

and  $\text{Te}^{123}$ . It is hoped that the examination of  $\text{In}^{113}$  may be possible as this isotope is present to approximately 5 percent of  $\text{In}^{115}$  and so is a nucleus more amenable to analysis than  $\text{Sn}^{115}$  or  $\text{Te}^{123}$ .

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### The Transmutation of Sulphur by Th C' Alpha-Particles

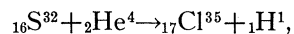
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The protons emitted from sulphur under bombardment of Th C' alpha-particles have been resolved into three groups corresponding to nuclear energy changes  $-2.4 \pm 0.3$ ,  $-2.85 \pm 0.3$  and  $-3.6 \pm 0.3$  MEV. The first of these values, together with Bainbridge's value for the mass of  $\text{Cl}^{35}$ , has been used to deduce a value for the mass of  $\text{S}^{32}$ ,

namely  $31.9812 \pm 0.0016$ . An excitation curve for the emission of protons has been plotted which obeys the Gamow penetration formula within the limits of error: from this formula the nuclear radius of  $\text{S}^{32}$  has been derived and has the value  $5.4 \pm 0.3 \times 10^{-13}$  cm.

THE determination of the energy levels of as many nuclei as possible is an essential preliminary to the foundation of a satisfactory nuclear theory. In particular, the binding energy of a nucleus, which depends on its mass defect, is of great importance and the present knowledge of nuclear masses as far as neon has already been used to test various suggested intranuclear forces and constitutions. Beyond neon the information is unsatisfactory and there is clearly need for an experimental determination of as many nuclear masses as possible in this region of heavier nuclei. The element sulphur offers an opportunity for investigation since the isotopic mass of its neighbor chlorine is known and a transmutation to chlorine, with a determination of the energy change involved, would enable the isotopic mass of sulphur to be found. Such a transmutation is feasible according to the reaction:



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in which the only unknown mass is that of sulphur.

The fact that sulphur has the relatively high nuclear charge of 16 means that its potential barrier is high and therefore that the proposed transmutation will not take place unless very energetic particles are used to bombard it. The alpha-particles emitted by Th C' are the most energetic available, having an energy of nine million electron volts, and are therefore the most likely to effect the required change. We therefore bombarded a sulphur target with Th C' alpha-particles with intent to detect emitted protons and measure their maximum energy. This has enabled the mass of sulphur to be derived. At the same time transmutations leaving a chlorine nucleus in an excited state occur, giving groups of shorter range protons, and measurement of the ranges of these groups has enabled the values of the excited energy levels to be found.

It is further possible to derive information

about the potential barrier of the sulphur nucleus by determining the variation of proton yield with alpha-particle energy and so to make an estimate of the nuclear radius.

The transmutation of sulphur by alpha-particles was first reported by Rutherford and Chadwick<sup>1</sup> who did not, however, measure the energy of the emitted protons. In a recent account Haxel<sup>2</sup> describes experiments on the three elements Mg, Si and S showing that the protons from each element are emitted in three groups and that the interval between corresponding values of the nuclear energy change appears to be the same for each element.

#### EXPERIMENTAL PROCEDURE

The technique for observing protons in the presence of ionizing radiations has been described by Wynn-Williams and Ward<sup>3</sup> and elaborated by Dunning, Haxel and Duncanson and Miller.<sup>4</sup> As detection apparatus we used a proportional counter following the design of Geiger and Zahn<sup>5</sup> consisting of a fine axial wire in a cylindrical case at a high negative voltage. The case, which was maintained at -3600 volts was filled with air at 25 cm pressure: the axial wire was of nichrome, one-half millimeter diameter. The wire was connected to the grid of an amplifier, the output from which actuated both a thyratron relay-Cenco counter system and a cathode-ray oscillograph. The grid leak on the first stage of the amplifier was small (one megohm), thus favoring the sharp proton deflections rather than the irregular wobble due to the gamma-ray background and a "tone control" device set to pass only frequencies above roughly a thousand was included to accentuate the shortening of the time for an impulse to record. The deflections were watched on the oscillograph and the thyratron bias set at the minimum value which did not respond to gamma-ray background fluctuations. We were unable to

count the fastest protons in this way because of their low specific ionization, but for the separation of groups this is no great disadvantage. (It must, however, be remembered, in considering the absorption curves given later, that a rise in the numbers counted at any absorption means only a rise in the number of *detected* particles.) We estimated from trial experiments that our counting level was such as to record all protons of residual range less than fifteen centimeters of air.

