Disintegration of Nitrogen and Boron and Possible Emission of Deuterons

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An account is given of the technique of detecting nuclear particles by the use of a proportional counter, and its advantages and limitations are discussed. The counter was used to investigate the disintegration of nitrogen and boron by alpha-particles. In the case of nitrogen no group which could be ascribed to deuterons was found while for boron the presence of a short range group of about the expected energy, first found by Heidenreich, was confirmed. The values of the magnetic deflections of particles from both elements were compared with those for known protons and deuterons of the same velocity and shown to be protons in each case. Further evidence for resonance was found in the experiments on nitrogen and the work of Heidenreich and Paton confirmed in the experiments on boron.

I. INTRODUCTION

At the present time three electrical methods of detection of nuclear particles are in use, namely, electrometer detection, linear amplifier, and proportional (Geiger-Klemperer) counter. The technique of the last method has not been fully developed and in the first part of this paper an account is given of the experimental arrangement and properties of a proportional counter suitable for detection of protons or alpha-particles produced in nuclear transmutations.

The Geiger-Klemperer counter[1] utilizes ionization by collision to multiply the initial ionization produced by a nuclear particle in a gas. It is operated with a negative voltage on the case surrounding a small ball, the voltage being just lower than that which is necessary for a momentary discharge if ions are produced near the ball. Under these circumstances the negative ions on their way to the ball produce further ions by collision, the multiplication of ions in this way being of the order of 1000. Such a counter has advantages for two reasons: First, the rapidity of collection of the ions renders it suitable for use with a high frequency oscillograph and hence for the detections of protons in the presence of γ-rays; second, the multiplication of ions gives a larger voltage rise to the ball than to the collecting electrode of a linear amplifier so that fewer stages of amplification by tubes are needed. This means the instrument is comparatively free from electrical disturbances which in crowded research laboratories are prime factors in slowing up the rate of work. Since the initial ionization is linearly multiplied, heavy charged particles can be detected in the presence of β, γ and x-rays.

II. EXPERIMENTAL ARRANGEMENT

The experimental arrangement for using a proportional counter has been described by several workers[1, 2] by whom it was developed for the purpose of counting in the presence of β and γ-rays. The general procedure is to detect the impulses with an oscillograph after amplification by a tube amplifier whose gain depends on the multiplication factor of the counter and the sensitivity of the oscillograph. With the exception of the input tube any standard design of amplifier giving proportional amplification is suitable (if counting in the presence of β and γ-rays is required the coupling between two of the tubes should be through a small condenser to give distortion favoring sharp impulses).[3] The amplifier here described has proved satisfactory and the details will be given briefly later on. The important points of construction are in the counter design and the input tube. Details of these follow.

The counter consists of a brass cylinder roughly 8 cm long and 4 cm diameter, closed at the end by a disk with a hole of about 2 cm diameter. A fine platinum wire ending in a small

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1 H. Geiger and O. Klemperer, Zeits. f. Physik **49**, 753 (1928).

2 H. Fränz, Zeits. f. Physik **63**, 370 (1930); H. Klarmann, Zeits. f. Physik **87**, 411 (1934); W. E. Duncanson and H. Miller, Proc. Roy. Soc. **A146**, 413 (1934).

