniformly distributed throughout the emulsion, and that they are emitted isotropically.

Consider those particles originating in a layer of thickness dx, parallel to the boundary planes and at a distance x from the median plane ($x = 0$), as shown in figure 16. If, now, $R<d$, and $x<d-R$, none of these particles escapes from the emulsion.

If $R<d$ and $d-R<x<d$, then a fraction of the particles escapes and this fraction is equal to the ratio (p) of the solid angle subtended by the upper segment in figure 16 to the entire solid angle of 4π. Thus

$$p = \frac{1}{2}\left(1-\frac{d-x}{R}\right).$$

The total number of particles produced in the domain $0<x<d$ and escaping from the emulsion, is as follows, provided that $R<d$:

$$n_1 = \int_{d-R}^{d} \frac{1}{2}\left(1-\frac{d-x}{R}\right) dx$$
$$= \tfrac{1}{4}R.$$

The fraction remaining inside the emulsion is then

$$f_1 = 1-\frac{R}{4d}.$$

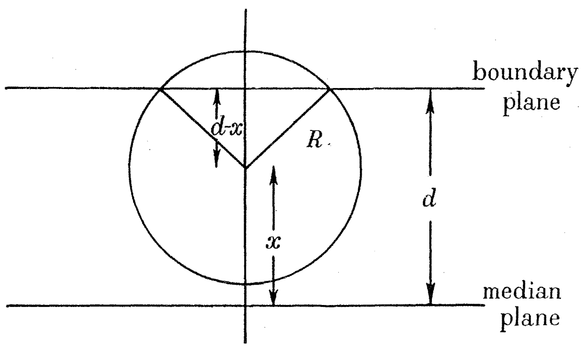


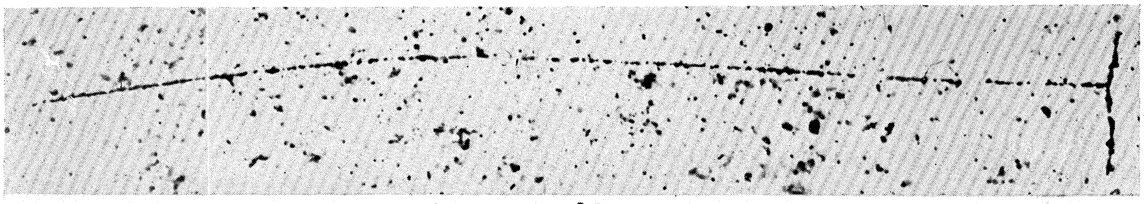
Figure 16

The same formula is applicable to the cases when $d<R<2d$, as may be seen by considering that the number escaping from the domain $0<x<d$ is then effectively:

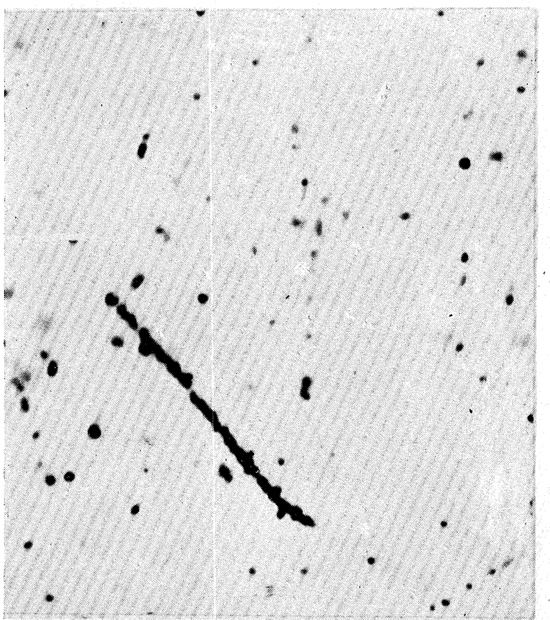
$$n_2 = \int_{-R+d}^{d} \frac{1}{2}\left(1-\frac{d-x}{R}\right) dx$$
$$= n_1.$$

