Nuclear Transmutations of the Lithium Isotopes

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Yield curves have been obtained for eight of the more important reactions of Li^6 and Li^7 produced by proton and deuteron bombardment in the energy range from 200 to 1000 kev. Each curve was obtained with an accuracy of about five percent, and a set of intercomparisons served to determine the relative yields between the different reactions within 20 percent. A special set of observations on a single reaction then gave the absolute yield for all the reactions within a factor of about three. An investigation was made of the reactions involved in the formation and decay of Li^8, the mass of which was found to be 8.02499 $\pm$0.00020. The range-distribution of the delayed alpha-particles from Li^8 was measured and the interpretation of this distribution is discussed. Observations on the reaction $Li^6+n\rightarrow He^4+H^3$ gave a value of 4.97 Mev for the reaction energy as compared to a value of 4.56 Mev derived from the masses involved. No evidence could be obtained for the production of Li^8 by the process $Li^7+n\rightarrow Li^8$. Measurements of the energies of the neutrons produced by deuteron bombardment of Li^6 indicated that Be^7 is formed by the reaction $Li^6+D^2\rightarrow Be^7+n$. Further experiments showed that Be^7 is radioactive and is converted to Li^7 by K-electron capture. It was found that this process is followed by gamma-ray emission in roughly ten percent of the transitions.

A. INTRODUCTION

LITHIUM was the first element to be disintegrated by artificially accelerated ions and its transmutation processes have been studied by many investigators, but at the time this work was undertaken there still remained much valuable information to be obtained from a more comprehensive investigation and correlation of the various processes involved in its transmutations. The products of disintegration have long been known and the energy balances have been measured for most of the possible modes of disintegration, but very little information was available concerning the yields of the various reactions and the variations of the yields with the energy of the bombarding particles. For a more complete understanding of the nucleus, measurements of the relative yields of different particles from the same compound nucleus are highly important. While the types of reactions which will occur and their energy balances can be predicted from a knowledge of the masses involved, and the general shape of most of the yield curves can be calculated by consideration of the penetration probabilities, the differences between yield curves for different isotopes or different projectiles probably depend on the arrangement of the nuclear constituents themselves, and thereby furnish definite criteria for testing the success of any theory of the structure of the nucleus.

It was the object of the present work to supplement the investigation of the transmutation processes of lithium previously reported[1] by obtaining accurate data on the yields of the various processes and by measuring quantitatively several other features of the reactions which had been studied only in a qualitative fashion. Some of the material covered by this paper has been reported briefly in the *Physical Review*.[2]

Because of the large number of possible reactions involved, they are collected in Table I for the convenience of the reader.

B. EXPERIMENTAL PROCEDURE

In general, the experimental procedure was similar to that of the previous work. The particles used in bombardment were accelerated by the two-meter electrostatic generator of the Department of Terrestrial Magnetism. Voltages were measured by the high resistance voltmeter,

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[1] L. H. Rumbaugh and L. R. Hafstad, Phys. Rev. **50**, 681–689 (1936).

[2] L. H. Rumbaugh, R. B. Roberts, and L. R. Hafstad, Phys. Rev. **51**, 1013 (1937); **51**, 1106–1107 (1937); R. B. Roberts, L. R. Hafstad, and L. H. Rumbaugh, Phys. Rev. **52**, 247 (1937); R. B. Roberts and N. P. Heydenburg, Phys. Rev. **53**, 929 (1938); R. B. Roberts, N. P. Heydenburg, and G. L. Locher, Phys. Rev. **53**, 1016 (1938).

previously described,[3] the absolute calibration of which has since been checked independently by observations of the scattering of protons in nitrogen and argon. The ion current could be varied at will from 0.2 to 5.0 microamperes, which was a great convenience in maintaining suitable counting rates. Molecular ions were employed for very low voltage work on deuteron produced reactions. The ion current was measured by a current integrator.

Targets

As in the previous work, a set of 18 isotopic targets which ranged from 10 micrograms to 4 milligrams in mass was available for use in any case where the disintegration products of the two isotopes might be confused. All yield curves were taken with thin targets.

Detecting apparatus

The apparatus used in detecting the various products of disintegration included several ionization chambers and Geiger-Müller counters with their associated amplifiers and counting circuits, a shielded Lauritsen electroscope, and a Wilson cloud chamber. Special features of the equipment will be described in conjunction with the experiment in which the apparatus was used.