3 C. E. Wynn-Williams and F. A. B. Ward, Proc. Roy. Soc. **A131**, 391 (1931).

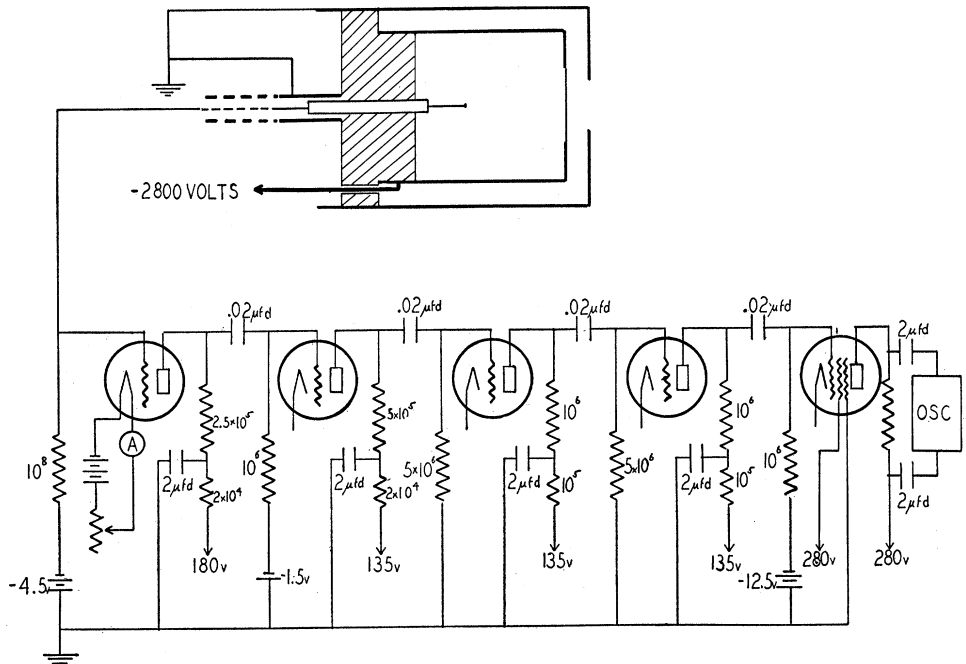

FIG. 1. Electric circuit of counter and amplifier.

ball is soldered into the end of a brass rod, and is placed axially in the cylinder, insulated from it by an ebonite plug. A guard ring is not necessary. The brass rod is connected by a shielded cable to the grid of a triode. Surrounding the counter case is a grounded brass cylinder whose end can be covered by copper disks with holes of various sizes; this grounded cylinder is not absolutely necessary but helps to reduce electrical disturbances. A high potential source which was either a bank of dry batteries or a smoothed transformer-rectifier device (designed by Dr. C. D. Bock of this laboratory) has the negative end connected to the counter case, the positive being grounded. A 2 microfarad condenser and resistance is placed across the battery as indicated in Fig. 1, which gives the complete connections of the counter and amplifier. The first tube is placed in a small copper box while the remainder of the amplifier is mounted on a small table on wheels—everything being cased in copper, with a shielded cable leading from the table to the first tube box. The second tube and connections are shielded in a separate copper case.

The ball used in the counter was made by melting fine platinum wire (about 1/20 mm in diameter) in an oxygen-gas flame. The ball is then clean and needs no further preparation. Several times during the work the counter began to give spontaneous discharges which were removed by reheating the ball in an ordinary blowpipe flame. The larger the ball the less likely the occurrence. The voltage at which the counter operates depends critically on the size of the ball. For a ball of diameter 100μ the critical voltage range lies in the region 1800–1900; of half a mm, in the region 2500–3000. In the first case, the sensitive voltage range covers about 30 volts; in the second case about 200 volts. These values apply to atmospheric pressure.

With a large counter opening (2 cm in diameter), the number of particles counted from a proton beam containing particles of all energies depends sharply on the voltage which must be kept perfectly steady. This is not so important with openings of less than 1 cm diameter but the exacting nature of this condition is the principal disadvantage of this method of counting. We found dry batteries sufficiently steady for experi-

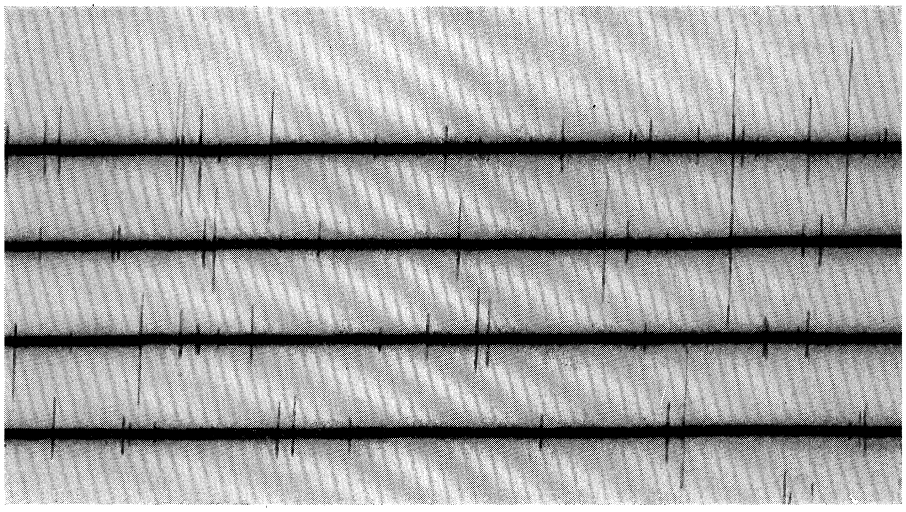

FIG. 2. Sample photographic record of oscillograph.

ments lasting a few days. The reason for the variation in numbers lies in the sensitivity of detection. In a completely inhomogeneous beam there is no lower limit to the size of a deflection since there is no lower limit to the ionization produced by each particle; thus conditions must be kept perfectly steady if consistent counts are to be obtained. It is therefore also important to keep the heater current of the first tube constant; this was watched with an open scale ammeter and kept at a fixed value with a good rheostat.

The counter opening could be made effective over the whole of 2 cm diameter by using a half mm ball set 4 cm from the end of the counter and keeping the voltage within 25 volts of that at which individual β-rays could be detected. It is doubtful if the particles entering at the edge of so large an opening could be detected in the face of heavy γ-radiation.

The first tube of the amplifier was of the type 76. This was chosen since it is a heater triode operating with a relatively low anode resistance (which reduces possible electrical pick-up where a long lead has to be used to connect to the next stage). Heater triodes are much less sensitive to mechanical and acoustic disturbances. The grid leak was 10^8 ohms made from ten 10-megohm resistors: a grid bias of -4.5 volts was used. The ratio of deflection to background is very sensitive to the heater current and this must consequently be adjusted carefully to give uniform results; the current depends on the grid leak and on the anode resistance, and the optimum value can be found only by trial and error.

The remainder of the amplifier is sufficiently described by Fig. 1. The second tube was type 75; third and fourth 240; fifth 41.[4] The counter can be made to record on the oscillograph if the third and fourth tubes are omitted providing the instrument's sensitivity is much lower (proportional to 1/frequency^2). For higher frequencies greater amplification is needed. The ratio of deflection to electrical disturbances is about the same for high and low frequencies. The oscillograph is of the type described by Wynn-Williams[3] which is easily constructed and reasonably simple to use. The deflections of the light spot were recorded photographically. The camera is of the rotating drum type, consisting of a brass drum about four inches in diameter rotating inside a brass cylinder on one side of which is a long adjustable slit whose width can be varied between 5/1000 and 100/1000 inch. The drum carries photographic paper (Eastman "News" Bromide) of width 8 inches (12-inch width can be taken if necessary) and is driven by a friction drive between a brass disk attached to the drum and a leather rimmed wheel fixed to a telechron

[4] For the general features of amplifier design see J. R. Dunning, Rev. Sci. Inst. **5**, 387 (1934).

synchronous motor. The rate of rotation can be varied with ease and the synchronous motor does away with the need of a time scale. The whole is placed on wheels and moves along rails, being driven slowly across the light spot by a rack and gear wheel, thus producing a helical trace on the paper wound around the rotating drum. We find that this type of recording is much more convenient than the use of strip film or paper and is much less expensive both in the initial cost of the camera and the running cost of the paper. Some of the records are shown in Fig. 2.

III. Emission of Deuterons in Nuclear Transmutations

The possibility of the emission of deuterons in nuclear reactions has been suggested by F. Perrin[5] who proposes in particular the existence of a group of about 7-cm range when polonium alpha-particles bombard nitrogen, the suggested reaction being

$$_2He^4+{}_7N^{14}={}_8O^{16}+{}_1D^2.$$

There is a general improbability of the detection of deuterons in all such reactions since the fractional part of the mass of the deuteron is so high that its formation requires considerable energy which must come either from the energy difference between the bombarded nucleus and product nucleus, or from the incident alpha-particle. Thus if we have a general reaction,

$$_2He^4+A=B+{}_1D^2$$

the difference of mass $_2He^4-{}_1D^2$ is almost always less than the difference of mass $B-A$, so that outside energy is needed to liberate the deuteron. In the nitrogen reaction $_2He^4-{}_1D^2$ is roughly 1.989 while the difference $_8O^{16}-{}_7N^{14}$ is 1.992 so that an alpha-particle bringing energy greater than 0.003 mass unit can liberate a deuteron. A polonium alpha-particle of range 3.9 cm has an energy of 0.005 mass unit and so has sufficient energy. Accurate calculation assuming the masses: $_2He^4=4.00216$; $_7N^{14}=14.0080$; $_8O^{16}=16.0000$; $_1D^2=2.01363$ gives a group of deuterons of range about 5 cm. F. Perrin uses the mass $_1D^2=2.0115$ and estimates 7 cm. Pollard[6] has found a group of particles of range 6.6 cm when nitrogen is bombarded by alpha-particles, but considers they are protons produced by resonance. An inspection of the various possible reactions taking place under alpha-particle bombardment shows the reaction:

$$_2He^4+{}_5B^{10}={}_6C^{12}+{}_1D^2$$

to be the one giving the greatest possible energy to the deuteron. The calculated range assuming the masses $_5B^{10}=10.0135$ and $_6C^{12}=12.0036$ is about 16 cm. A group of particles of range 14 cm has been found by Heidenreich.[7] This group could therefore be explained as being deuterons. It is of interest that in both these reactions the angular momentum would be conserved and hence on Goldhaber's[8] suggestion they should be classed as "probable." Other nuclear reactions do not give sufficient energies to the deuteron to make its detection feasible.

IV. The Reaction $_2He^4+{}_7N^{14}={}_8O^{16}+{}_1D^2$

The products from the bombardment of a layer of nitrogen by polonium alpha-particles were first examined for the presence of a short range group, not due to resonance, produced by full energy alpha-particles. The nitrogen (air) was placed in a tube whose end was closed by a gold foil of air equivalent about 9 mm and the whole placed about 2 cm distant from the opening of the counter. By moving the source relative to the foil, different thicknesses of air could be bombarded. With the source near the foil alpha-particles penetrating it could bombard and possibly disintegrate nitrogen nuclei in the air outside. By placing screens of aluminum between counter and foil the yield of the disintegration particles could be plotted against absorption in their path. Such curves are shown for three positions of the source, namely, 3.0, 1.4 and 0.8 cm from the foil (Fig. 3). In the first case a group of 6.5 cm is weakly shown; in the second this group is entirely absent; in the third it appears strongly. The explanation of this group is therefore that it is due to resonance at alpha-particle ranges of about 2.2 cm, which are effective in the

[5] F. Perrin, Comptes rendus **194**, 2211 (1932).
[6] E. Pollard, Proc. Roy. Soc. **A141**, 377 (1933).
[7] F. Heidenreich, Zeits. f. Physik **86**, 675 (1933).
[8] M. Goldhaber, Proc. Camb. Phil. Soc. **30**, 561 (1934).

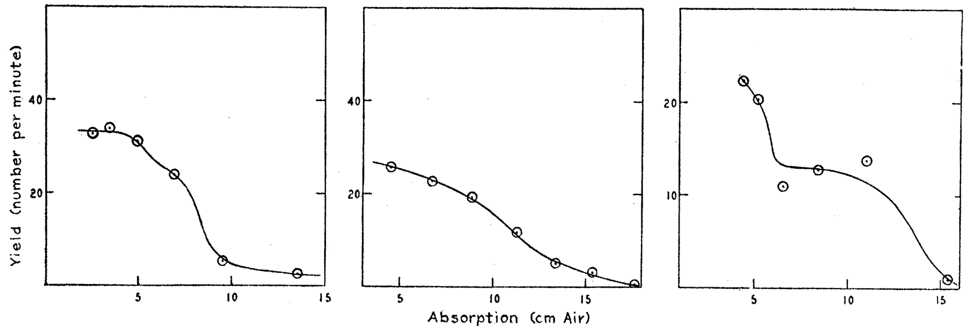

FIG. 3. Range distribution of disintegration particles from nitrogen bombarded by polonium alpha-particles.

layer near the counter in the third case, ineffective in the gold foil in the second, and effective inside the tube in the first case. The geometrical conditions explain the variation in definiteness of the group since it is greatly favored in the third case, by being produced so near the counter. The rapid fall at range 8 cm in the first experiment is due to this reason also, since the longer range protons are produced near the source and hence have a smaller solid angle for entering the counter. From the first two curves it is possible to deduce that the maximum width of the resonance level extends from 1.7 cm to 2.4 cm alpha-particle range and that its yield is 40 percent of the main group which is known to be excited by particles of ranges 3.0 cm to the full range of 3.87 cm.[6] These experiments therefore show no short range group which is not explainable by resonance with alpha-particles of range 2.2 cm. Any group of deuterons produced by alpha-particles of range 2.2 cm would have a range too small to detect and hence the emission of deuterons is unlikely. A more direct test was made (as described later) in which the magnetic deflection of the particles was investigated.

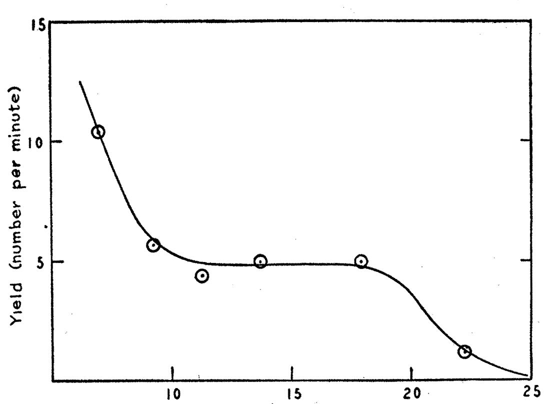

FIG. 4. Range distribution of disintegration particles from boron bombarded by polonium alpha-particles.

V. THE REACTION $_5B^{10}+_2He^4=_6C^{12}+_1D^2$

The experiments of Heidenreich[7] showed the existence of a group of range 14 cm when full range polonium alpha-particles bombard boron. To verify this a layer of boron was deposited on gold foil from a suspension in alcohol and placed at the end of a glass tube through which oxygen could be passed. The source was placed 6 mm from the layer and the counter was brought up near the outside of the gold foil. As in the previous experiment the interposition of aluminum screens enabled the yield *versus* range curves to be plotted. The resulting curve is shown in Fig. 4. The presence of two groups is clearly shown, one of range 11 cm, the other of range 25 cm. Since the maximum range of the alpha-particles impinging on the target is here 3.3 cm the ranges of the groups would be expected to be less than those found by Heidenreich. The 25 cm group corresponds to a nuclear energy change which was first found by Bothe and has been confirmed by several workers.[9] The products of total range less than 16 cm were therefore examined by magnetic deflection to see if they were deuterons.

[9] See H. Miller, W. E. Duncanson and A. N. May, Proc. Camb. Phil. Soc. **30**, 549 (1934).

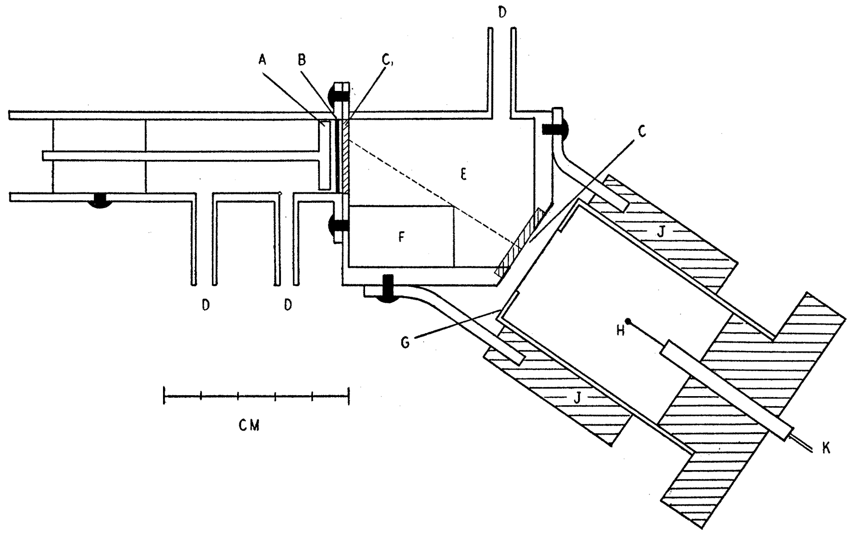

FIG. 5. Arrangement of magnetic deflection apparatus.

VI. Magnetic Deflection Apparatus

The early experiments of Rutherford and Chadwick[10] showed that the magnetic deflection of particles resulting from alpha-particle impacts with four elements (N, F, Al, P) was consistent with their being protons. These experiments were made before the knowledge of the existence of groups of particles of different velocities and hence would not decide whether a particular group is deuterons or protons. Moreover, boron was not investigated. In designing a method for observing magnetic deflection the principle of Rutherford's method was followed, with the refinements of electrical counting and of isolation of a limited velocity band for deflection. In the case of nitrogen we made the added improvement of placing the bombarded gas outside the space in which deflection occurred. The arrangement of apparatus is shown in Fig. 5. E is an evacuated flat brass box about an inch thick, with windows at C_1 and C_2 covered by thin foils of aluminum of about 2 cm air equivalent. The aluminum was supported by thin strips of brass let endwise into the box as indicated. A brass block, F, which just fitted inside the box was placed as shown so that a straight line from the center of the right-hand window past the tip of F intersects the left-hand window about a quarter of its width from the top. This is shown by a dotted line in the figure. At B could be placed a solid target such as thin paper, $Ca(OD)_2$, or powdered boron, which was bombarded by the alpha-particles from the source at A. Or else a gas could be circulated between the source and the window by means of the tubes DD. As a result of the disintegrations being produced in the target, particles moving in all directions enter the space E across which magnetic fields up to 13,700 gauss can be applied. With no field, a relatively small number of particles can pass through the right-hand window and reach the counter G, which is supported in an ebonite mounting J. A field which bends the paths of the particles downwards (designated as a positive field) increases the number striking the window until a maximum is reached when any further increase in field causes as many particles to be deflected past the window as are deflected into it. The position of this maximum cannot be simply calculated from the ill-defined geometrical conditions, but comparison with experiments using known protons in a given velocity range showed it to be clearly marked. A negative field of course deflects the particles away from the window C_2. The distance FC_2 was made about one-half FC_1, this being the ratio of the areas of the window openings. The results of applying

[10] Rutherford, Proc. Roy. Soc. **A97**, 374 (1920); Rutherford and J. Chadwick, Phil. Mag. **44**, 417 (1922).

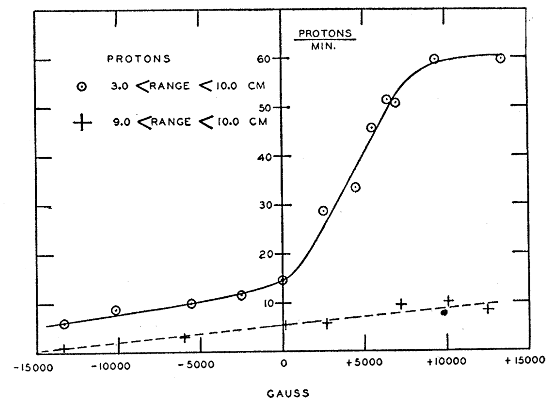

FIG. 6. Results of applying different fields to known protons from onion skin paper.

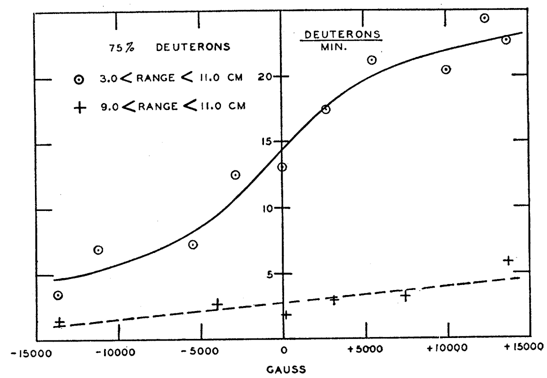

FIG. 7. Effect of applying different fields to a beam of 75 percent deuterons.

different fields to known protons from onion skin paper are shown as circles in Fig. 6.

In order to isolate reasonably narrow velocity bands use was made of the rapid variation of range with velocity. With the apparatus as in Fig. 4 a particle could be detected with a range (after entering the field) of 3 cm. The velocity corresponding to this is 1.5×10^9 cm/sec. for a proton, or 1.2×10^9 cm/sec. for a deuteron.[11] If a screen of absorption 6 cm air equivalent is placed between C_2 and the counter the minimum detected range is 9 cm corresponding to a proton velocity of 2.1×10^9 cm/sec. of a deuteron velocity of 1.7×10^9 cm/sec. Then if curves are plotted for the difference between the counts with and without screen a velocity band of $1.8\pm0.3\times10^9$ cm/sec. is chosen for protons and $1.45\pm0.25\times10^9$ cm/sec. for deuterons. Although for a given range, deuterons have only 80 percent the velocity of protons, the ratio of the masses makes the radius of curvature of the deuteron paths 1.6 times that of the proton paths. Thus for a given range group of particles it is possible to distinguish clearly between protons and deuterons. In Fig. 6, the crosses give the curve for protons of range greater than 9 cm while in Fig. 7, similar curves are given for a beam of 75 percent deuterons produced by bombarding a layer of $Ca(OD)_2+Ca(OH)_2$ with Po alpha-particles. That the protons are deflected more easily as the field ranges from high negative to high positive values is clearly shown by a comparison of the curves.

It is of interest that if the maximum range of particles greater than 9 cm range is about 25 cm then the highest velocity present is 2.8×10^9 cm/sec. for protons or 2.4×10^9 cm/sec. for deuterons. Hence these particles also lie in a reasonably defined velocity band, namely, $2.45\pm0.35\times10^9$ cm/sec. for protons and 2.05 ± 0.35 for deuterons.

The method of investigation therefore consists in placing the layer of element to be disintegrated at B and observing whether the difference between the yields of emitted particles for 3.0 and 9.0 cm range varies with the field like protons or deuterons as deduced from Figs. 6 and 7.

Results for boron

A layer of boron of about 2 cm air equivalent was deposited on gold foil of 0.9 cm air equivalent by spraying a suspension in absolute alcohol. This gives rather more uniform layers than deposition from suspension. The absorption when no screen was placed before the counter was 8.3 cm so that the short group was detected. The difference between the yields with and without screen was found for six values of the field, the accuracy being greater for the three points with positive field. The values are shown as circles in Fig. 8 with known proton and deuteron curves for comparison. It will be seen that the fit is much closer with the proton curve than with the one for deuterons. The high values for negative

[11] W. E. Duncanson, Proc. Camb. Phil. Soc. **30**, 110 (1934).

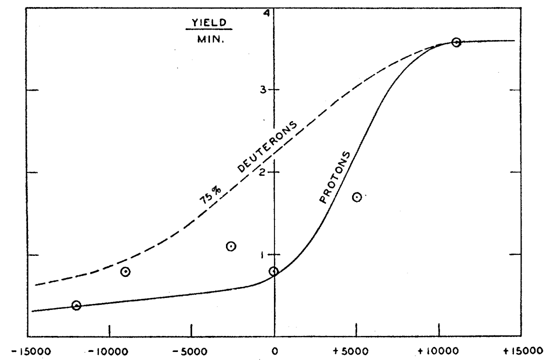

FIG. 8. The circles show the difference in yield of disintegration products from boron with and without absorbing screen for different field strengths.

fields are probably accounted for in part by fluctuations and also by the fact that the beam from boron contained more protons near the upper limit of velocity than the comparison beam. Even if these suggestions are discounted it is possible to say that no more than 10 percent of the particles counted can have been deuterons. Since natural protons were known to be present in amounts roughly 50 percent of the boron particles this means that only one-sixth of the boron group can have been deuterons. The explanation of the short group must therefore be in terms of protons.

While these experiments were in progress Paton has shown[12] that under bombardment by 8.6 alpha-particles boron emits particles of range 25.5 cm in a direction at right angles to the alpha-particles. The corresponding range for this group if produced by 3.3 cm particles along their original direction would be 11.7 cm, which is in good agreement with the value here found. The value found for the nuclear energy change is -1.68 mev which compares with -1.86 given by Paton and -1.76 calculated from Heidenreich's data. Thus while the results of these three separate experiments agree with one another providing the group is explained as being protons, the deuteron hypothesis applied to Paton's findings leads to a group of 24 cm range under the conditions of our experiment which is widely different from the 11 cm as found; or to 28 cm for full range alpha-particles which again differs from the value of 14 cm found by Heidenreich.

It is of interest that the particles of range sufficient to penetrate the screen and which, as explained previously, fall in the velocity range $2.45 \pm 0.35 \times 10^9$ cm/sec. if protons or $2.05 \pm 0.35 \times 10^9$ cm/sec. if deuterons are deflected in the same way as protons. Hence the group whose nuclear energy change is $+0.2$ mev can also be said to consist of protons.

Results for nitrogen

To test the particles emitted from the bombardment of nitrogen the source was placed 2 cm from the window so that the resonance group was produced, the gold foil being removed to reduce the total absorption in the path of the particles to less than 6 cm. The nitrogen in the air served as target, and a yield curve was plotted. Since the particles were known to have a maximum range of less than 18.5 cm a screen was not interposed, the velocity band being already reasonably narrow. For a comparison curve illuminating gas (containing a high proportion of hydrogen) was passed through the space in front of the source and a yield curve again plotted against various values of the field. It is to be expected that these particles will have an average velocity 7 percent less than the nitrogen particles (since their distribution with range is towards lower energies, and their maximum range is less) and hence should be bent slightly more easily. The two curves are shown in Fig. 9. It will be seen that there is reasonable agreement, allowing for experimental

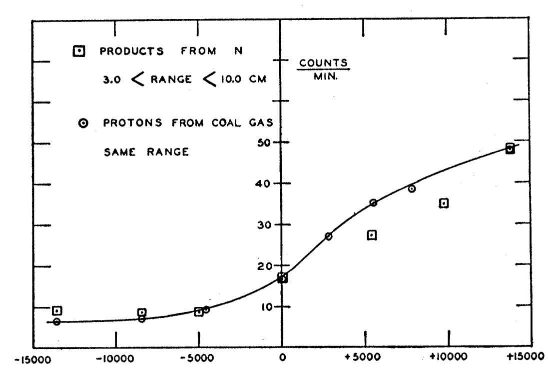

FIG. 9. The squares show the difference in yield of disintegration products from nitrogen for various field strengths.

[12] R. F. Paton, Zeits. f. Physik **90**, 586 (1934).

error (about 100 particles only were counted at each point for the nitrogen curve). We do not consider that this curve alone is sufficient to disprove the presence of deuterons but that taken with the fact that the expected short range group for bombardment by full range alpha-particles is absent, it enables a final conclusion to be drawn in favor of protons.

It may be pointed out that the difference between the proton curve and that obtained by bombarding $Ca(OD)_2$ is direct evidence that the projected particles are deuterons, as concluded from measurements of their range by Rutherford and Kempton.[13]

In conclusion we wish to express our thanks to Professor A. F. Kovarik for his interest and advice, to Dr. C. T. Lane for advice in running the magnet, to Dr. Donald Cooksey for assistance in the counter design, and to Professor H. C. Urey for a gift of heavy water.

[13] Rutherford and A. E. Kempton, Proc. Roy. Soc. **A143**, 724 (1934).

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Nuclear Shells: Angular and Magnetic Momenta of Nuclei

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Part I. Angular momenta. The experimental data on the number of isotopes per atom show marked regularities, which suggest closed shells in the nucleus. These regularities have been rigorously followed in arranging the first thirty elements into an isotopic system, with proton and neutron shells. In order to correlate both the angular and magnetic momenta of nuclei, it is necessary to choose certain j values of the terms arising from the proton and neutron configurations, as the deepest terms. After this choice has been made, the assumption, that the j values of the lowest terms are added vectorially with the j's oppositely directed, makes it possible to account for all of the observed i values. Apparently the S states are exceptions to the general rule and have their j's in the same direction.

Part II. Magnetic momenta. Nuclear magnetic moments are discussed from the viewpoint of proton and neutron shells in the nucleus. The generalized g-formula is used to calculate the proton and neutron contributions to the g-factor. These two contributions are then combined by jj coupling to give the nuclear g values. A magnetic moment of $+2.7$ nuclear magnetons for the proton, and $+1.75$ nuclear magnetons for the neutron are used in the calculations. The deepest proton and neutron terms fix the values of l, j and s, so that, once having chosen these deepest terms, no arbitrariness exists in the calculations. A discussion of the correlated data is given.

PART I

FORTY-EIGHT elements have been found to have a nuclear spin greater than zero. Three (He^4, C^{12} and O^{16}) are known to have zero spin. Twenty of these elements, about which definite information is known, are among the first thirty elements of the periodic table. For this reason, an attempt to correlate proton and neutron shells with nuclear momenta has been made.

In addition to the known nuclear "i" values, the number of known isotopes per atom provide valuable information which can be used as a guide for deciding where nuclear shells are filled. Isotopic regularities are nicely shown by the Chart of Isotopes which has been compiled by Bartlett.[1] Part of those data which are pertinent to the present discussion is given in Table I. The elements are listed in the first column, the number of known isotopes in the second, and the number of expected isotopes in the third. Numbers larger than one mean that that number of isotopes occur with consecutive mass numbers. Where the notation 1, 1 or 1, 1, 1 (Cl, A) is used, each comma denotes a missing mass number between known mass numbers for that element.

Table I suggests[2] that one isotopic regularity

[1] Bartlett, Rev. Sci. Inst. **6**, 61 (1935).
[2] Bartlett (Nature **130**, 165 (1932)) has suggested shells similar to these.

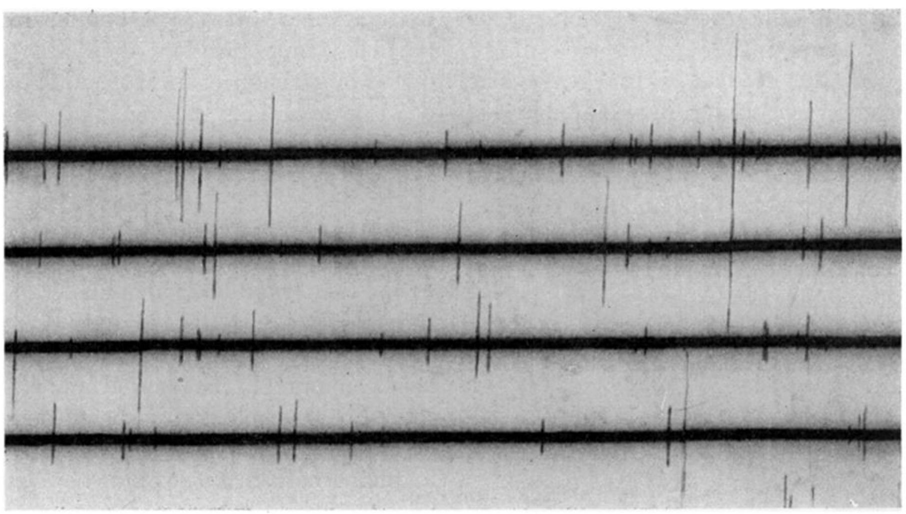

Fig. 2. Sample photographic record of oscillograph.