If $R>2d$, the fraction f may be determined directly. For all values of x between $-d$ and $+d$, the sphere of radius R cuts both boundary planes, and the area of the segment enclosed by the planes is constant, being equal to $4\pi Rd$. The escape fraction is therefore

$$f_2 = \frac{d}{R}.$$

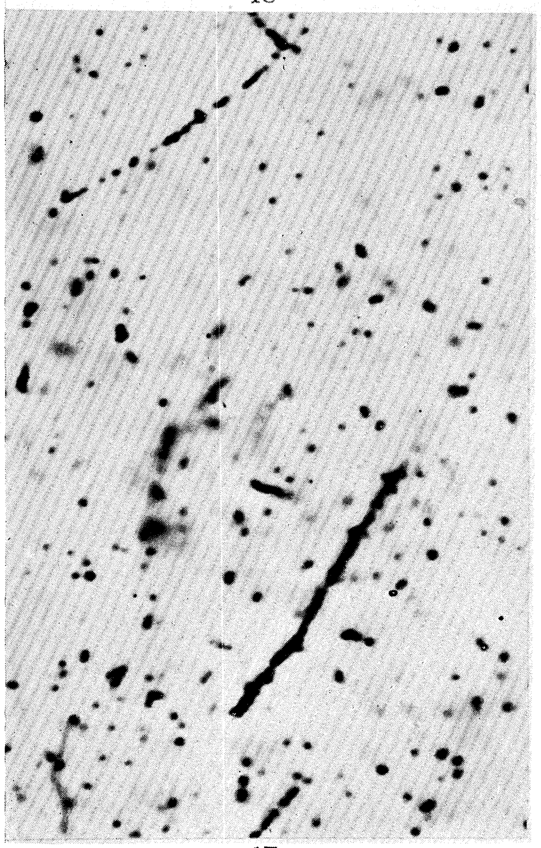


20

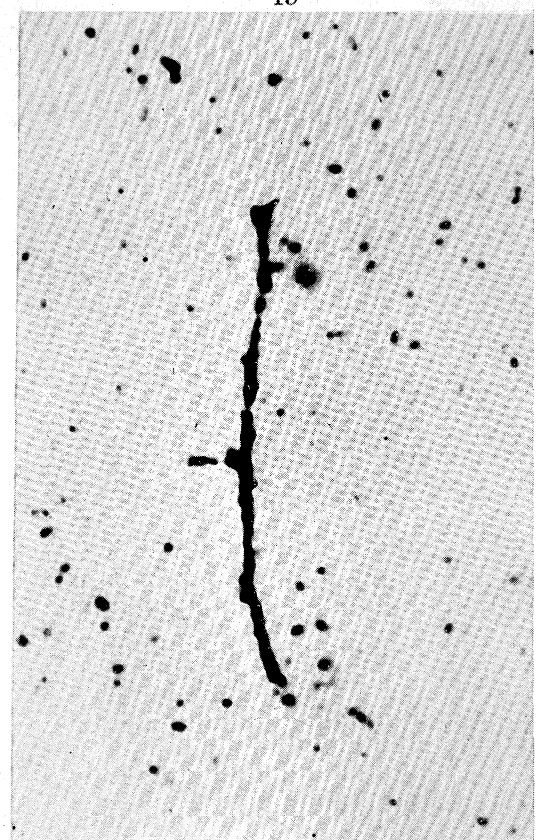


18

19



17



21

FIGURES 17 TO 21

(*Facing p.* 343)

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

References

Bøggild, Brostrøm & Lauritsen 1940 *K. danske. vidensk. Selsk.* **18**, no. 4.
Bohr 1940 *Phys. Rev.* **58**, 654.
Bohr 1941 *Phys. Rev.* **59**, 270.
Bohr & Wheeler 1939 *Phys. Rev.* **56**, 426.
Cassels, Dainty, Feather & Green 1947 *Proc. Roy. Soc.* A, **191**, 428.
Demers 1946*a* *Phys. Rev.* **70**, 86.
Demers 1946*b* *Phys. Rev.* **70**, 974.
Farwell, Segré & Wiegand 1947 *Phys. Rev.* **71**, 327.
Feather 1947 *Nature*, **159**, 607.
Green & Livesey 1946 *Nature*, **158**, 272.
Green & Livesey 1947 *Nature*, **159**, 332.
Knipp & Teller 1941 *Phys. Rev.* **59**, 659.
Lark-Horowitz & Miller 1941 *Phys. Rev.* **59**, 941.
Lattes, Fowler & Cüer 1947 *Proc. Phys. Soc.* **59**, 883.
Plutonium Project Report 1946 *Rev. Mod. Phys.* **18**, 513.
Powell, Occhialini, Livesey & Chilton 1946 *J. Sci. Instrum.* **23**, 102.
Present & Knipp 1940 *Phys. Rev.* **57**, 751.
Tsien, Chastel, Ho & Vigneron 1946 *C.R. Acad. Sci., Paris*, **223**, 986.
Tsien, Ho, Vigneron & Chastel 1947 *Nature*, **159**, 773.
Tsien 1947 *C.R. Acad. Sci., Paris*, **224**, 1056.

