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Transmutation Effects Observed with Heavy Hydrogen

M. L. E. Oliphant, P. Harteck and Lord Rutherford

Proc. R. Soc. Lond. A 1934 **144**, 692-703
doi: 10.1098/rspa.1934.0077

Summary.

(1) The optical rotatory power of an asymmetric atom attached to four small radicals has been calculated and is expressed in a practical formula. The rotation is dependent on the refractivities and the sizes of the radicals.

(2) The formula has been confirmed by the calculation of the rotatory powers of amyl alcohol, amylamine, sec-butyl alcohol, and sec-butylamine.

(3) The conclusions drawn from this calculation with regard to optical activity and chemical substitution, and optical activity and physical conditions agree with the characteristic behaviour of optically active compounds.

(4) An exact formula connecting the rotatory dispersion of the compound with the refractive dispersion of the radicals has been obtained.

Transmutation Effects Observed with Heavy Hydrogen

By M. L. E. Oliphant, Ph.D. (Messel Research Fellow of the Royal Society), P. Harteck, Ph.D., and Lord Rutherford, O.M., F.R.S.

(Received April 14, 1934.)

[Plate 16.]

In our paper "Transmutation of Elements by Protons,"* we showed that the transformation of some of the light elements by protons could be conveniently studied by the use of comparatively low voltages—of the order of 100,000 volts—by generating an intense narrow beam of protons which fell on the target of small area of about 1 sq. cm. In the light of experience of the past year, the installation has been modified in several particulars and entirely reconstructed. By the addition of another 100,000-volt transformer in tandem and the use of appropriate condensers the D.C. voltage available has been raised from 200,000 to 400,000 volts. The main change, however, consists in the use of a horizontal instead of a vertical discharge tube. In place of glass, a corrugated porcelain wall bushing capable of withstanding high voltages has been used to insulate the positive electrode, while the earthed metal casing forming the negative electrode projects through a brick wall. The arrangement of the internal electrodes is, in general, similar to that used in the

* 'Proc. Roy. Soc.,' A, vol. 141, p. 259 (1933).

earlier apparatus. The oil cooling circulation has been improved as the electrodes cannot now be cooled by radiation alone. As before a magnetic field is applied to sort out the various types of ions generated in the discharge tube. The use of the horizontal tube has many advantages, not only for assembling the controls at convenient points but also in the ease of handling the counting apparatus and absorbing screens.

The new installation has worked smoothly and satisfactorily and we have been able to increase the number of disintegration particles available for study by a factor of 10 to 50. The thick brick wall acts as a complete screen for the X-radiation generated in the system.

In our last paper* we gave an account of the transformations produced in lithium by the ions of heavy hydrogen. The heavy water used for this purpose was generously presented to us by Professor G. N. Lewis. For our present experiments we have depended on a supply of concentrated heavy water prepared in the Cavendish Laboratory by Dr. P. Harteck.† For preliminary requirements a weak concentration of diplogen‡ of about 12% was generally used. Stronger concentrations up to 30% mixture with helium§ were necessary in order to study the emission of neutrons and protons. The action of diplons on diplons was studied by observation of the effects produced when diplons were used to bombard targets covered with a thin layer of a preparation containing heavy hydrogen. These were ammonium chloride, ammonium sulphate, and orthophosphoric acid in which the normal hydrogen had been largely replaced by diplogen. The method of preparation was very simple. A small quantity of the normal ammonium salt or the phosphoric pentoxide was added to an excess of heavy water. An equilibrium was at once established between the concentration of hydrogen and of diplogen in the compound and in the water,‖ and if a drop of the solution was placed upon a warm iron target and allowed to evaporate a stable but non-uniform layer of a salt containing diplogen was left behind. The ND_4Cl was also deposited upon the target in the form of a very thin and a uniform layer by means of sublimation, but this very property renders the target unstable and liable to disappear rapidly

* 'Proc. Roy. Soc.,' A, vol. 141, p. 722 (1933).

† 'Proc. Phys. Soc. Lond.,' vol. 46, p. 277 (1934).

‡ In a discussion on Heavy Hydrogen before the Royal Society on December 14, 1933 (see 'Nature,' p. 955 (1933)), the names diplogen (D) for the new isotope of hydrogen and diplon for its nucleus seemed to find favour. Also 'Proc. Roy. Soc.,' A, vol. 144, p. 1 (1934).

§ *Cf.* Oliphant, Kinsey, and Rutherford, 'Proc. Roy. Soc.,' A, vol. 141, p. 722 (1933).

‖ Lewis, 'Proc. Amer. Chem. Soc.,' vol. 55, p. 3502 (1933).

under bombardment. The phosphoric acid remained as a liquid film over the surface of the target. Exceedingly small quantities of the substances are required.

The Action of Diplons on Diplons

The Emission of Charged Particles—The most interesting and important reaction which we have observed is that of heavy hydrogen on heavy hydrogen itself. Experiment has shown* that diplogen is not appreciably affected by bombardment with α-particles from polonium, and we have been unable to detect any specific action of protons on diplogen for energies up to 300,000 e-volts. We were therefore surprised to find that on bombarding heavy hydrogen with diplons an enormous effect was produced. Fig. 4, Plate 16, shows a reproduction of portion of an oscillograph record obtained in our first experiment. We assumed at first that this was an effect due to radiation passing through the counting chamber as previous experiments had shown that X-rays could produce just the result observed, but subsequent observation at much lower bombarding potentials showed that we were dealing in reality with a very large emission of protons. Examples of an oscillograph record obtained under these conditions are given in figs. 5 and 6, Plate 16. The original observations were made on ND_4Cl, but in order to establish that the effects observed came from the action of D on D and not from the nitrogen or chlorine, we bombarded targets of $(ND_4)_2SO_4$ and of D_3PO_4. The absorption curves obtained for the three substances are given in fig. 1. The shape of these curves is due to the fact that protons gave too small a deflection in the oscillograph to be easily counted except over the last five centimetres of their path.

It is evident from fig. 1 that there are present in each case two very prominent groups of particles of ranges 14·3 and 1·6 cm. respectively. Careful counting of the records established that the numbers of these particles were identical within the errors of measurement. The maximum size of the deflections produced on the oscillograph record by the particles in each group indicated that they both consisted of singly charged particles. On these data it is natural to assume that the particles are emitted in pairs opposite one another, and that the difference in range arises from a difference in mass, and hence of the velocity and energy. The simplest reaction which we can assume is

$$_{1}D^{2} + {}_{1}D^{2} \rightarrow {}_{2}He^{4} \rightarrow {}_{1}H^{1} + {}_{1}H^{3}.\dagger \qquad (1)$$

* Rutherford and Kempton, 'Proc. Roy. Soc.,' A, vol. 143, p. 724 (1934).

† It has been decided to put the symbol for the nuclear charge to the left and below the symbol for the element.

While from the point of view of both experiment and theory, we have no information of the details of the nuclear changes involved and can only study the final result, yet it is often useful and may even be more correct to assume that the first step in the process is the capture of the bombarding particle to form a new and heavier nucleus. If this proves unstable, it then breaks up, possibly in a variety of ways. The recent discovery by M. and Mme. Curie-Joliot,* and by Cockcroft and Walton,† that a type of radioactive nucleus is

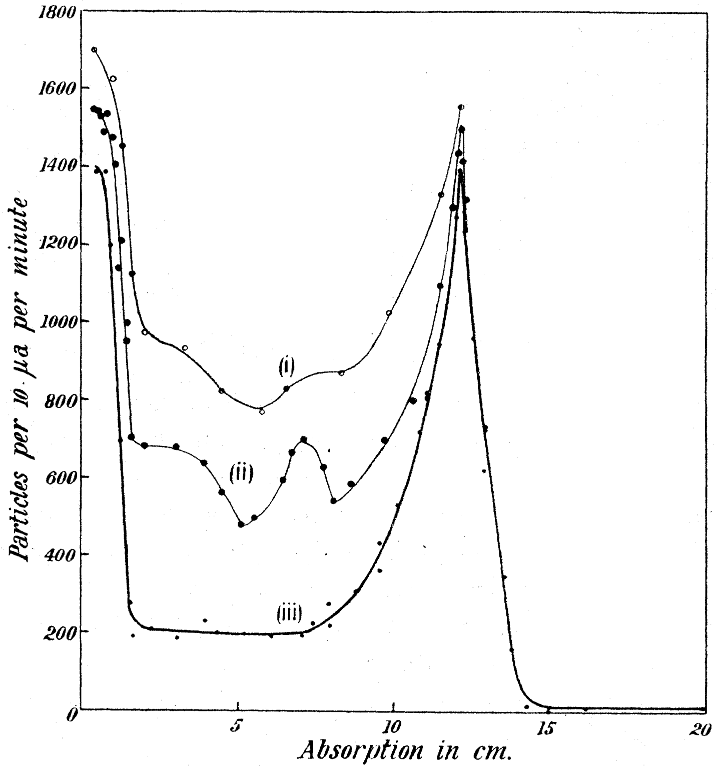

FIG. 1.—(i) ND_4Cl; (ii) $(ND_4)_2SO_4$; (iii) D_3PO_4.

formed by the capture of an α-particle, diplon, or proton lends support to this point of view. The time of transformation may vary over very wide limits and may sometimes be so short that the process cannot be experimentally followed. In the case which we are considering we are inclined to interpret the observations in the following way. The initial process is the union of two diplons to form a new nucleus of charge 2 and mass 4, *i.e.*, a helium nucleus.

* ' C. R. Acad. Sci. Paris,' vol. 198, p. 254 (1934).
† ' Nature,' vol. 133, p. 328 (1934).

If we neglect the energy of the bombarding particle and assume the mass of D to be that given by Bainbridge,* the mass of this helium atom must be 4·0272, and it therefore possesses an excess energy over the normal helium atom, of mass 4·0022, of about 23 million volts. This atom is unstable and may lose its energy in a variety of ways, some of which are considered later. We are considering here the breaking up of the helium nucleus into a proton and a hydrogen isotope of mass 3. The transformation follows so rapidly after the capture that no evidence of the existence of the excited helium nucleus has so far been obtained.

The mass and energy relations on the right-hand side of equation (1) can be obtained in the following way. The ${}_1H^1$ particle possesses an energy of $3 \cdot 0 \times 10^6$ e-volts, corresponding to protons of the observed range of 14·3 cm. Then, from momentum considerations, the energy of the ${}_1H^3$ particle which is emitted in the opposite direction will be $1 \cdot 0 \times 10^6$ e-volts, *i.e.*, the total energy of the two particles will be $4 \cdot 0 \times 10^6$ e-volts, corresponding to a mass-change of 0·0043 units. Hence the mass of the ${}_1H^3$ atom will be

$$4 \cdot 0272 - (1 \cdot 0078 + 0 \cdot 0043),$$

i.e., 3·0151. The ionization produced by the ${}_1H^3$ particle will be at every point of its path identical with that produced by a proton possessing the same velocity. However, owing to its greater momentum the ${}_1H^3$ particle will travel three times as far as the proton for a given reduction of velocity. Consequently it will have three times the range of a proton of the same initial speed. The initial velocity of the ${}_1H^3$, corresponding to an energy of 10^6 e-volts, is 8×10^8 cm./sec. The range of a proton with this velocity is 5·8 mm. according to data given to us by Feather. Hence the range of the ${}_1H^3$ particle should be $3 \times 5 \cdot 8 = 1 \cdot 74$ cm. Considering the nature of the data available and the difficulties of determining the range of the short 1·6 cm. group accurately, we feel that this agreement is very satisfactory.

Additional evidence of the truth of our assumption is afforded by observation of the way in which the ionization, as measured from the magnitude of the oscillograph deflections, varies near the end of the range of the particles. In fig. 2 we have plotted the number of deflections above a given size against the absorption in the path of the particles, using a chamber 3 mm. deep. It is seen that the14 cm. group shows a very sharp peak which occurs about 2 cm. short of the end of the range. The 1·6 cm. group, on the other hand, rises rapidly to what appears to be a much broader maximum. This is just what

* 'Phys. Rev.,' vol. 44, p. 56 (1933).

would be expected from a particle of greater mass, the velocity of which varies less rapidly with the length of path in the material than the proton.

Still further confirmation of the truth of this mode of transformation would be provided if it could be shown that the ${}_1H^1$ and ${}_1H^3$ particles recoil in opposite directions. This point has been carefully examined by Dee using the Wilson expansion chamber method, and in a recent letter to ' Nature '* he concludes that there is no doubt of its correctness.

It seems clear that the production of this isotope of hydrogen of mass 3 in these reactions is established beyond doubt. The mass of the ${}_1H^3$ atom is consistent with its possessing a stability of the same order as ${}_1H^2$. The possible existence of this isotope has been discussed by several writers and although a

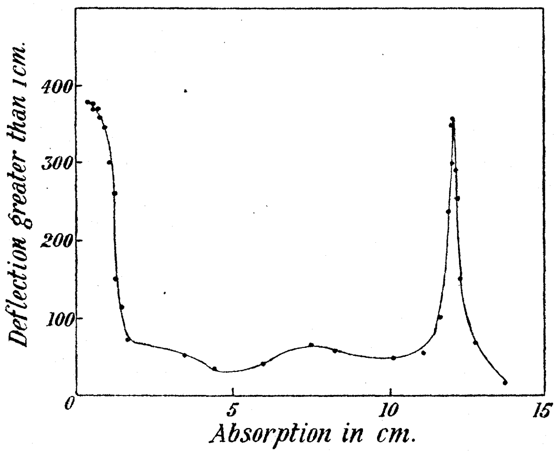

FIG. 2.—$(ND_4)_2SO_4$.

careful search has been made no evidence of its presence has been found. It seems probable, however, that it could be formed by the process we have considered in sufficient quantity to be detected ultimately by spectroscopic or positive-ray methods.

Voltage Variation and Absolute Yield.—The variation with energy of the bombarding diplons, of the emission of 14·3 cm. protons, has been measured over a limited range of voltage. The yield is so great that the number of particles entering the chamber soon became inconveniently large, and also we found that at high bombarding energies the heavy hydrogen compound is rapidly removed from the target. Fig. 3 gives the curve obtained with $(ND_4)_2SO_4$. It is evident that particles are detected at energies as low as 20,000 volts

* ' Nature,' vol. 133, p. 564, April 14th (1934).

and that the number increases rapidly with increase of accelerating potential.* The nearly linear rise beyond 100,000 volts is probably due to the fact that, for potentials greater than this, the chance of disintegration in a collision is nearly constant, and the number of transformations therefore increases proportionally with the penetration of the bombarding particle into the target material.

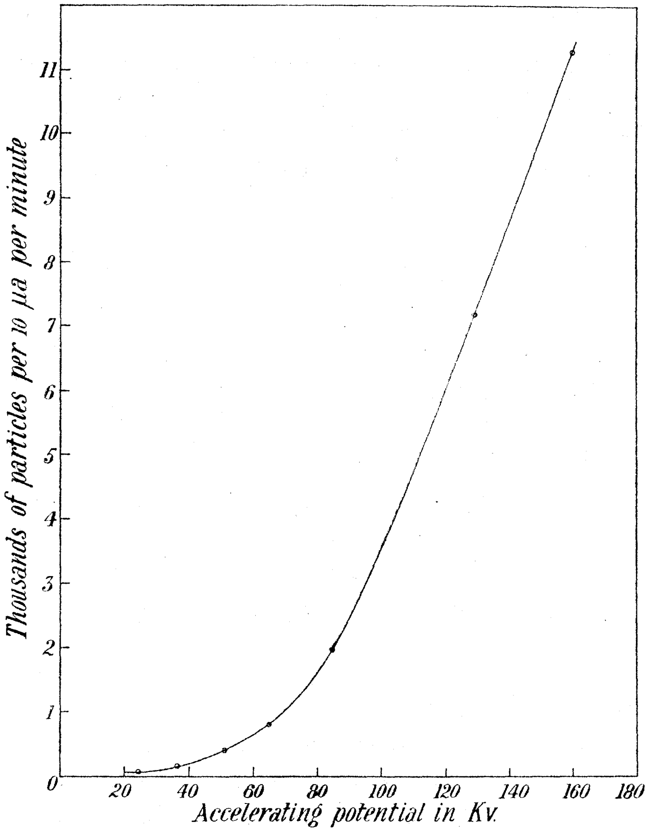

Fig. 3.—$(ND_4)_2SO_4$, 12 cm. absorber.

From the solid angle subtended by the window, and the known composition of the target, we estimate that at 100,000 volts the absolute yield of disintegrations for collisions between D and D is of the order of magnitude of 1 in 10^6. This is a far greater yield than that obtained for any other disintegration process, even at much higher potentials. Simple calculation shows that even a monomolecular layer of diplogen under the conditions of our

* It should be emphasized that the energy available in a collision between two particles of equal mass is only half the energy of the bombarding particle, since half the kinetic energy is retained by the composite nucleus formed by capture.

experiment would give a large effect. Consequently every substance we have bombarded with diplons begins, after a short time, to show effects which are clearly due to traces of diplogen absorbed by or driven into the target. We have also observed that when a compound containing hydrogen, such as ammonium sulphate or lithium hydroxide, is bombarded with a strong beam of diplons, the hydrogen in the compound is gradually replaced in part by diplogen. After some time such compounds begin to behave as diplogen targets of appreciable concentration, and great care must be exercised to separate these effects from any others which are under investigation. This deposition of diplogen from the beam itself, while easily distinguished by the appearance of protons of characteristic range, is much more difficult to allow for when observations are made on the neutron emission of elements. For example, it is difficult to disentangle the emission of neutrons characteristic of lithium and beryllium from the spurious effect due to contamination by diplogen.

The Emission of Neutrons.—We have pointed out that the appearance of the oscillograph records obtained in our initial experiments on the effects produced on bombarding diplogen with diplogen were suggestive of the presence of a strong radiation. The unstable form of He of mass 4·00272, formed by the union of two diplons, might be able to revert to the normal form of He of mass 4·0022 by losing the additional mass as energy of a gamma ray or rays of 23×10^6 e-volts energy. Accordingly we searched for such a radiation with a Geiger-Müller counter. It was at once evident that there was present a very intense radiation capable of producing an undiminished effect on the counter through 20 cm. of lead. As a check on this a search was made for recoil nuclei with the linear counter, and it was found that neutrons are emitted in numbers comparable with the number of 14 cm. protons. It is known that the Geiger counter is affected by neutrons both by the recoil nuclei produced in the counter itself, and also through the action of secondary radiations* produced when neutrons pass through matter. Under these conditions it is impossible for us to decide, on the basis of our experiments, whether a gamma ray of high energy is present. In order to establish the existence of such a radiation it will be necessary to search for high speed photo-electrons, either with the expansion chamber or with a system of coincidence counters.

We have endeavoured to determine the properties of the neutrons by various methods. The absorption in lead has been measured with the linear counter

* *Cf.* Lea, 'Nature,' vol. 133, p. 24 (1934). Experiments on these points have been made with the assistance of Mr. Westcott, who provided the counters, and details will be published elsewhere.

by observation of the number of recoil kicks produced with or without an absorbing screen of 5·5 cm. of lead between the chamber and the target. The average of three measurements showed that the number of kicks was reduced by 44%, corresponding to a radius of cross-section for a collision between a neutron and a lead nucleus of $8 \cdot 4 \times 10^{-13}$ cm. This is to be compared with the value found by Chadwick for the neutrons from beryllium bombarded by polonium α-particles, *i.e.*, 7×10^{-13} cm.* Chadwick states that the cross-section does not appear to change rapidly with the energy of the neutron except when the energy is very low, so that the result tells us nothing except that the neutrons appear to behave in very much the same way as the neutrons from beryllium, which we know to possess a large range of energies.

It is possible to obtain an approximate value for the maximum energy of the neutrons by observation of the maximum size of the kicks produced by recoil nuclei in the linear counting chamber. From the known size of the kicks produced by α-particles, and our knowledge of the energy loss of such particles in traversing the chamber, we can estimate the ionization produced by, and hence the energy of the recoil nuclei. Thus we find that the maximum size of kick produced in the chamber by the recoil nuclei in air is about 6 mm., if we neglect the very large deflections of 2 cm. and over, which almost certainly result from disintegrations. Assuming that the energy loss of the recoil particle per ion produced along its path is the same as for an α-particle, we find an energy for this nucleus of about $0 \cdot 5 \times 10^6$ e-volts. Application of momentum considerations to a head-on collision between a nitrogen nucleus and a neutron leads to the conclusion that the energy of the colliding neutron must be about 2×10^6 e-volts. We have also made experiments on the energy of recoil nuclei produced in helium resulting in a slightly higher value of $2 \cdot 2 \times 10^6$ e-volts. This gas is especially suited for neutron recoil observations as it does not suffer disintegration by neutrons.

In order to account for the production of neutrons of the observed energy and number we have been led to assume the transformation

$$ {}_1D^2 + {}_1D^2 \rightarrow {}_2He^4 \rightarrow {}_2He^3 + {}_0n^1, \qquad (2) $$

in which the unstable ${}_2He^4$ nucleus first formed breaks up into a helium isotope of mass 3 and a neutron. The assumption of the formation of ${}_2He^3$ as a product of such a transformation is not without a precedent, as we have already con-

* 'Proc. Roy. Soc.,' A, vol. 136, p. 693 (1932).

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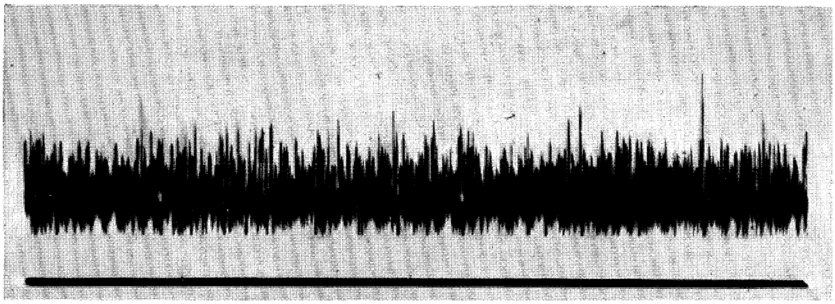

FIG. 4.—First observation of 14 cm protons from D + D.

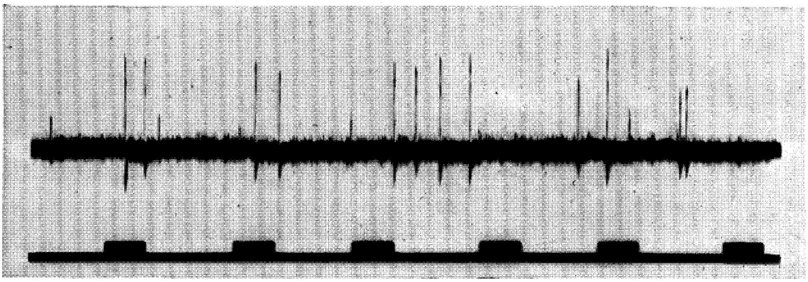

FIG. 5.—14 cm protons from D + D (12 cm absorber).

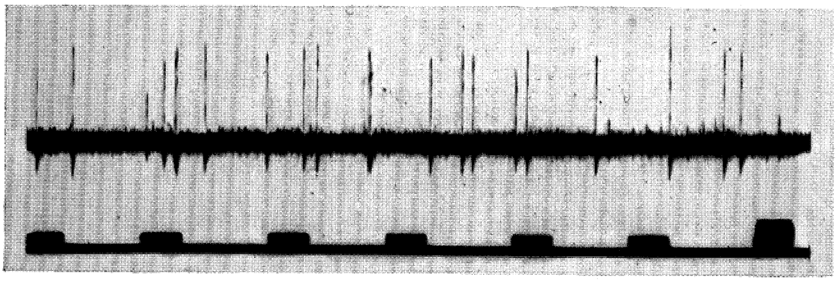

FIG. 6.—$_1H^3$ particles from D + D (1·1 cm absorber).

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cluded that it is produced, together with a normal α-particle, as a result of the bombardment of $_3Li^6$ with protons.* We assumed

$$_3Li^6 + {}_1H^1 \rightarrow {}_4Be^7 \rightarrow {}_2He^3 + {}_2He^4, \tag{3}$$

and found that the observed ranges of 11·5 and 6·8 mm. were in good accord with the application of momentum considerations to this reaction. Dee† has obtained very definite evidence that the two short-range particles are emitted in opposite directions, and we have confirmed that it is the $_3Li^6$ isotope which is concerned in this transformation.‡ It seems quite clear, therefore, that $_2He^3$ can exist under these conditions, and from the observed ranges of the particles we are able to calculate its mass in the following way. The longer range of 11·5 mm. is reasonably well known and it will correspond with the $_2He^3$ particle. For the same velocity an α-particle would go 4/3 times as far because of its greater energy. Hence the velocity of the $_2He^3$ is the same as the velocity of an α-particle the range of which is 1·53 cm., corresponding with an energy of $2·5 \times 10^6$ e-volts. The energy of the $_2He^3$ will be three-quarters of the energy of this α-particle, *i.e.*, $1·88 \times 10^6$ e-volts. The total energy produced in the transformation is thus $3·30 \times 10^6$ e-volts, or the corresponding change in mass is 0·0035 units. Hence the mass of the $_2He^3$ is

$$(6·0145 + 1·0078) - (4·0022 + 0·0035) = 3·0166,$$

using Bainbridge's (*loc. cit.*) value for the mass of Li^6. On the other hand, we can use the mass 6·0157 calculated from the disintegration of $_3Li^6$ into two α-particles of 13·2 cm. range under bombardment by diplons,§ the data for this transformation being extremely good. From this transformation we obtain for $_2He^3$ a mass of 3·0178 units.

Substitution in reaction (3) of the masses calculated in the above manner leads to the appearance of excess masses of

$$(2·0136 + 2·0136) - (3·0166 + 1·0067) = 0·0039,$$

and

$$(2·0136 + 2·0136) - (3·0178 + 1·0067) = 0·0027,$$

respectively, corresponding to energies of $3·6 \times 10^6$ and $2·5 \times 10^6$ e-volts. The neutron would receive three-quarters of this energy, *i.e.*, $2·7 \times 10^6$, or $1·9 \times 10^6$ e-volts. Either of these two values are in good accord with the

* 'Proc. Roy. Soc.,' A, vol. 141, p. 259 (1933).

† 'Nature,' vol. 133, p. 564 (1934).

‡ 'Nature,' vol. 133, p. 377 (1934).

§ Oliphant, Kinsey, and Rutherford, 'Proc. Roy. Soc.,' A, vol. 141, p. 722 (1933).

approximate value of 2×10^6 e-volts found from our experiments, but Dee* has now obtained expansion chamber photographs of the recoil nuclei, analysis of which suggests that the lower of the two figures is more nearly correct, and that the neutrons emitted at right angles to the bombarding beam of diplons are homogeneous in velocity, as required by our reaction.

The recoiling ${}_2He^3$ nucleus possesses an energy of about $0 \cdot 7 \times 10^6$ e-volts, *i.e.*, a range of 5–6 mm. as a maximum.† The thinnest mica window we have been able to use has a stopping power of 3 mm. of air, and the residual range of 2–3 mm. is not sufficient for the particle to enter our counting chamber and produce a deflection of appreciable size. We have searched very carefully for such a doubly charged particle, both with a special type of counting chamber of small depth, and by looking for scintillations produced on a screen of zinc sulphide placed inside the apparatus itself and covered with aluminium of 2 mm. stopping power to prevent light and scattered diplons from reaching it. In both cases the presence of a very intense radiation which is strongly absorbed in a fraction of a centimetre of air, gave rise to so much disturbance as to render counting impossible. Thus, while we have not yet detected the ${}_2He^3$ particles which we believe to be present, we have not yet obtained any evidence that they do not exist.

No evidence of the existence of an ${}_2He^3$ isotope has been obtained by ordinary methods, although the possibility of its existence has been suggested at various times. It is not unlikely that while the new isotope may prove to be unstable over long periods it may yet have a sufficiently long life to be detected by counting methods and in the expansion chamber. We have not detected any after-effects lasting for a few seconds or more, suggesting the expulsion of a positive electron or other charged particle. If the ${}_2He^3$ nucleus is unstable, there are a number of possibilities as to the mode of transformation.

It is evident that the experiments we have described suggest very strongly that the neutrons resulting from the bombardment of diplogen with diplogen are homogeneous in velocity, and since large yields are obtainable at comparatively low bombarding potentials they should serve as an almost ideal group for experimental work on the properties of neutrons.

In addition to a study of these transformations, we have made a number of observations on the neutrons, protons, and also α-particles emitted from

* Not yet published.

† The energy and range of the particle depends to some extent on the direction of emission relative to the bombarding particle, and will be greater in the forward direction.

lithium, beryllium, carbon, and other elements under diplon bombardment. We hope to give an account of these experiments in a subsequent paper.

In conclusion, we have to express our thanks for help with some of the experiments to Mr. Kempton and Mr. Westcott, and we acknowledge our indebtedness to Mr. G. R. Crowe for his technical assistance throughout. Dr. P. Harteck is indebted to the Rockefeller Foundation for a grant.

Summary

An account is given of the effects observed when diplons are used to bombard targets of compounds containing heavy hydrogen. It is found that a group of protons of 14·3 cm. range is emitted in very large numbers. A shorter 1·6 cm. range group of singly charged particles is also observed, and it is shown that the two groups contain equal numbers of particles. A discussion of the reaction which gives rise to them is given, and reasons are advanced for supposing that the short-range group consists of nuclei of a new isotope of hydrogen of mass 3·0151. The number of particles emitted has been investigated as a function of the energy of the bombarding diplons, and the absolute yield for a pure diplon beam hitting a pure diplogen target is estimated to be about 1 in 10^6 at 100,000 volts.

Neutrons have been observed in large numbers as a result of the same bombardment. It is shown that the energy of the neutrons is about 2×10^6 e-volts, and it is suggested that they arise from an alternative mode of breaking up of the unstable form of helium nucleus formed initially by the union of two diplons. This other mode results in the expulsion of a neutron and a helium isotope of mass 3 in directions opposite to one another. If we calculate the mass of $_2He^3$ from energy and momentum considerations of the ranges of the short-range groups emitted from $_3Li^6$ when bombarded by protons, the energy of the neutron can be deduced and agrees well with experiment.

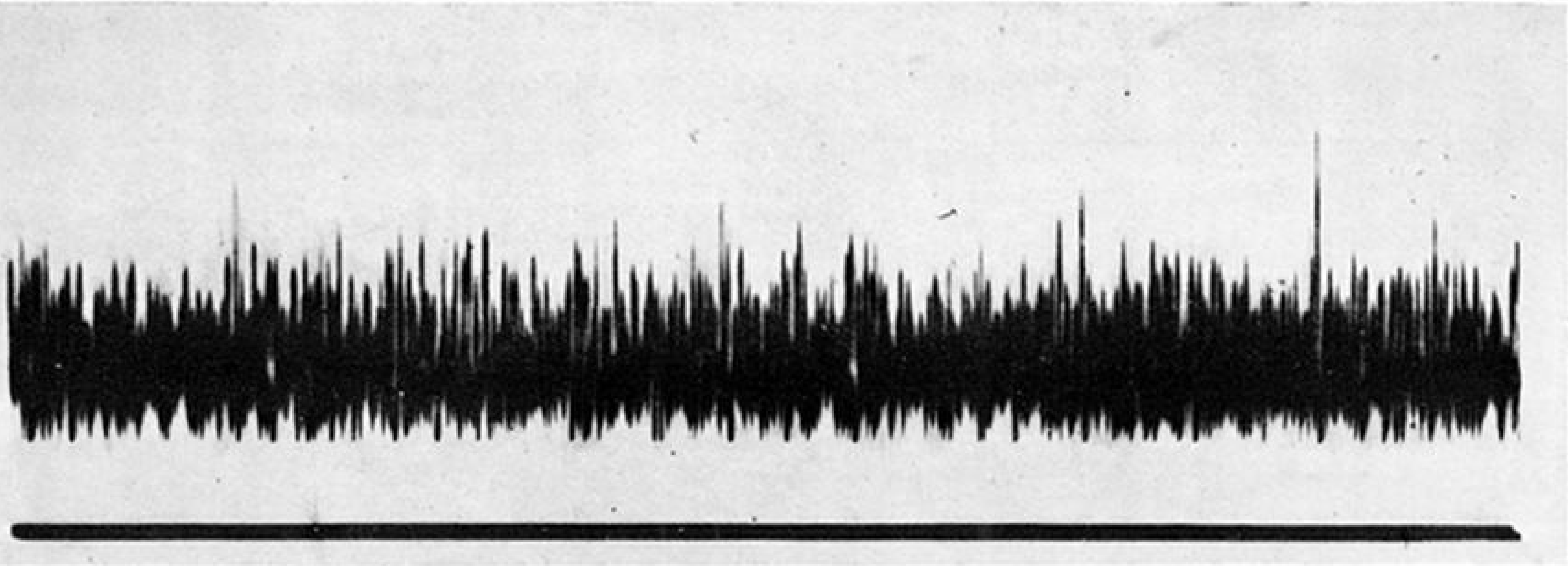

FIG. 4.—First observation of 14 cm protons from D + D.