In experiments like this there is some choice in the geometrical conditions of bombardment and detection. If  $\theta$  is the angle between an incident alpha-particle causing a transmutation and the ejected proton and if  $Q$  is the nuclear energy change in the process then we have the equation:

$$2M_N Q = M_P(M_P + M_N)V_P^2 - M_\alpha(M_N - M_\alpha)V_\alpha^2 - 2M_\alpha M_P V_P V_\alpha \cos \theta, \quad (1)$$

where  $M_N$  = Mass of residual nucleus,  
 $M_P$  = Mass of proton,  
 $M_\alpha$  = Mass of alpha-particle,  
 $V_\alpha$  = Initial velocity of alpha-particle,  
 $V_P$  = Velocity of emergent proton.

This equation simply expresses the conservation of energy and momentum. (The quantity " $Q$ " for *maximum energy* protons represents the difference between the mass of sulphur plus alpha-particle and chlorine plus proton. If the resultant chlorine nucleus is formed in an excited state, the corresponding value of  $Q$  is less: the residual energy being supposed emitted as a gamma-ray quantum.) If the protons are observed in the same direction as the incident alpha-particles  $\theta$  is zero and the proton velocity is maximum for a given alpha-particle energy. This would be the most advantageous arrangement except for the presence of "natural" protons from the source of range 40 cm which mask all groups of less than this range. If observation is made at an angle of ninety degrees between alpha-ray and proton a shield can be placed between the source and the counter, eliminating the natural protons and also any projected hydrogen nuclei from hydrogen in the target. For a full survey of the groups the so-called "right angles" method of observation is therefore the best, while for determining the

<sup>1</sup> E. Rutherford and J. Chadwick, Proc. Phys. Soc. London **36**, 417 (1924).

<sup>2</sup> O. Haxel, Physik. Zeits. **36**, 804 (1935).

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

<sup>4</sup> J. Dunning, Rev. Sci. Inst. **5**, 387 (1934); O. Haxel, Zeits. f. Physik **83**, 323 (1933); W. E. Duncanson and H. Miller, Proc. Roy. Soc. **A146**, 396 (1934).

<sup>5</sup> H. Geiger and H. Zahn, *Handbuch der Physik*, Vol. XXII, [2] p. 163.

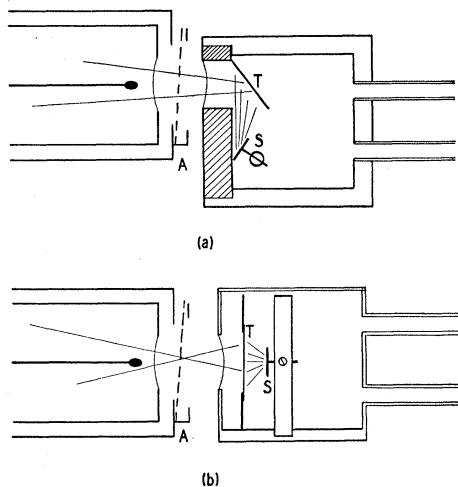


FIG. 1. Arrangements for bombardment of target and detection of protons.

maximum range of the protons the "forwards" method (protons in same direction as the alpha-rays) is more satisfactory. The forwards method allows a greater solid angle between the source and the target and consequently an increased yield of protons.

These two arrangements are illustrated in Fig. 1(a) and (b). (a) shows the "right angles" method: *S* is the alpha-particle source deposited on a monel metal button 9 mm diameter by exposure to the emanation from radiothorium, *T* is the sulphur target on cardboard backing. The activity of the source varied between 2 and 3 millicuries. Both are enclosed in a brass box which can be evacuated: the shaded wall of the box is of lead of one centimeter thickness (which served to diminish the gamma-rays reaching the counter) with a 5/8" hole covered by an aluminum foil of 4.1 cm air equivalent. The whole box was placed in a magnetic field of about 5000 gauss to bend back (and so remove) secondary electrons produced in the target. The counter opening was sealed with a mica window of 4.8 cm air equivalent. With this arrangement the minimum angle between alpha-ray and detected proton is 80° and the maximum 120° giving a solid angle of observation (the product of the solid angle between source and target and that between target and counter) of 1/1400.

The "forwards" method is shown in (b). The sulphur target was deposited on a thick gold foil by spraying a suspension of sulphur in carbon

tetrachloride over the gold. The foil had an absorption greater than 8.6 cm so that the aluminum window closing the box was not bombarded by alpha-particles. In this arrangement a magnetic field is essential to bend out the beta-rays. The solid angle of observation was approximately 1/400.

In both arrangements absorption curves of the protons were plotted by interposing aluminum screens between the opening of the box and that of the counter as at *A*. The air equivalent of these screens was determined by weighing, 1.64 mg per square centimeter being taken as equivalent to 1 centimeter of air.

## EXPERIMENTAL RESULTS

### Absorption curves and nuclear energy changes

Absorption curves plotted using the right angles arrangement are shown in Fig. 2. The upper curve is for full range alpha-particles; the lower for 7.5 cm range. The sharp drop in yield shows that the 4.9 cm alpha-particles also present are not effective in causing transmutations. It will be seen that the protons form three groups of ranges 24 cm, 32 cm, and 38 cm air equivalent when full range alpha-particles are used, the values being considered good to within 1.5 cm. For this curve about one hundred particles were counted at each point. We have not included here in the diagram several runs in which the counting level was different which also showed the presence of the three groups at the ranges given. The value 8.4 cm is used for the alpha-particle range rather than the maximum value of 8.6 cm since the last two millimeters contain

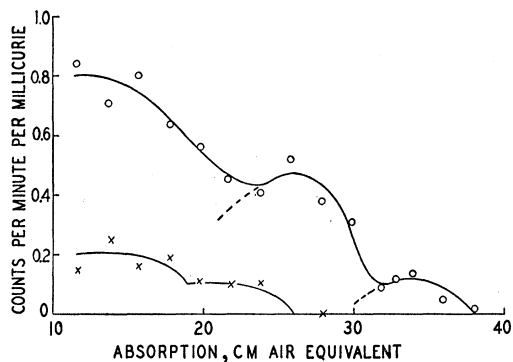


FIG. 2. Absorption curve for protons emitted at 90° to incident alpha-particles.

a very small proportion of the particles from the source. 8.4 cm is taken as a limit which is more nearly that responsible for the ends of the experimentally found proton groups. Using the range-velocity data from the Cavendish laboratory curves the nuclear energy changes "Q" corresponding to the three groups are found to be

$$-3.6 \pm 0.3; \quad -2.85 \pm 0.3; \quad -2.35 \pm 0.3 \text{ MEV,}$$

as calculated from the upper curve, and  $-3.6$  and  $-2.90$  MEV from the lower, less reliable, curve. (The numbers in the third group were too small to be detected without long counting.)

In consequence of the extra momentum imparted by the recoiling nucleus the 38 cm group found above should give a group of protons in the forward direction having a range beyond that of the natural protons (40 cm). A more accurate determination of the highest "Q" value can therefore be made using the forward arrangement. The results of experiments in this way are shown in Fig. 3. Here the maximum range alpha-particles were used. A single group is seen whose range is 46 cm correct to 1.5 cm. A careful investigation beyond this range showed no signs of an additional group so this value of 46 cm presumably corresponds to a transition from the ground state of sulphur to the ground state of chlorine and can be used to compare the two masses. On this basis the calculated value of "Q" is  $-2.4 \pm 0.2$  MEV, in good agreement with that found previously. Expressing this energy change in terms of atomic weight units we have  $Q = -0.0026$ . The equation

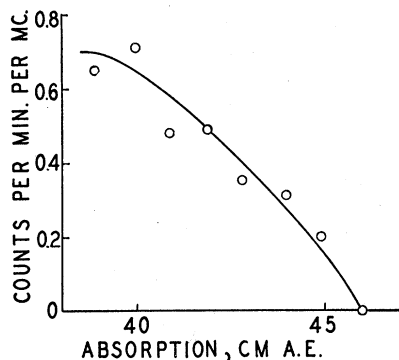
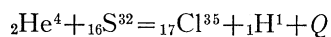


FIG. 3. Absorption curve for protons emitted in the same direction as the incident alpha-particles.

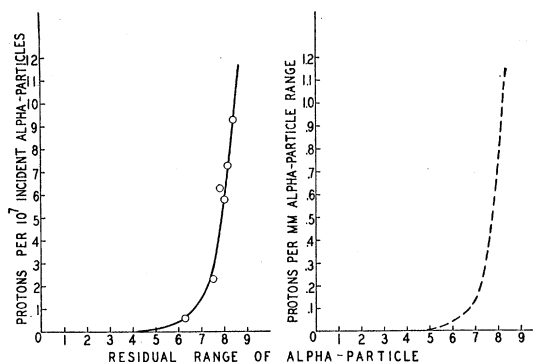


FIG. 4. Excitation curve and derived differential curve.

then becomes

$$4.0039 + {}^{32}_{16}\text{S} = 34.9796 + 1.0081 - 0.0026$$

$$(\pm 0.0002) \quad (\pm 0.0012) \quad (\pm 0.0002)$$

and we derive the value  $31.9812 (\pm 0.0016)$  for the mass of the neutral sulphur atom. The masses for helium and hydrogen are those found recently by Aston and quoted by Oliphant,<sup>6</sup> while the mass of chlorine is the value found by Bainbridge.<sup>7</sup> The energy values reported by Haxel<sup>2</sup> are  $-3.85$ ,  $-2.90$ ,  $-2.25$  MEV. Our results, which are in good agreement with these, thus give a confirmation of his work.

### Excitation curve and nuclear radius

Fig. 2 shows that there is a marked diminution in the yield as the energy of the incident alpha-particles is reduced. This is due to the fact that fewer alpha-particles are able to penetrate the barrier of the sulphur nucleus. In order to determine the form of the excitation curve experiments were made at six different alpha-particle energies—the residual range being reduced by placing thin gold or aluminum screens over the source. The procedure adopted was to plot a rough absorption curve for the emitted protons in each case and to take a mean value of the yield in the flat part of the curve between eleven and sixteen centimeters air equivalent absorption. By so doing a true excitation curve for the shortest range group is plotted, not merely a curve showing diminution of yield at one absorption which may be due to the shortening of range of a proton group to less than the

<sup>6</sup> M. L. Oliphant, Nature **137**, 396 (1936).

<sup>7</sup> K. T. Bainbridge, Phys. Rev. **43**, 378 (1933).

detected absorption. The six rough absorption curves showed that the yields of all three groups changed simultaneously, a behavior different from that found by Haxel<sup>8</sup> for the excitation of protons from aluminum.

The resulting excitation curve is shown in Fig. 4. As this is plotted for a thick target the excitation for a thin layer is obtained by differentiation and is shown as a dotted curve. The ordinates give the total yield in all directions for  $10^7$  alpha-particles incident on the target for the thick curve and yield per  $10^7$  alpha-particles for one millimeter target thickness for the differential curve.

The numbers available at the lower counts were not very large and therefore the curve is necessarily somewhat rough. It is interesting, nevertheless, that a reasonable estimate of the radius of the sulphur nucleus can be made even from these data, since absolute yields are determined. Thus if we assume that the sulphur nucleus has a barrier of the Gamow type and that the Gamow law for penetration holds we can say at once that the continued rise at 8 cm alpha-particle range means the barrier height exceeds the corresponding energy of  $12 \times 10^{-6}$  erg (7.5 MEV) or that the nuclear radius cannot be greater than  $6.0 \times 10^{-13}$  cm. At the same time the fact that there is an appreciable yield at 7.0 cm range means that the radius cannot be less than  $4.5 \times 10^{-13}$  cm for then the expected

yield on the Gamow theory would be too small by a factor of one hundred. In an appendix a detailed account of a use of our excitation curve to derive a probable value for the nuclear radius is given—the value arrived at is  $(5.4 \pm 0.3) \times 10^{-13}$  cm. This is slightly higher than would be expected on the basis of Gamow's values for the radioactive nuclei but is nevertheless not unreasonable.

### Nuclear spin and this reaction

According to a suggestion made by Goldhaber<sup>9</sup> a nuclear reaction will be probable if the nuclear spin is conserved. The present reaction is one in which a very considerable change of spin takes place since both sulphur and helium have zero spin while chlorine has a value of  $5/2$  and hydrogen  $1/2$ . We estimate that our yield is similar in magnitude to that from other nuclear reactions if account is taken of the difference in penetration of the barrier. It is possible that at high energies of bombardment the conservation of nuclear spin plays a less important part than at the low energies used for the transmutation of light nuclei such as lithium and boron.

In conclusion we wish to express our thanks to Professor A. F. Kovarik for his interest and advice, to Dr. C. D. Bock for help in the setting up of a voltage stabilizer of his design and to Mr. H. L. Schultz for the construction of the amplifier.

### APPENDIX

#### Derivation of the nuclear radius of $_{16}\text{S}^{32}$

We suppose the nucleus has a potential barrier of the Gamow type consisting of a cut-off Coulomb barrier falling sharply to negative potentials at a radius defined as the nuclear radius  $r_0$ . The process of disintegration is considered to be as follows:

- (1) The alpha-particle must collide with a nucleus.
- (2) It must penetrate into the region  $r < r_0$ .
- (3) It must then fall to a stable level giving energy to a proton which is ejected.

If the probabilities of these processes are called  $P_1$ ,  $P_2$  and  $P_3$  then the yield of protons observed is the product  $P_1 P_2 P_3$ . Of these three probabilities,  $P_1$  and  $P_3$  vary slowly with the nuclear radius while  $P_2$ , which follows the Gamow formula, is a rapidly varying function of it: thus a change of ten percent in the radius changes  $P_2$  by a factor of five.

Then if we can make an estimate of  $P_2$  correct within a factor of five we can derive the nuclear radius correct to ten percent. We give below methods of estimating  $P_1$  and  $P_3$  which, together with the experimentally observed values of  $P_1 P_2 P_3$  allow us to estimate the corresponding values of  $P_2$ , from which we can deduce the nuclear radius. The estimates of  $P_1$  and  $P_3$  are not accurate but since wide limits of error are permissible it is still possible to give a reasonable value for the nuclear radius.

#### Probability $P_1$

This may be estimated in two ways, the first following a theoretical approach, the second in terms of experimentally observed data. The quantum mechanical approach to the problem involves regarding a beam of alpha-particles as a plane wave which can be split into a number of partial waves of increasing angular momentum. Only that of zero

<sup>8</sup> O. Haxel, *Zeits. f. Physik* **90**, 373 (1934).

<sup>9</sup> M. Goldhaber, *Proc. Camb. Phil. Soc.* **30**, 561 (1934).

order is supposed to be effective in disintegrating (meaning only head-on—or nearly so—collisions cause disintegration). This wave has a collision cross-section equal to the square of the wave-length of the alpha-particles which is

$$\sigma = \left[ \frac{h}{mv} \right]^2 = \left[ \frac{6 \times 10^{-27}}{6.6 \times 10^{-24} \times 2.0 \times 10^9} \right]^2 = 2 \times 10^{-25} \text{ sq. cm.}$$

Or alternatively, adopting the experimental approach, we can suppose  $\sigma$  to be the actual cross-section of a nucleus whose radius we know lies between  $4.5$  and  $6.0 \times 10^{-13}$  and so can be estimated as  $5 \times 10^{-13}$  cm giving  $\sigma = 0.8 \times 10^{-18}$  sq. cm.

Now a target one square centimeter in area and of one millimeter air equivalent thickness contains  $3.0 \times 10^{18}$  atoms. These present an area  $3.0 \times 10^{18} \sigma$  to the incident alpha-particles for disintegration purposes. This represents the fraction of area which is effective and so is a measure of  $P_1$  the chance that an alpha-particle will collide with a nucleus in passing through one millimeter air equivalent of sulphur.