Description of Plate 11

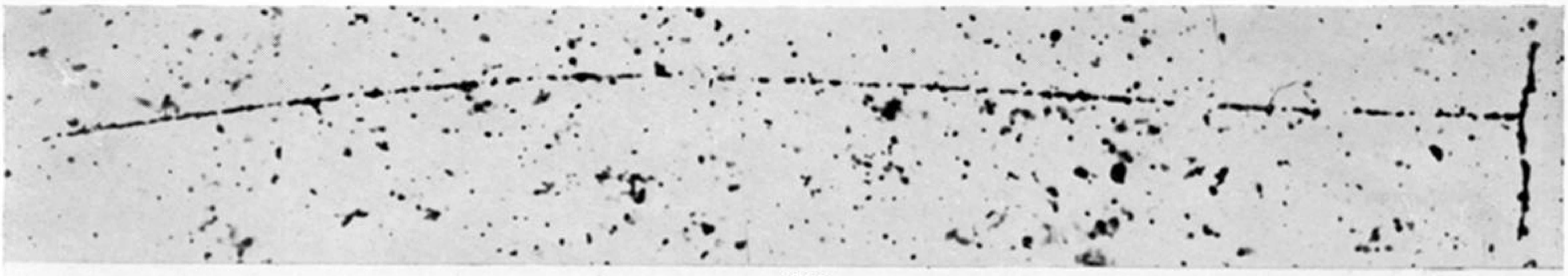
Figure 17. Fission fragment track and α-particle track in plate impregnated with 1% solution of uranyl acetate.

Figure 18. Fission fragment track and α-particle tracks in plate impregnated with 4% solution of uranyl acetate.

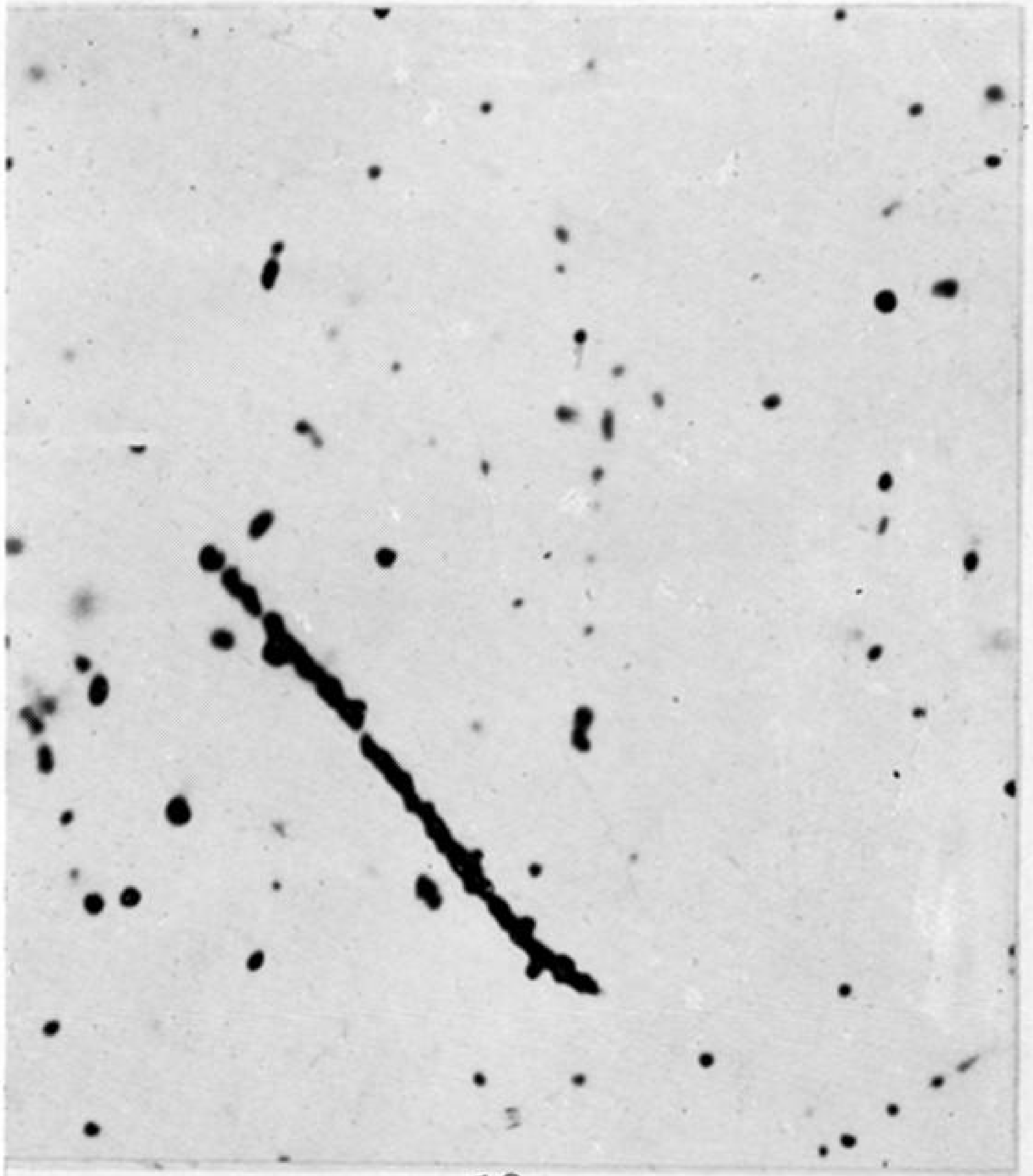
Figure 19. Typical fission fragment track showing close nuclear collisions.