TABLE I. *Nuclear reactions of lithium.**

$Li^6+H^1\rightarrow \mathbf{He^3}_{1.2\text{ cm}}+\mathbf{He^4}_{8\text{ cm}}+3.72$ Mev
$Li^7+H^1\rightarrow \mathbf{He^4}_{8\text{ cm}}+\mathbf{He^4}_{8\text{ cm}}+17.13$ Mev
$Li^7+H^1\rightarrow (Be^8)^*\rightarrow Be^8+\boldsymbol{h\nu}+(17$ Mev)
$Li^6+D^2\rightarrow \mathbf{He^4}_{13\text{ cm}}+\mathbf{He^4}_{13\text{ cm}}+22.07$ Mev
$Li^6+D^2\rightarrow Li^7+\mathbf{H^1}_{30\text{ cm}}+5.03$ Mev
$Li^6+D^2\rightarrow (Li^7)^*+\mathbf{H^1}_{25\text{ cm}}+4.58$ Mev
$(Li^7)^*\rightarrow Li^7+\boldsymbol{h\nu}+0.455$ Mev
$Li^6+D^2\rightarrow He^3+He^4+\boldsymbol{n}+(1.56$ Mev)
$Li^6+D^2\rightarrow Be^7+\boldsymbol{n}+(3.1$ Mev)
$Be^7+e_K\rightarrow Li^7+\eta+(1$ Mev) } (half-life 43 days)
$Be^7+e_K\rightarrow (Li^7)^*+\eta+(0.55$ Mev) } (half-life 43 days)
$(Li^7)^*\rightarrow Li^7+\boldsymbol{h\nu}+0.45$ Mev
$Li^7+D^2\rightarrow Be^8+\boldsymbol{n}+(14.5$ Mev)
$Li^7+D^2\rightarrow He^5+\mathbf{He^4}_{7\text{ cm}}+(14.3$ Mev)
$(He^5)\rightarrow \mathbf{He^4}_{2-6\text{ cm}}+\boldsymbol{n}+(0.8$ Mev)
$Li^7+D^2\rightarrow \mathbf{He^4}_{0-8\text{ cm}}+\mathbf{He^4}_{0-8\text{ cm}}+\boldsymbol{n}+(14.9$ Mev)
$Li^7+D^2\rightarrow Li^8+H^1-0.2$ Mev
$Li^8\rightarrow \mathbf{He^4}_{0-6\text{ cm}}+\mathbf{He^4}_{0-6\text{ cm}}+\beta^-+\eta+15.9$ Mev (half-life 0.85 sec.)
$Li^6+n\rightarrow \mathbf{He^4}_{1\text{ cm}}+\mathbf{H^3}_{6\text{ cm}}+4.9$ Mev
$Li^6+n\rightarrow He^6+H^1-2.8$ Mev (probable)
$Li^7+n\rightarrow Li^8+h\nu$ (not observed)
$Li^7+n\rightarrow He^6+H^2-2.7$ Mev (probable)

* The ranges given are for identification only, and are taken from the well-known original work in which the existence of such groups were established, no adjustment being made for differences in bombarding energies in different experiments. The disintegration products which have been observed are shown by boldface type. Those reaction energies included in parentheses are approximate.

[3] L. R. Hafstad, N. P. Heydenburg, and M. A. Tuve, Phys. Rev. **50**, 504–514 (1936).

Range measurements

In all measurements of the ranges of the immediate products of disintegrations, the observations were made at 90° to the incident beam. The smallest aperture which would give adequate counting rates was used. Two sets of calibrated mica windows mounted on wheels provided a means of rapidly varying the stopping power in 5-mm steps. A screw on the ionization chamber furnished a fine adjustment. In all cases where accurate range measurements were desired the stopping power of the window of the target chamber and of the effective air path to the ionization chamber was calibrated by placing a clean polonium source at the target position and measuring the range of the emitted alpha-particles. In converting to energy, the revised Cornell range-energy curves of 1937 were used for protons and those of 1938 for alpha-particles.

Elimination of spurious effects

In the first attempt to obtain accurate yield curves, some difficulty was encountered in getting reproducible results. The source of the difficulty was soon traced to the current measurements which recorded as effective current some ions which entered the target chamber but were not effective in producing observable disintegrations; that is, ions missing the target area or striking parts of the target shielded from the detector. To correct this condition the target chamber was placed at the end of a 60-cm collimating tube. At each end of this tube was a diaphragm with a $\frac{3}{16}$-inch hole, which insured a well-defined beam at the target which was located approximately ten cm below the last diaphragm. The collimating tube was built in insulated sections so that it also served as a trap for the electrons liberated from the diaphragms. Very little ion current was sacrificed by this arrangement due to the sharp focus of the accelerating tube. Fluctuations of the current caused no errors since the current was measured by an integrator. Voltage fluctuations