These two estimates therefore yield the values  $P_1^A$ ,  $P_1^B$  for the probability of collision given by

$$P_1^A = 2 \times 10^{-25} \times 3 \times 10^{18} = 6 \times 10^{-7},$$

$$P_1^B = 0.8 \times 10^{-24} \times 3 \times 10^{18} = 2 \times 10^{-6}.$$

**Probability  $P_2$**

The probability of penetration after collision is supposed to be given by the Gamow formula assuming the component of angular momentum zero is alone effective. The formula is

$$P_2 = A \exp \frac{-2(2m)^{\frac{1}{2}}}{\hbar} \int_{r_0}^r \left( \frac{2Ze^2}{r_0} - \frac{2Ze^2}{r} \right)^{\frac{1}{2}} dr,$$

$2Ze^2/r$  = energy of incident particle in the nuclear field at radius  $r$ ,

$2Ze^2/r_0$  = energy at the top of the barrier.

$A$  is a quantity depending on the energy of the bottom of the nuclear potential well which can be treated as unity for a first approximation. The evaluation of this integral gives the final numerical value

$$P_2 = e^{-375.8 \times 10^5 r^{\frac{1}{2}} [2u_0 - \sin 2u_0]},$$

where  $\cos u_0 = (r_0/r)^{\frac{1}{2}}$ .

**Probability  $P_3$**

Having entered, how often does an alpha-particle eject a proton? This again can be approached theoretically or

experimentally. Theoretically, taking  $P_1^A$  based on the wave-length of the alpha-particle the order of magnitude of  $P_3$  is the velocity of the proton divided by the nuclear radius (assuming the proton clears the barrier). This is already well in excess of unity. Hence if we use  $P_1^A$  we can assume  $P_3^A = 1$ .

Experimentally, Blackett and Lees have found for nitrogen disintegration that between one in two and one in ten sufficiently close collisions eject a proton. The general run of disintegration experiments indicate that there is no wide deviation from these limits. Hence using  $P_1^B$  we can take  $P_3^B$  as between  $1/2$  and  $1/10$ . In either case the product  $P_1 P_3$  is of the order  $10^{-6}$ . We wish to emphasize that in spite of an error of a factor of five which is possible in making this estimate, a value for the nuclear radius can nevertheless be derived.

**Application to our experiments**

We find that at 8.4 cm range we get 1.2 protons per  $10^7$  alpha-particles incident on a target of thickness one millimeter air equivalent. Then we have  $P_1 P_2 P_3 = 1.2 \times 10^{-7}$ . The limits of  $P_1 P_3$  can be put as  $10^{-6}$  to  $2 \times 10^{-7}$  with a mean  $6 \times 10^{-7}$  from which we derive  $P_2 = 0.2$  with limits 0.12 and 0.6. We now compare our experimental curves with theoretical curves for the variation of  $P_2$  with energy seeking one which gives  $P_2 = 0.2$  for the point at 8.4 cm range and estimating the limiting values of the radius from curves which pass through  $P_2 = 0.12$  and 0.6. Comparative figures are given below (alpha-particle energies have been corrected for recoil motion by multiplying experimental values by 32/36)

Energy of alpha-particle	$P_2$	Experimental
$r_0 = 5.2 \times 10^{-13}$ cm		
12.0 $\times 10^{-6}$ erg	.110	.120
11.5 $\times 10^{-6}$ erg	.040	.045
11.0 $\times 10^{-6}$ erg	.015	.019
$r_0 = 5.4 \times 10^{-13}$ cm		
12.0	.200	.200
11.5	.110	.080
11.0	.030	.034
$r_0 = 5.7 \times 10^{-13}$ cm		
12.0	.60	.60
11.5	.32	.24
11.0	.09	.10

Hence we can set  $5.4 \pm .3 \times 10^{-13}$  as the nuclear radius of sulphur.

It is of some interest that the Gamow penetration formula for angular momentum zero appears to be obeyed although we do not claim that our experiments give a very rigorous test.