Figure 20. Fission event in which a long-range light particle is emitted.

Figure 21. Fission event in which a short-range light particle is emitted.

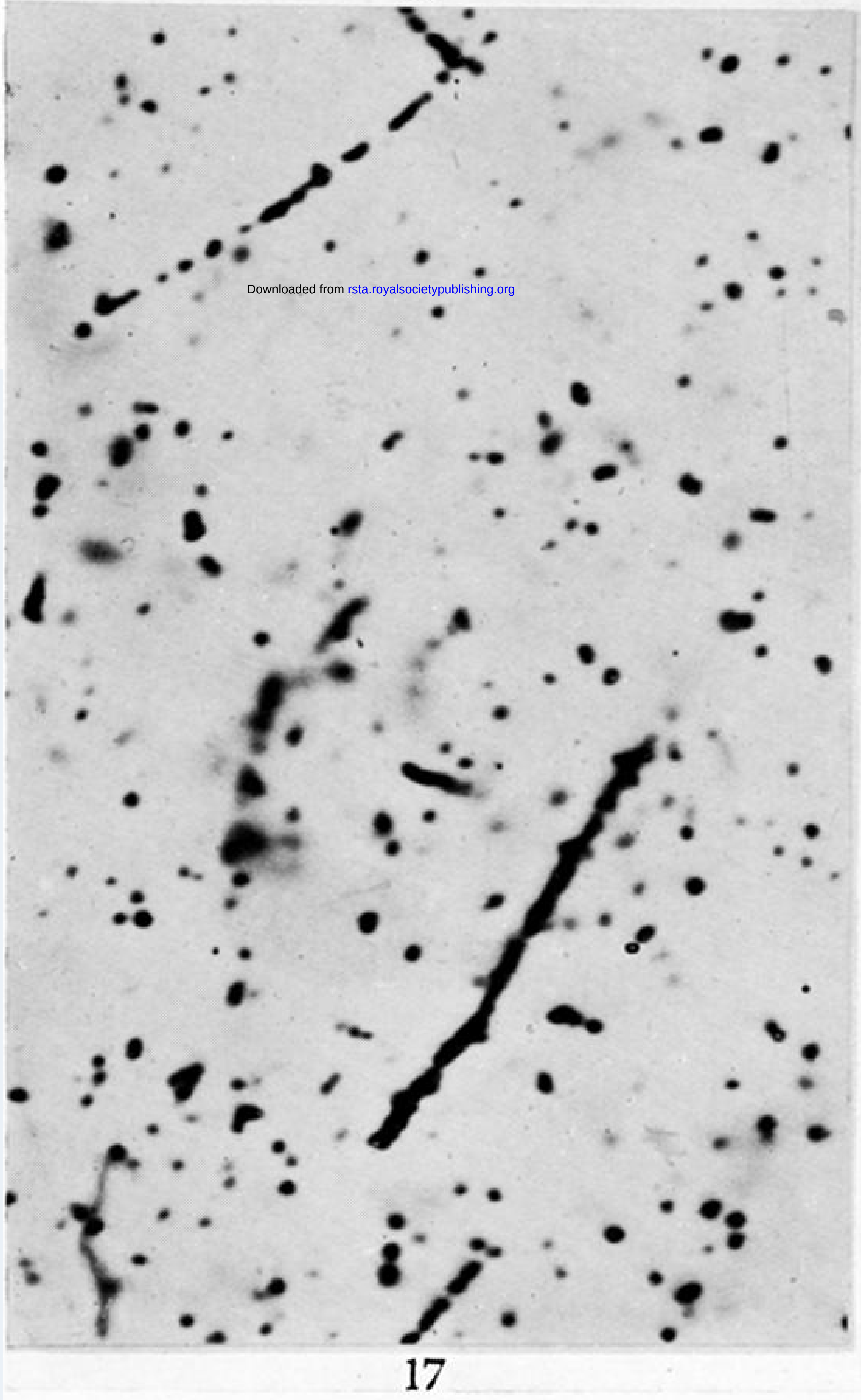


