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The Transmutation of Heavy Hydrogen Investigated by the Cloud-Track Method

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[Plates 4 and 5]

§ 1—Introduction

One of the most important cases of artificial transmutation which have been investigated is that in which compounds containing heavy hydrogen are bombarded with high velocity ${}_1H^2$ ions. It was first shown by Oliphant, Harteck and Lord Rutherford* that such bombardment resulted in a copious emission of two proton groups of ranges 15·0 cm and 16·0 mm the relative numbers of particles in the two groups being equal within the errors of measurement. They postulated as a mechanism for the process involved the reaction

$$ {}_1H^2 + {}_1H^2 \rightarrow {}_1H^1 + {}_1H^3 \qquad (1)$$

and showed that if one assumed the ${}_1H^1$ particles to constitute the 15·0 cm group then the range of the ${}_1H^3$ particles, calculated by the application of conservation of momentum to this process, was in approximate agreement with the shorter range. Expansion chamber photographs obtained by one of us† showed that in this bombardment the particles of 15·0 cm and 16·0 mm range were emitted in pairs in opposite directions, this result providing strong evidence in favour of the proposed reaction. In addition to this proton emission, however, it was shown by Oliphant, Harteck and Lord Rutherford* that an intense neutron emission resulted from this bombardment and from an expansion chamber investigation by one of us‡ of helium nuclei recoiling from these neutrons it was shown that the neutrons were probably homogeneous and that their energy was $1 \cdot 8 \times 10^6$ electron volts. An investigation by Oliphant,

* 'Proc. Roy. Soc.,' A, vol. 144, p. 692 (1934).
† Dee, 'Nature,' vol. 133, p. 564 (April 14, 1934).
‡ Dee, 'Proc. Roy. Soc.,' A, vol. 148, p. 628 (1935).

Harteck and Lord Rutherford using a linear counting chamber gave a maximum value for the neutron energy of about 2×10^6 electron volts. The reaction proposed by Oliphant, Harteck and Lord Rutherford to account for these neutrons was

$${}_1H^2 + {}_1H^2 \rightarrow {}_0n^1 + {}_2He^3. \qquad (2)$$

The mass of the ${}_2He^3$ atom was obtained by them from the data for the disintegration of ${}_3Li^6$ by protons, $\{{}_3Li^6 + {}_1H^1 \rightarrow {}_2He^3 + {}_2He^4\}$, and using the value thus derived (3·0166) and a neutron mass of 1·0067 they showed that the energy of the neutrons calculated from (2) was $2 \cdot 5 \times 10^6$ electron volts in approximate agreement with the observed value. Attempts were made by counting methods to detect the ${}_2He^3$ nuclei which are to be expected according to the above reaction but were unsuccessful. The range to be expected from the above data is only 5 mm and the detection of particles of so short a range is very difficult and could not be expected under their experimental conditions. We first tried to detect these particles by passing a beam of ${}_1H^2$ ions into an expansion chamber on to a solid target of heavy ammonium sulphate, but this experiment also failed probably on account of exchange of the heavy hydrogen in the target with the hydrogen of the water used in the chamber.

It was decided therefore to build a small sealed-off expansion chamber which could be filled with heavy hydrogen (the necessary saturation being produced by the addition of a drop of heavy water) and to bombard this gas with a beam of ${}_1H^2$ ions admitted through a thin mica window. It was also decided to look for the ${}_2He^3$ nuclei which may be ejected in the direction of the bombarding ${}_1H^2$ ions since, as will be shown in what follows, their range in this direction is appreciably greater than for those emitted at right angles to the incident beam, and their detection thereby greatly facilitated.

§ 2—Experimental Procedure

The application of the laws of conservation of energy and momentum to the proposed reaction (2) leads to a value for the velocity v_3 of the ${}_2He^3$ nucleus emitted at an angle θ_3 with the direction of the incident ${}_1H^2$ ion of velocity v given by

$$v_3 = \tfrac{1}{2}v \cos \theta_3 + \sqrt{\tfrac{1}{4}v^2 \cos^2 \theta_3 + \tfrac{1}{6}(E - v^2)},$$

where E = the energy liberated in the reaction (ergs ÷ mass of proton).

Using values for $E = 2\cdot4$ and $2\cdot8 \times 10^6$ electron volts in approximate agreement with the data on the energy of the neutrons and taking $\theta_3 = 0°$, the curves *c*, *d* of fig. 1 have been plotted giving the ranges to be expected for the $_2He^3$ nuclei emitted in the forward direction for different energies of the incident $_1H^2$ ion. The curves *a*, *b* show the ranges of $_1H^2$ and $_1H^1$ nuclei plotted against the accelerating voltage.

Now since in this work no magnetic analysis of the beam could easily be applied and since the beam contains both $_1H^2$ and $_1H^1$ nuclei it is evident that with $_1H^2$ and $_1H^1$ particles of 400 electron kv energy there

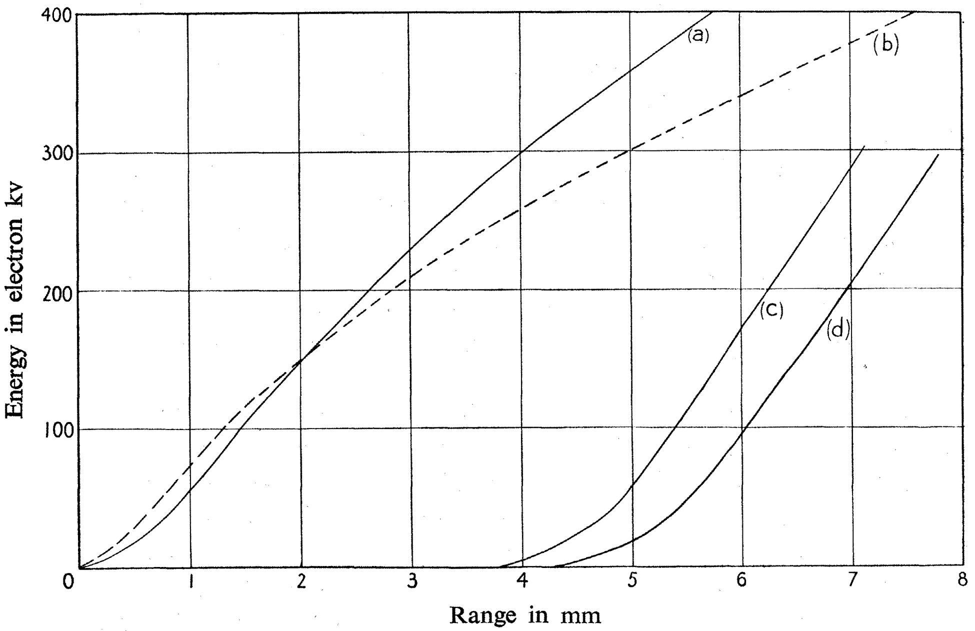

FIG. 1—(*a*) $_1H^2$ ions; (*b*) $_1H^1$ ions; (*c*) $E = 2\cdot4 \times 10^6$ e-volts; (*d*) $E = 2\cdot8 \times 10^6$ e-volts

is no hope of detecting the $_2He^3$ particles since their range is then not greater than that of the bombarding beam. It also appears that up to 150 kv the *difference* between the ranges of the $_2He^3$ particles and the $_1H^2$ or $_1H^1$ particles should not vary much and is equal to about 4 mm.

Above this voltage the excess range rapidly decreases. It was obviously important therefore that the energy of the $_1H^2$ ions entering the chamber should be kept below this value. It is further obvious from this curve that the best results would be obtained by the use of the thinnest possible mica window between the accelerating tube and the chamber since the range of the $_1H^1$ ions increases more rapidly than the range of the $_1H^2$

for accelerating voltages > 150 kv and since for a given energy of the ${}_1H^2$ ions entering the chamber the proton range must be kept as small as possible. The great difficulty of detecting the ${}_2He^3$ nuclei emitted at right angles to the direction of the bombarding beam is also apparent. In this direction the range of the ${}_2He^3$ nucleus is only 3·8 mm for zero bombarding energy, and actually decreases as the energy of the incident particle is increased. Thus even if sufficient numbers of disintegrations

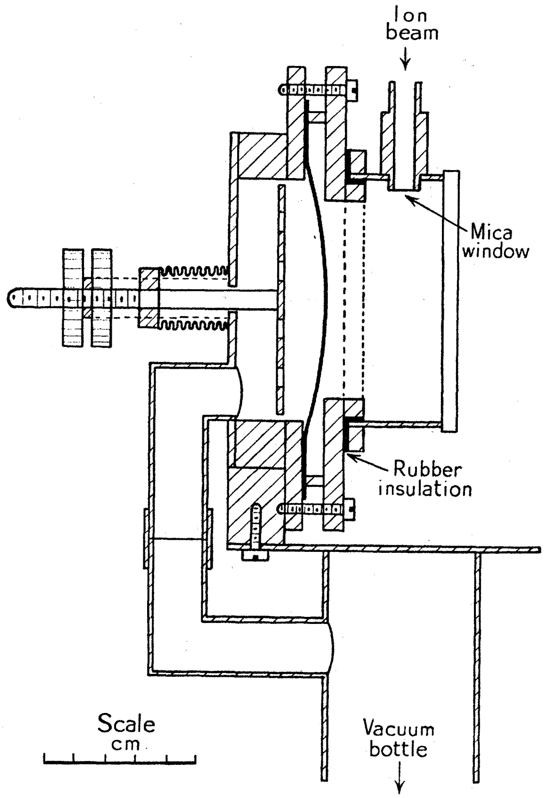

FIG. 2

could be obtained with ${}_1H^2$ ions of 150 electron kv energy and range equal to 2 mm there would be an excess range of the ${}_2He^3$ nucleus of less than 1·8 mm available for detection. The window finally used had an α-particle stopping power of 2·8 mm and was supported upon the plane end of a cylindrical tube containing a large number of holes of diameter 0·33 mm.

A diagram of the expansion chamber is shown in fig. 2. A rubber diaphragm and wire gauzes were used in the manner recommended by

Professor C. T. R. Wilson.* It was found convenient to make a gas-tight rubber seal by clamping the rubber between a flat circular brass plate and a circular brass ring of 2 mm wall thickness. The chamber proper was a separate unit made of brass with glass windows and carrying insulated wire gauzes. This unit was insulated from the brass rings clamping the rubber and made gas-tight with plasticene. The circular rubber sheet in its initial position made contact with a wire gauze and on expansion was drawn back against a perforated brass plate. The position of the latter plate could be readily adjusted, and determined the expansion ratio. An electric field of about 10 volt/cm was maintained between the gauze and the chamber. The expansion was made by the sudden evacuation of the space behind the rubber. The beam was admitted to the chamber at the completion of each expansion during an interval of 1/30 second by means of an electrically operated shutter contained in the evacuated tube connecting the accelerating tube to the expansion chamber. The procedure was as described in previous papers.

The expansion chamber was filled to a pressure of about 40 cm with a mixture of approximately 50% He, 25% ${}_1H_2^2$, 25% air and at the completion of a run the stopping power of the gas was determined by admitting α-particles from a source of ThC through a small window in the base of the chamber. Under these conditions the track length photographed was about five times its length at N.T.P.

§ 3—Experimental Results

A preliminary run of photographs at different accelerating voltages gave clear evidence of the existence of a group of tracks of about the range sought. Tracks which passed right across the chamber were also observed, their reduced range being > 15 mm, and must doubtless be attributed to ${}_1H^1$ and ${}_1H^3$ nuclei produced according to reaction (1). Subsequent experiments were made keeping the bombarding energy as low as possible consistent with obtaining a few tracks per photograph. Typical photographs are shown in figs. 4–7, Plates 4 and 5. The plates were measured by replacing them in the cameras and reprojecting their images in space. For this purpose a dummy chamber was constructed with the same dimensions as the one actually employed but without the glass plates. By reprojecting the images of the illumination apertures on to this chamber it was possible to decide with precision whether an apparent short track actually ended within the illumination or whether it was to be attributed to one of the longer tracks passing out of the illuminating beam. Since under these conditions of use of an expansion

* 'Proc. Roy. Soc.,' A, vol. 142, p. 88 (1933).

chamber dense condensation is produced throughout the whole chamber making the operation very critical only the best plates were measured. In a large number of cases it was obvious that tracks ended in the chamber from the small deflections visible at their ends, apart from the further test as to the position of this end relative to the illuminating beam. It was of course, impossible to observe the point of origin of any particular track within the sphere of ionization due to the entering beam. The " length " of each track was therefore determined by projecting its direction back to the mica window where the beam entered, and measuring from this point. The ranges thus obtained are therefore only true ranges for those tracks which actually originated at the window. Measure-

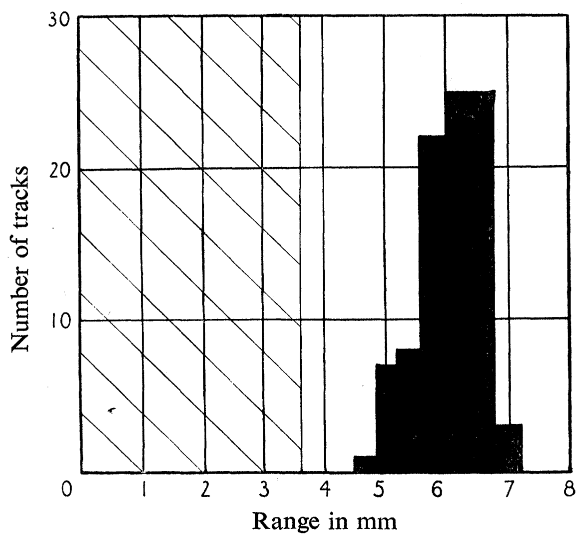

FIG. 3

ment of the tracks was confined to those emitted within 30° of the forward direction and as will be shown later it is easy to deduce the true range from the " range " distribution obtained in this manner. The " range " distribution measured directly in this manner is shown in fig. 3 where the abscissa scale has been adjusted to correct these ranges to air at N.T.P.

The reduced range corresponding to the edge of the beam was 3·6 mm. The mica window through which the beam passed had a stopping power of 2·8 mm whence the maximum range of the particles in the beam was 6·4 mm. These particles of range 6·4 mm were therefore protons of energy 355 electron kv. The accelerated ${}_1H^2$ ions had therefore a total range of 4·9 mm and a range of 2·1 mm in the chamber.

The existence of a group of particles of maximum range 6·8 mm is clearly shown. Using the curves of fig. 1 and a range of the ${}_1H^2$ ions

$\doteq$ 2·1 mm the expected range of the $_2He^3$ particles would be 6·6 mm for an energy release in the reaction of $2 \cdot 8 \times 10^6$ electron volts. This mode of comparing the theoretical and experimental results is not obviously justifiable. It would be correct only if all the disintegration particles were produced at the mica window where the $_1H^2$ ions entered. In order to obtain a more accurate value for the energy balance and to justify this comparison it is necessary to consider the range of $_2He^3$ nuclei produced at all distances from this window up to the range of the entering $_1H^2$ ions. This calculation is made in § 4.

No tracks having ranges between 6·8 mm and 1·5 cm were observed, and the fall in number on the short range side suggests also that the observed group is not continuous on the low energy side, since it would have been possible to detect tracks on these photographs up to within a reduced distance of 0·5 mm of the edge of the beam. The ratio of the numbers of tracks in this group to the numbers passing out of the chamber was $0 \cdot 41 \pm 0 \cdot 06$ corresponding to a relative probability of the processes of disintegration into neutrons and $_2He^3$ nuclei and disintegration into $_1H^1$ and $_1H^3$ particles of $0 \cdot 8 \pm 0 \cdot 1$. Two short control runs were taken with ordinary hydrogen in the chamber instead of heavy hydrogen the same proportions of air and helium being present. The first run showed several tracks of about the range of the $_2He^3$ particles and also tracks going right across the chamber. The numbers were about a quarter of those obtained with heavy hydrogen in the chamber. In the second run, taken a day later, the number of tracks obtained per photograph was less than one-tenth of the number with heavy hydrogen in the chamber although the $_1H^2$ ion beam was stronger than in previous runs. The high number obtained in the first run was almost certainly due to exchange occurring between the hydrogen introduced and heavy hydrogen from the heavy water which remained from the main experiments. This contamination effect was considerably reduced on allowing the chamber to stand overnight after introducing excess of ordinary water. It seems clear from these results that the short range group of particles is due to the bombardment of heavy hydrogen with its own ions.

§ 4—The Energy Balance in the Reaction

As we have shown in the above paragraph it is necessary to consider the fact that disintegration may occur at any distance from the mica window up to a value equal to the range of the $_1H^2$ ions (*i.e.*, 2·1 mm), the "ranges" occurring in the distribution diagram being true ranges only for those $_2He^3$ nuclei produced at the mica window. In Table I, row 2 gives the actual ranges of the $_2He^3$ nuclei obtained from the curves of fig. 1 using the value $E = 2\cdot4 \times 10^6$ e-volts and different energies of the $_1H^2$ ions corresponding to the energies which these ions (of initial range 2·1 mm) would have at distances D from the plane of entry.

In row 4 the true range of row 2 has been added to the distance D giving the " ranges " as measured.

Rows 3 and 5 give the same data for an assumed value of $E = 2\cdot8 \times 10^6$ e.v.

TABLE I

	mm	mm	mm	mm
(1) D	0	0·7	1·4	2·1
(2) Calculated range of $_2He^3$ nuclei for $E = 2\cdot4 \times 10^6$ e.v.	5·9	5·3	4·7	3·8
(3) Calculated range of $_2He^3$ nuclei for $E = 2\cdot8 \times 10^6$ e.v.	6·6	6·0	5·3	4·3
(4) Theoretical " range " as measured for $E = 2\cdot4 \times 10^6$ e.v.	5·9	6·0	6·1	5·9
(5) Theoretical " range " as measured for $E = 2\cdot8 \times 10^6$ e.v.	6·6	6·7	6·7	6·4

It is obvious that the ranges measured in the manner described are practically unaffected by changes in the point of origin, any increase in the distance of this point from the window being counterbalanced by the reduction in the effect of the energy of the bombarding $_1H^2$ ion. The maximum observed range was 6·8 mm agreeing closely with the maximum value of row 5. Thus the value for the energy balance in the reaction is $E = 2\cdot8 \times 10^6$ e.v. The observed distribution is from 4·8 mm to 6·8 mm the main peak lying between 5·6 mm and 6·8 mm, whereas the theoretical width is from 6·4 to 6·7 mm. Part of this difference must be attributed to the fact that the tracks measured made angles up to 30° with the incident beam. For this angle the range of the $_2He^3$ nucleus would be 6·1 mm. The remaining difference may be partly due to straggling of the $_1H^2$ and $_2He^3$ particles. The estimated error in the value of E is $0\cdot2 \times 10^6$ e-volts and lies mainly in the determination of the stopping power of the gas mixture used.

Thus from these experiments we obtain for the energy balance in the reaction

$$_1H^2 + {}_1H^2 \rightarrow {}_2He^3 + {}_0n^1,$$

a value $2\cdot8 \pm 0\cdot2 \times 10^6$ e-volts. It follows from the application of the law of conservation of momentum to this process that the neutron and

${}_2He^3$ nucleus are emitted in opposite directions for zero bombarding energy. In this case also the energy of the neutron would be

$$\tfrac{3}{4}(2\cdot8 \pm 0\cdot2 \times 10^6) \text{ e.v.} = 2\cdot1 \pm 0\cdot2 \times 10^6 \text{ e.v.}$$

and the energy of the ${}_2He^3$ nucleus $\tfrac{1}{4}(2\cdot8 \times 10^6) = 0\cdot7 \times 10^6$ e.v. Thus the range of the ${}_2He^3$ nucleus for zero bombarding energy would be 4·3 mm.

The investigation of the neutron emission* using the expansion chamber gave a value for the energy of the neutrons $= 1\cdot8 \pm 0\cdot2 \times 10^6$ e-volts, which agrees with the value deduced above within the limits of error. From the latter value we obtain for the energy release in the process a value $\tfrac{4}{3}(1\cdot8 \pm 0\cdot2 \times 10^6)$ e.v. $= 2\cdot4 \pm 0\cdot3 \times 10^6$ e.v. Thus the mean value for the energy release from the two sets of experiments is $2\cdot6 \pm 0\cdot2 \times 10^6$ e-volts or $0\cdot0028 \pm 0\cdot0002$ mass units. For the reaction in question the only mass known with precision is that of ${}_1H^2$ which, according to Bainbridge,† is $2\cdot0136 \pm 0\cdot00007$. Applying the conservation of energy to the process we obtain for the sum of the masses of ${}_2He^3$ and ${}_0n^1$ a value

$$2(2\cdot0136 \pm 0\cdot00007) - (0\cdot0028 \pm 0\cdot0002) = 4\cdot0244 \pm 0\cdot0003.$$

It is possible, however, to obtain the mass of ${}_2He^3$ from another set of transmutation experiments.

In the disintegration of lithium by protons a group of particles of range < 2 cm was detected by Cockcroft and Walton.‡ Subsequent work by Oliphant, Kinsey and Lord Rutherford§ showed these particles to consist of two groups one of range 11·5 mm and the other of range 6–8 mm. They proposed for the nuclear reaction

$${}_1H^1 + {}_3Li^6 \rightarrow {}_2He^3 + {}_2He^4,$$

and expansion chamber photographs by one of us‖ showed that these particles were emitted in opposite pairs as is required by the application of momentum considerations to the process assumed. Still further proof of the correctness of these views has been obtained by the bombardment of the separated isotopes of lithium when the short range products were shown to arise from the ${}_3Li^6$ isotope.¶ It follows from the energies

* Dee, 'Proc. Roy. Soc.,' A, vol 148, p. 628 (1935).
† 'Phys. Rev.,' vol. 44, p. 57 (July 1, 1933).
‡ 'Proc. Roy. Soc.,' A, vol. 137, p. 229 (1932).
§ 'Proc. Roy. Soc.,' A, vol. 141, p. 722 (1933).
‖ Dee, 'Nature,' vol. 132, p. 818 (1933).
¶ Oliphant, Shire and Crowther, 'Proc. Roy. Soc.,' A, vol. 146, p. 922 (1934).

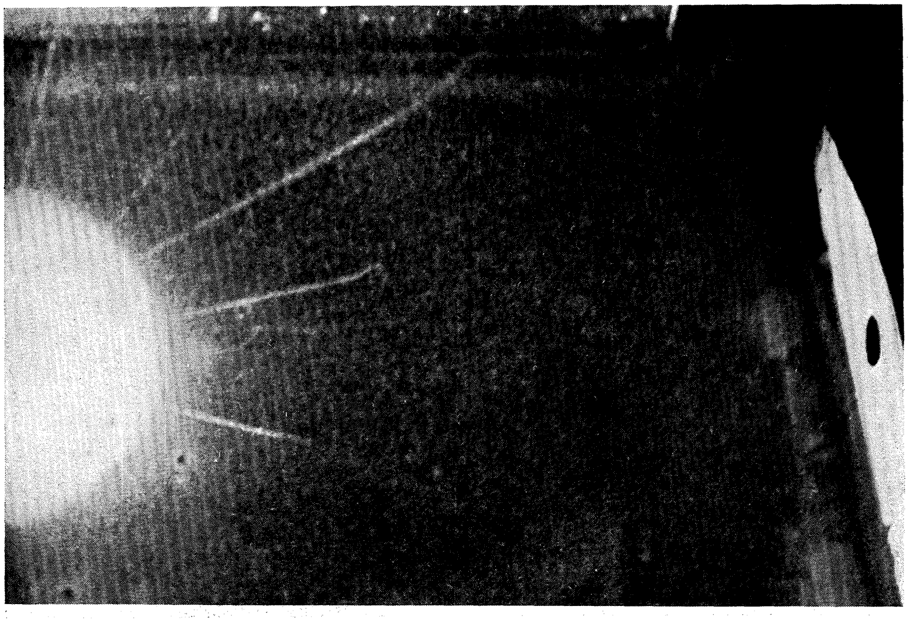

Fig. 5

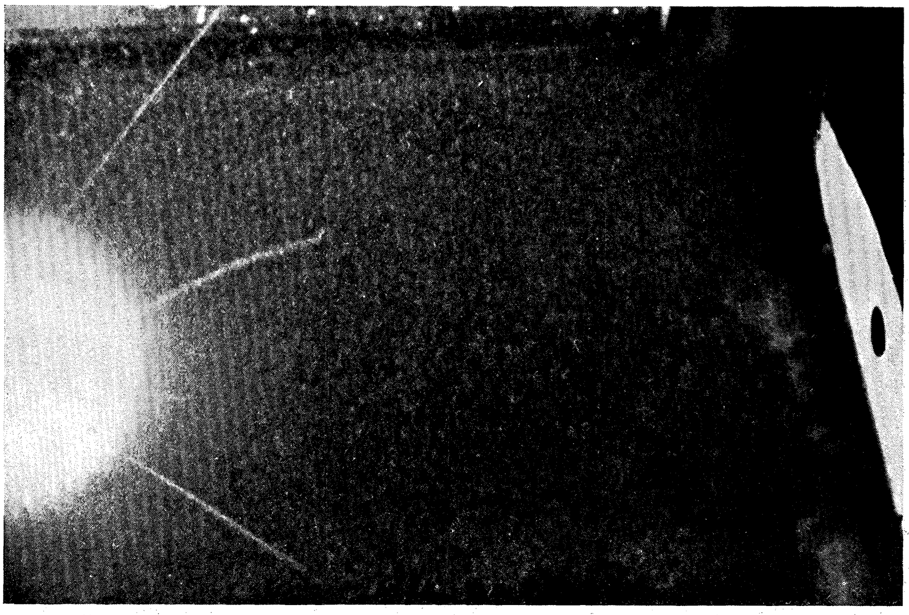

Fig. 4

(*Facing p.* 208)

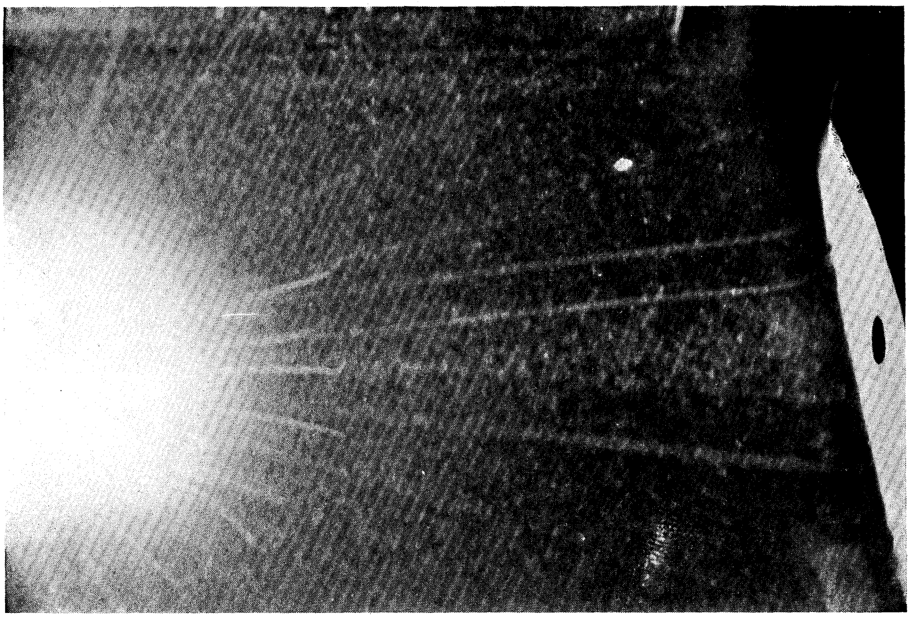

FIG. 7

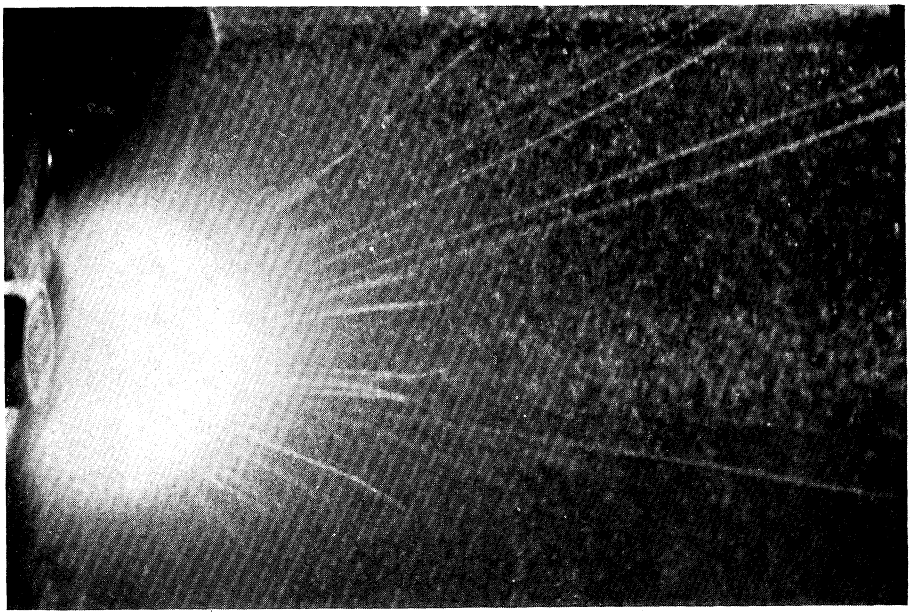

FIG. 6

of these short range particles that the mass of $_2He^3$ is $3 \cdot 0164 \pm 0 \cdot 0003$ and hence we obtain for the mass of the neutron, from the experiments of this paper, a value

$$(4 \cdot 0244 \pm 0 \cdot 0003) - (3 \cdot 0164 \pm 0 \cdot 0003) = 1 \cdot 0080 \pm 0 \cdot 0004,$$

in good agreement with the value obtained by Chadwick and Goldhaber* from the disintegration of heavy hydrogen by γ-rays.

We are indebted to Lord Rutherford for his interest and encouragement in this research.

One of us (C. W. G.) wishes to acknowledge a Senior Research Award by the Department of Scientific and Industrial Research. We wish to thank Mr. Birtwhistle for much technical assistance.

Summary

The $_2He^3$ nuclei produced in the reaction

$$_1H^2 + {}_1H^2 \rightarrow {}_2He^3 + {}_0n^1$$

have been detected in the expansion chamber by passing a beam of artificially accelerated $_1H^2$ ions into a gas mixture containing heavy hydrogen.

The range of this group of particles has been measured and is $4 \cdot 3 \pm 0 \cdot 2$ mm for zero bombarding energy. The neutrons produced in the same bombardment have an energy of $1 \cdot 8 \pm 0 \cdot 2 \times 10^6$ e-volts, and it is shown that these results are in agreement with the application of the conservation of momentum to the process assumed. A value of $1 \cdot 0080 \pm 0 \cdot 0004$ is thereby deduced for the mass of the neutron.

Description of Plates

The reproductions are $1 \cdot 5$ times natural size.

The length of a track in the gas is five times its range reduced to N.T.P.

The white fan at the top of each photograph is due to the intense ionization of the entering beam. Figs. 4, 5 show tracks of range about $6 \cdot 8$ mm, the small deflections often visible at their ends is evidence that they end in the gas. Some longer tracks passing out of the chamber are probably due to protons produced according to reaction (1).

Figs. 6, 7 taken with a more intense $_1H^2$ ion beam show clearly the two groups of particles.

* 'Nature,' vol. 134, p. 237 (1934).

FIG. 4

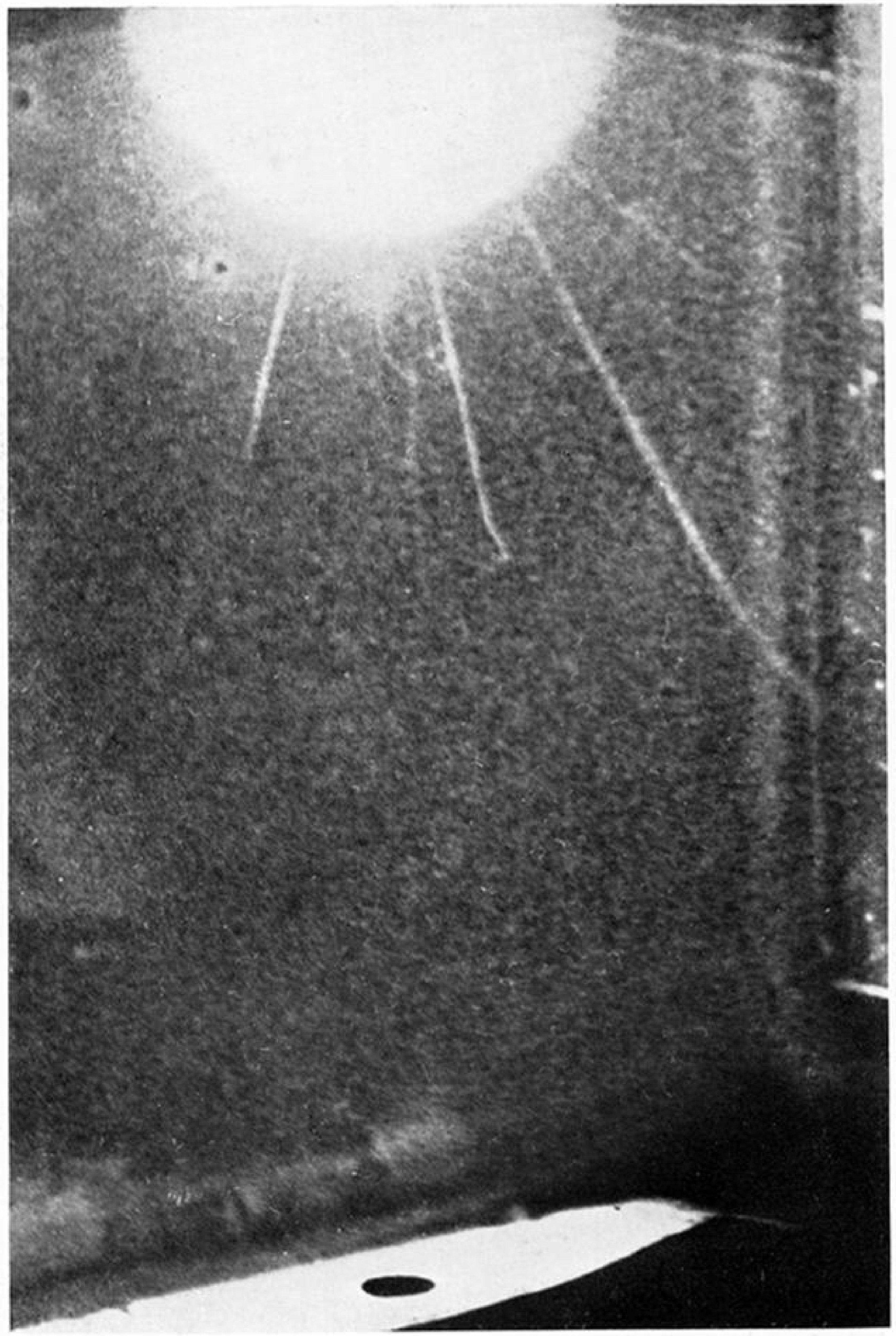

FIG. 5

DESCRIPTION OF PLATES

The reproductions are 1·5 times natural size.

The length of a track in the gas is five times its range reduced to N.T.P.

The white fan at the top of each photograph is due to the intense ionization of the entering beam. Figs. 4, 5 show tracks of range about 6·8 mm, the small deflections often visible at their ends is evidence that they end in the gas. Some longer tracks passing out of the chamber are probably due to protons produced according to reaction (1).

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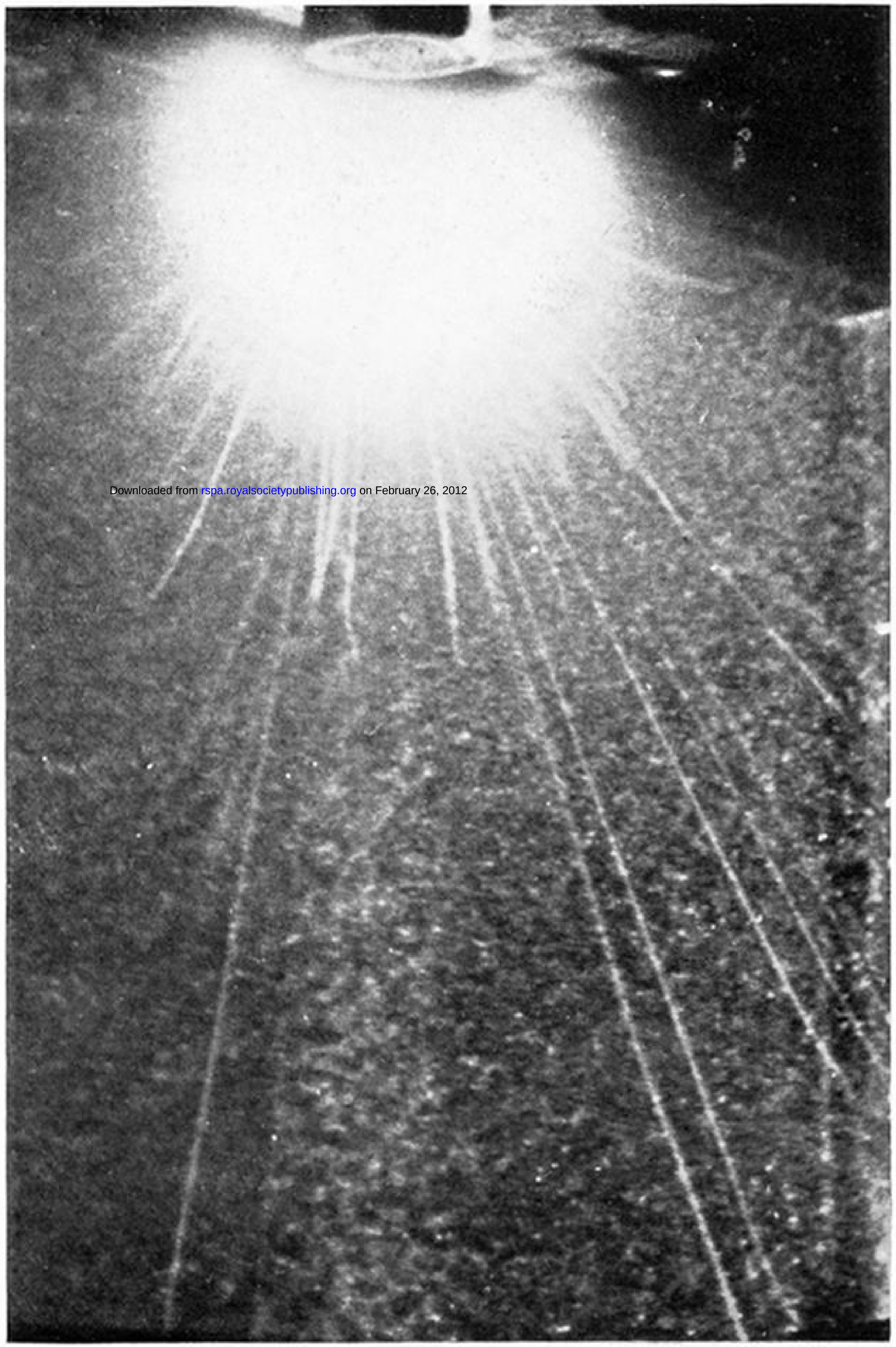

FIG. 6

FIG. 7