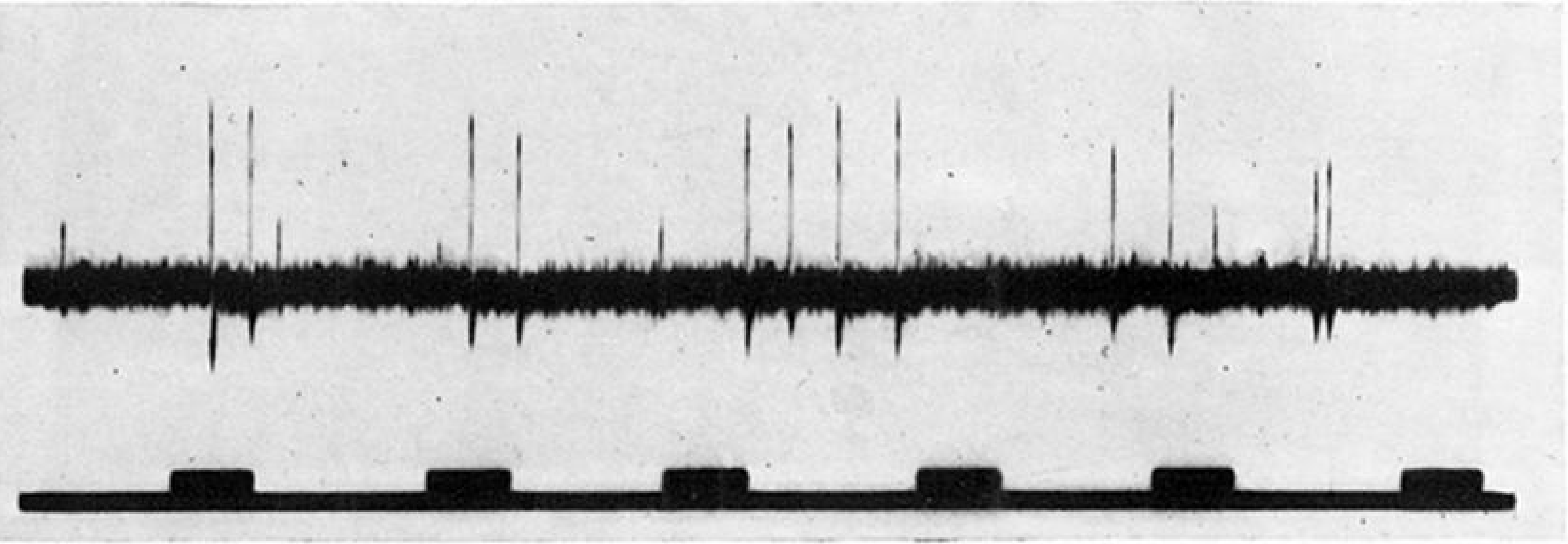

FIG. 5.—14 cm protons from D + D (12 cm absorber).

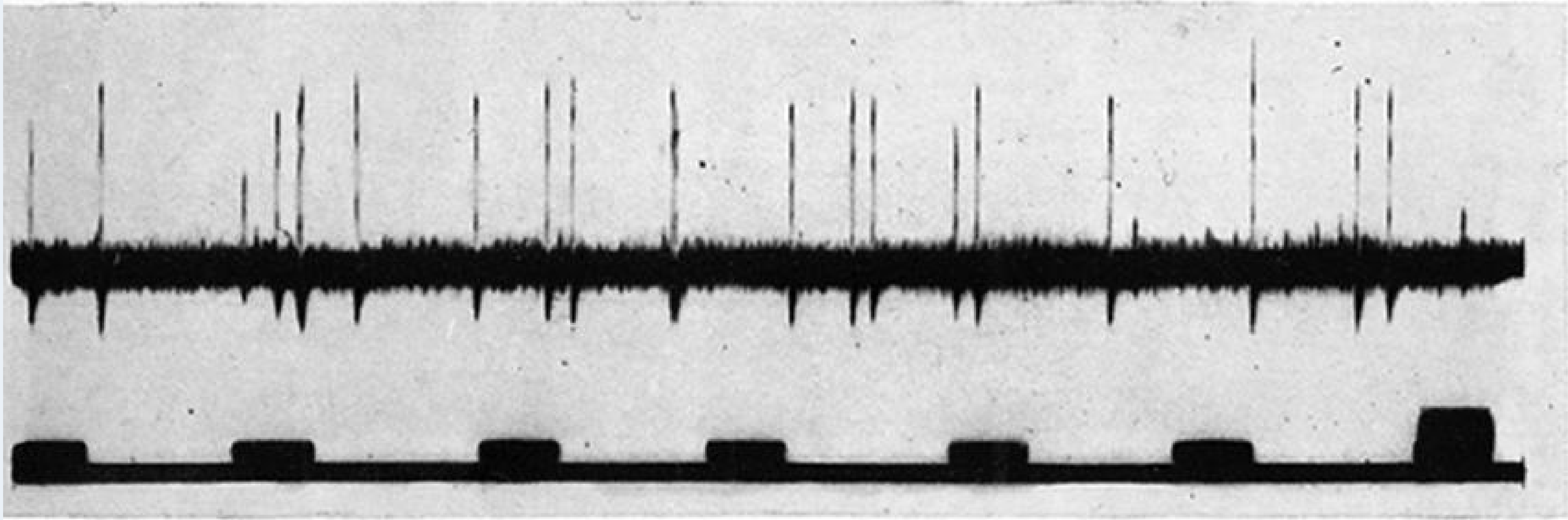

FIG. 6.—${}_1H^3$ particles from $D + D$ (1·1 cm absorber).