20

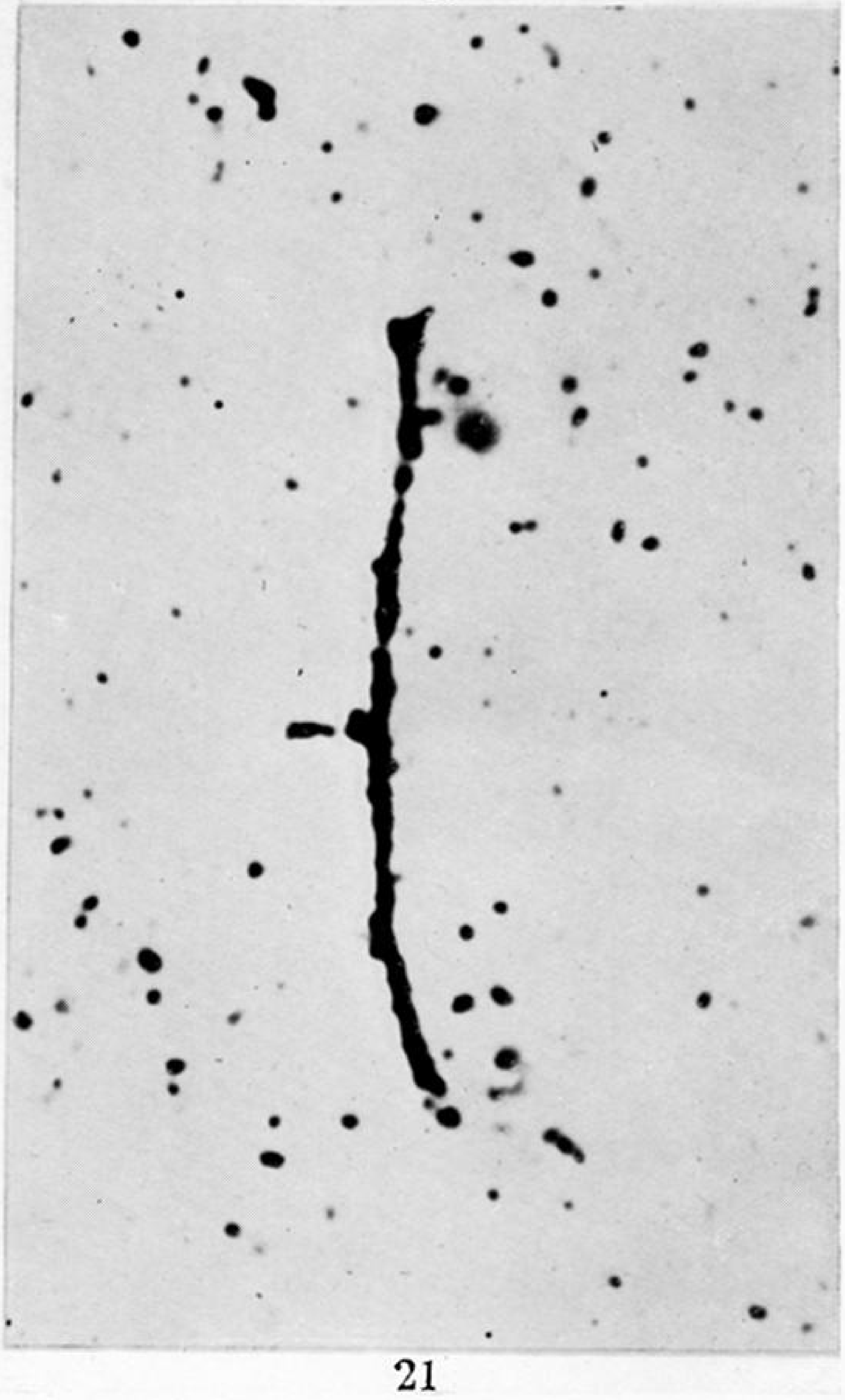


18

19



17



21

Figures 17 to 21

(*Facing p.* 343)

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES