on the high voltage side as fast as would be expected for an important resonance, but the estimated error of 25 percent exclusive of systematic errors might be large enough to conceal such a trend. However since it seems sure that any large errors in the cross section would be in the direction of underestimation, such an explanation does not seem probable.

On the other hand, the unusually high value found for the cross section introduces another question. On the assumption that the Coulomb radius of the beryllium nucleus is  $5 \times 10^{-13}$  the geometric cross section of the nucleus is approximately the same as the disintegration cross section. This suggests that protons having L>0may be taking part in the reaction. Since protons of L=1 would not contribute to the yield at 90°, even higher order collisions would be called for. These would imply unsymmetrical distribution of the resulting  $\alpha$ -particles, with probably a minimum distribution at 90°. No such unsymmetrical distribution has been reported, so that the interpretation is very questionable.

It is hoped that further work in this laboratory on the problem of angular distribution will clarify this matter.

## Acknowledgments

This work was carried out under the general direction of Professor S. K. Allison. Construction of the apparatus and the taking of the data were made possible only by the invaluable assistance of Professor Allison, Mr. L. S. Skaggs, and Mr. Nicholas M. Smith, Jr.

## AUGUST 1, 1938

## PHYSICAL REVIEW

VOLUME 54

## A Precise Measurement of the Energy Change in the Transmutation of Beryllium into Lithium by Proton Bombardment

SAMUEL K. ALLISON, LESTER S. SKAGGS AND NICHOLAS M. SMITH, JR. University of Chicago, Chicago, Illinois (Received May 27, 1938)

The very high yield of alpha-particles from the reaction  ${}_{4}\text{Be}{}^{9}(p,\alpha){}_{3}\text{Li}{}^{6}$  has made it possible to measure their energies by deviating a beam through 90° in an electrostatic field. A steady source of high voltage, which could be measured to 0.1 percent up to 40,000 volts, was used to deflect the alpha-particles. Two experiments are reported in which the beryllium was bombarded by 320 kv and 383 kv protons, respectively. The energy distribution curves of the alphas show a sharp high energy limit, and this limit

THE nuclear transformations induced in beryllium by proton bombardment have been the subject of several investigations since the first extended report by Oliphant, Kempton, and Rutherford<sup>1</sup> appeared. In this report previous experiments and exploratory work by Cockcroft, Dee, Döpel, Kirchner, etc. are mentioned. More recently papers<sup>2</sup> by Döpel, Kirchner increases with bombarding energy of the protons. The experiment shows that the energy balance of the reaction is  $2.152\pm0.04$  Mev, corresponding to  $2.310\pm0.04\times10^{-3}$  atomic weight unit. This establishes the difference in mass between Be<sup>9</sup> and Li<sup>6</sup> as  $2.99804\pm0.00009$ . If we assume that of the two, the mass of Li<sup>6</sup> is more accurately known, and adopt Livingston and Bethe's value of 6.01686  $\pm 0.00020$ , the experiment shows that the mass of Be<sup>9</sup> is 9.01491 $\pm 0.00025$ .

and Neuert, Zipprich, Allen, and Williams, Haxby, and Shepherd have appeared. G. T. Hatch, in this laboratory, has recently extended the yield curve of alpha-particles up to 400 kilovolts bombarding energy of protons.

It seems well established that two reactions occur simultaneously, one producing alpha-particles, the other, a singly charged particle which is probably a deuteron.

$$_{4}\text{Be}^{9} + _{1}\text{H}^{1} = _{3}\text{Li}^{6} + _{2}\text{He}^{4} + Q,$$
 (1)

$$_{4}\text{Be}^{9} + _{1}\text{H}^{1} = _{4}\text{Be}^{8} + _{1}\text{H}^{2} + Q'.$$
 (2)

The reported work previous to that of Hatch was

<sup>&</sup>lt;sup>1</sup>Oliphant, Kempton, and Rutherford, Proc. Roy. Soc. Lond. A150, 241 (1935).

<sup>&</sup>lt;sup>2</sup> R. Döpel, Zeits. f. Physik **91**, 796 (1934) and **104**, 666 (1937); B. Zipprich, Zeits. f. Physik **96**, 337 (1935); Kirchner and Neuert, Physik. Zeits. **36**, 54 (1935) and **38**, 969 (1937); J. S. Allen, Phys. Rev. **51**, 182 (1937); Williams, Haxby, and Shepherd, Phys. Rev. **52**, 1031 (1937); G. T. Hatch, Phys. Rev. **54**, 165 (1938).





done at proton bombarding energies below 0.25 Mev. Under these conditions, the relative values of Q and Q' are such that the deuterons and alphaparticles have nearly the same range. The difficulty of their separate detection is thus increased. Table I shows the estimates of several observers concerning this range.

Because of the difficulty in showing that actually two different kinds of particles are present, Oliphant, Kempton, and Rutherford tried to separate the alpha's from the deuterons by passing the disintegration particles through an electric field between two parallel plates. The electric deflector was of low resolving power, and the particles showed no sign of separating into two groups. From this they concluded that  $\frac{1}{2} Mv^2/ze$  (where ze is the charge of the particle) is the same for the alphas and deuterons. This would mean  $E_{\alpha} = 2E_d$ , where E is the energy of the particles. They assigned to  $E_{\alpha}$  the value 1.1 Mev.

The measurement of the range of such low energy particles as these is an uncertain process, because of the necessity of introducing films, correcting for the depth of the ionization chamber, and straggling. At 7 mm range, an error of 1 mm represents roughly 200,000 volts, which is 15 percent of the total energy.

Hatch<sup>2</sup> has called attention to the remarkably large yield of alpha-particles from a solid target of beryllium under proton bombardment; at 400 kv a yield roughly 30 times that from a solid lithium target is obtained. Because of this high yield and low energy, it seemed to us worth while to try and increase the accuracy of the energy measurement by deflecting the particles in an electrostatic analyzer.

 
 TABLE I. Ranges of particles from beryllium under proton bombardment.

| Observer                         | PROTON ENERGY<br>(Mev) | ESTIMATED MEAN RANGE<br>(cm) |  |  |  |
|----------------------------------|------------------------|------------------------------|--|--|--|
| Oliphant, Kempton,<br>Rutherford | 0.18                   | 0.74                         |  |  |  |
| Döpel                            | 0.12                   | continuous, out to 0.68      |  |  |  |
| Kirchner and<br>Neuert (1935)    | 0.16                   | discrete, 0.75               |  |  |  |
| Kirchner and<br>Neuert (1937)    | 0.14                   | α's 0.75; d's 0.8            |  |  |  |
| Zipprich                         | 0.05                   | 0.66                         |  |  |  |

We chose to construct an electrostatic deflection device instead of a magnetic analyzer largely because we had available in the laboratory a source of very constant high voltage which could be very accurately measured on a potentiometer-bridge circuit. The primary currrent for the high voltage is obtained from a 540-cycle generator, turned by a three phase synchronous motor mounted on the same shaft. The 540-cycle current is raised to a voltage up to 100,000 volts by a specially built transformer, rectified by kenetrons, and smoothed by a 0.1 microfarad condenser. The only appreciable current drain in our experiments was about 1 ma drawn through the Taylor wire-wound resistor of 2.8 megohms for measurement of the voltage. By means of a vacuum tube stabilizing circuit operating in the generator field, the output voltage of the system could be held constant to 2 volts in 30,000 over extended periods of time.<sup>3</sup>

The inner surfaces of the deflecting plates of the analyzer are sections of surfaces of two coaxial cylinders. The mean radius of the cylinders is 25.40 cm, and the space between them, 0.635 cm. The angle subtended by the deflecting plates at the axis is 90°. The plates are constructed of aluminum and are 8.26 cm high. They are held apart by two Bakelite insulators which pass through slots in the inner faces and are attached to the deflecting plates at their outsides. Thus the deflecting voltage is not supported over merely the 0.635 cm of insulator between the plates, and the contact between the plates and the separating insulator takes place in a relatively field-free region. After construction, a test of the spacing between the plates with a precision gauge showed a maximum variation of 0.005 cm from the mean. The plates were mounted in a vacuum chamber constructed of brass tubing.

The constants of such a deflector can be readily computed from the following equations, which are rather obvious approximations to the rigorous treatment:

$$r = Mv^2/Eze,$$
(3)

$$E = V / \{ \frac{1}{2} (r_1 + r_2) \log r_1 / r_2 \}, \quad r_2 < r < r_1, \quad (4)$$

where *r* is the radius of curvature of the path of the particle through the analyzer,

 $r_1, r_2$  are the radii of the analyzer plates, V is the potential drop across the region between the analyzer plates.

For the beam of particles which travels along the average radius of the analyzer, we have r=25.400 cm, and  $r_1$  and  $r_2$  have the values 25.717 and 25.083 cm respectively. Using these values in the above equations, we find

$$E_2 = 20.00 Vz$$
 (5)

where  $E_2$  is the energy of the particles in electron volts and V the deflecting potential in volts.

In a preliminary experiment, the analyzer was used on the beam of protons accelerated by our Cockcroft-Walton type circuit. The high voltage produced by the Cockcroft-Walton circuit could be accurately read by measuring the current through the high resistor whose calibration has been described by Hatch.<sup>2</sup> This voltage should be numerically equal to  $E_2$ , and in the region of 400 kv we found satisfactory agreement (2 to 3 percent) between the necessary analyzing voltage V and the  $E_2$  measured with the  $7.5 \times 10^9$  ohm resistor. More precise agreement could not be obtained due to the ripple in the output of the Cockcroft-Walton circuit.

Such an electrostatic deflector acts as a lens, whose focal points may be calculated by equations developed by Hughes and Rojansky, Herzog, and Dempster.<sup>4</sup> These equations are:

$$\begin{aligned} f^2 &= (l'-g)(l''-g), \\ f &= \frac{1}{2}(r_1+r_2)/\{\sqrt{2} \sin 2^{\frac{1}{2}}\phi\}, \\ g &= \{\frac{1}{2}(r_1+r_2)/\sqrt{2}\} \cot 2^{\frac{1}{2}}\phi, \end{aligned}$$

where l' is the distance from the entrance slit to the entrance of the analyzer, and l'' is the distance from the exit of the analyzer to the focal point.  $\phi$  is the angle subtended by the analyzer plates at the axis. Particles of the same speed, but divergent in direction, which enter at the entrance slit, pursue different orbits through the analyzer, which, however, converge at the exit focus. With the numerical values of our analyzer, we find, as a special solution, l'=l''=8.90 cm.

<sup>&</sup>lt;sup>3</sup> We are indebted to Mr. F. V. Stearns for the careful construction of the potentiometer-bridge circuit, and to Mr. R. B. Bowersox for the successful stabilizing circuit.

<sup>&</sup>lt;sup>4</sup>A. L. Hughes and V. Rojansky, Phys. Rev. **34**, 284 (1929); R. Herzog, Zeits. f. Physik **89**, 447 (1934); A. J. Dempster, Phys. Rev. **51**, 67 (1936).

A slit 1 mm wide was placed at this distance from the entrance to the field, and a 3 mm slit was placed at the same distance from the exit. The entrance slit was 2.3 cm from the beryllium target. The 3 mm slit was a slot in the front cap of the ionization chamber in which the particles were counted. It was covered with screen and a thin foil and served as a wall of the vacuum chamber in which the analyzer operated. The height of this 3 mm slot was 2 cm.

The bursts of ionization due to the passage of charged particles through the detecting ionization chamber (3 mm deep) were amplified by a circuit of the Dunning<sup>5</sup> type, which was constructed by J.S. Allen.<sup>2</sup> The pulses were observed on the screen of an oscillograph. When the deflecting voltage of about 35,000 volts was put on the analyzer, the oscillograph showed disturbances of very high frequency, presumably connected with incipient discharges in the analyzer. Much experience in observing the oscillograph, however, made confusion of these disturbances with true particle pulses impossible. Two observers watched the oscillograph screen and counted independently the number of pulses due to particles in a given time. The two independent counts checked closely.

The resulting curves are shown in Fig. 2. The curve marked 320 kv corresponds to particles getting through the analyzer when 320 kv protons (magnetically selected) were directed at the beryllium target. Formation of carbon deposits on the target was prevented by the pre-heating method described by Hatch.<sup>2</sup> In taking the 320 kv curve, attention was concentrated on defining accurately the upper energy limit of the distribution, as the relatively large number of points along the upper limit indicates. Not much reliance can be placed on the shape of the 320 kv curve below the upper limit.

In taking the 383 kv curve, the observers selected visually pulses which were known to be of alpha-particle height. Deuteron pulses are only about 40 percent as intense as alpha-pulses at the maxima of specific ionization for both types, according to Kirchner and Neuert.<sup>2</sup> Thus it was established that the sharp upper limit found was due to alpha's. More confidence can



FIG. 2. The energy distribution of alpha-particles from  ${}_{4}\text{Be}^{9}$  (p,  $\alpha$ )  ${}_{3}\text{Li}^{6}$  at proton energies of 320 and 383 kv. The energies of the alpha-particles may be obtained by multiplying the kilovolt values by 40.00.

be placed in the shape of the curve below the upper limit for this curve.

Although many kicks of deuteron size were seen, the noise level was too high to enable the observers to reach accurate conclusions. It seems certain, however, that there is no peak of comparable height for deuterons, and that they must be distributed over a wider energy range than the alphas. The energy spectrum of the deuterons is now under investigation.

The voltage at which the particles were describing' exactly the average radius of the deflector was selected in the following manner. Let us consider a beam of particles, homogeneous in energy, admitted to the deflecting system through the 1 mm wide entrance slit. Due to the focusing action, this beam will again be one mm wide at the 3 mm slit, and changing the deflecting voltage will sweep this 1 mm beam across the 3 mm slit. When the center of the beam is displaced 2 mm from the center of the 3 mm slit, no particles will get through. From the equations of the analyzer,

$$r = \{\frac{1}{2}Mv^2(r_1+r_2) \log (r_1/r_2)\}/zeV,$$

whence dV/V = -dr/r.

Setting dr equal to 2 mm, the calculated correction dV is subtracted from the deflection voltage corresponding to the intersection of the extrapolated linear high energy edge with the kilovolt axis. The results are given in Table II.

A solution of the equations of conservation of

<sup>&</sup>lt;sup>5</sup> J. R. Dunning, Rev. Sci. Inst. 5, 387 (1934).

energy and momentum for the special case of particles leaving at right angles to the proton beam gives

$$E_{\alpha} = \{E_{p}(1 - M_{p}/M_{\text{Li}}) + Q\}/(1 + M_{\alpha}/M_{\text{Li}}),$$

where  $E_{\alpha}$  is the energy of the alpha-particle,

 $E_p$  is the energy of the proton,

Q is the energy balance in the raction,

 $M_p$ ,  $M_\alpha$ ,  $M_{\text{Li}}$  are the masses of the proton, alpha-particle and the lithium six isotope, respectively.

From this equation the values of *Q* in Table II were computed. It also follows that

$$dE_{\alpha}/dE_{p} = (1 - M_{p}/M_{\text{Li}})/(1 + M_{\alpha}/M_{\text{Li}}) = 0.500.$$

For the two curves shown in Fig. 2,  $dE_p$  is 0.063 Mev, from which the value of  $dE_\alpha$  is computed to be 0.032 Mev. From Eq. (5) the corresponding dV is 0.794 kv. The difference in the intercepts on the axis of Fig. 2 is 0.92 kv, which is in agreement with the computed value within the limit of error. It is estimated that the experiment gives the energy of alpha-particles passing through the analyzer at a given deflecting voltage to about 2 percent.

The value of *Q* recommended by Livingston and Bethe,<sup>6</sup> is 2.28 Mev. The more precise value found in this work has a much higher internal consistency with other disintegration experiments. Consider the following cycle of reactions:

$${}_{4}Be^{9}+{}_{1}H^{1}={}_{3}Li^{6}+{}_{2}He^{4}+Q,$$
  
 ${}_{4}Be^{9}+{}_{1}H^{2}={}_{3}Li^{7}+{}_{2}He^{4}+7.19$  Mev,  
 ${}_{3}Li^{6}+{}_{1}H^{2}={}_{3}Li^{7}+{}_{1}H^{1}+5.02$  Mev.

| TA | BLE | 11. | Comput | tations. | from | curves | of | Fig. 2. | • |
|----|-----|-----|--------|----------|------|--------|----|---------|---|
|----|-----|-----|--------|----------|------|--------|----|---------|---|

| Proton<br>En-<br>ergy<br>(kv) | Maximum<br>Deflecting<br>Voltage<br>(kv) | Corrected<br>Deflecting<br>Voltage<br>(kv) | Alpha-<br>Particle<br>Energy<br>(Mev) | Q<br>(Mev)                                  | Mass<br>Equiva-<br>Lent of<br>$Q \times 10^3$ |
|-------------------------------|--|--|---------------------------------------|---|---|
| 320<br>383                    | 36.30<br>37.22                           | 36.02<br>36.93                             | 1.441 $1.477$                         | $\begin{array}{c} 2.148\\ 2.155\end{array}$ | 2.306<br>2.315                                |

<sup>6</sup> Livingston and Bethe, Rev. Mod. Phys. 9, 246 (1937).

The energy balances quoted above for the last two reactions are from Livingston and Bethe's report, and the ranges of the particles are long enough so that range determinations are quite reliable. The third of the above reactions subtracted from the second leaves the first. Thus the predicted Q for the first reaction is 2.17 Mev, which is in excellent agreement with the average of our two results, namely, 2.15 Mev.

The data from our experiment can be used to give a precise difference between the masses of Be<sup>9</sup> and Li<sup>6</sup>, since the masses of the proton and alpha-particle are known with much higher accuracy than is the mass of either of the heavier atoms concerned. We have

$$M_{\rm Be} - M_{\rm Li} = M_{\alpha} + M_Q - M_p$$

where  $M_Q$  is the energy balance in mass units. We have used the factor 931 Mev equals 1 atomic weight unit to make this transformation. Using  $M_{\alpha} = 4.00386 \pm 0.00006$ ,  $^7 M_p = 1.00813 \pm 0.00002^6$ and taking an average value of the mass equivalent of Q as  $2.316 \pm 0.04 \times 10^{-3}$ , we obtain

$$M_{\rm Be} - M_{\rm Li} = 2.99804 \pm 0.00009.$$

Although neither the mass of Be<sup>9</sup> nor Li<sup>6</sup> is known with satisfactorily high accuracy, we may assume that of the two, Li<sup>6</sup> is the more accurate. Adopting Livingston and Bethe's value of  $6.01686 \pm 0.00020$ , we get

$$M_{\rm Be} = 9.01490 \pm 0.00025$$

This is considerably lower than Livingston and Bethe's adjusted value of  $9.01504 \pm 0.00025$ , and is much lower than Jordan and Bainbridge's<sup>8</sup> mass-spectroscopic value of  $9.01517 \pm 0.00016$ .

We wish to thank Mr. G. T. Hatch and Mrs. E. R. Graves for assistance in performing the experiments.

<sup>&</sup>lt;sup>7</sup> K. T. Bainbridge, Bull. Am. Phys. Soc. **13**, No. 2 (1938). <sup>8</sup> E. B. Jordan and K. T. Bainbridge, Phys. Rev. **51**, 385 (1937).