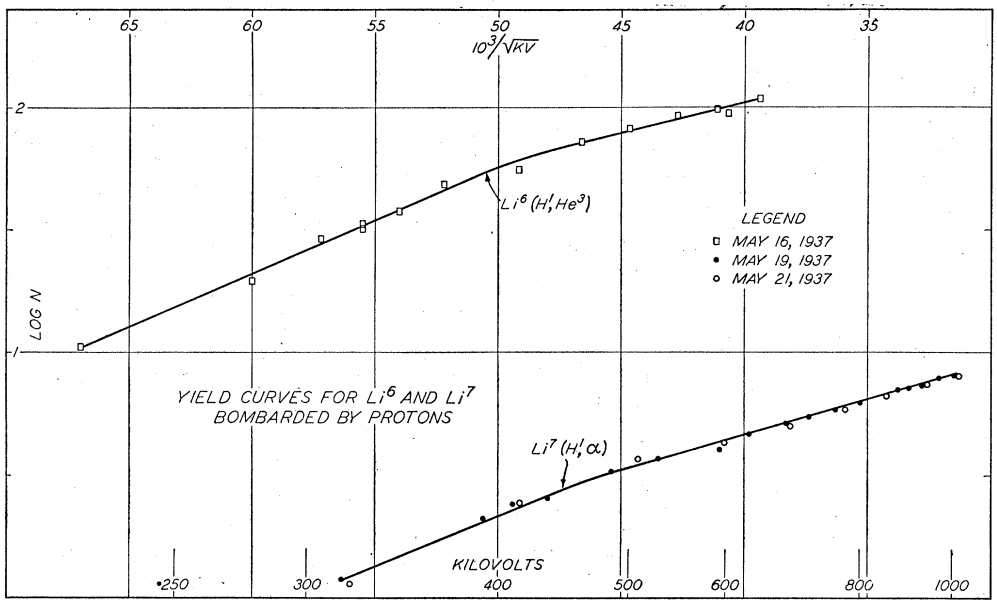

FIG. 1. Yield curves for Li^6 and Li^7 bombarded by protons.

were also eliminated because of the high resolution given by the magnetic field in combination with the slit system. As soon as this arrangement was installed, there was no further difficulty in repeating any observation with agreement better than five percent.

The possibility of changes in the calibration of the voltmeter was eliminated by checking the voltmeter occasionally with a measurement of the lithium gamma-ray resonance at 440 kev. The same measurement applied to the thin targets of Li^7 used for the yield curves showed the thicknesses of the targets and the amount of carbon (contamination) which had accumulated on them.

Despite the magnetic analysis of the ion beam it was possible, of course, to have molecular hydrogen present in the mass 2 spot. Its presence readily could be detected and its percentage estimated by observing the current carried by the mass 1, 3, and 4 spots. This error was never serious as the mass 2 spot was almost invariably better than 95 percent deuterons.

Confusion due to contamination by carbon or oxygen on the target was easily avoided in most cases since the characteristic groups of charged particles arising from these elements are well known. In counting the beta-rays from lithium, however, it was necessary to use a coincidence pair of Geiger-Müller counters shielded by lead to avoid spurious effects from x-rays produced in the tube and from beta- and gamma-rays from carbon. Carbon neutrons also had an appreciable effect when counting neutrons at the higher voltages.

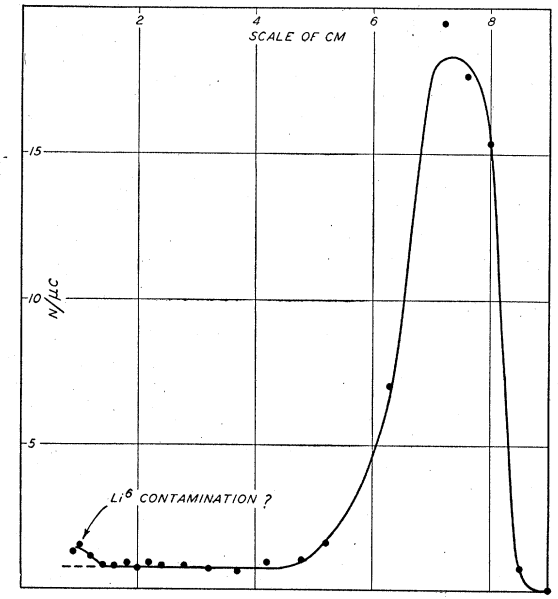

FIG. 2. Alpha-particles from Li^7+H^1.

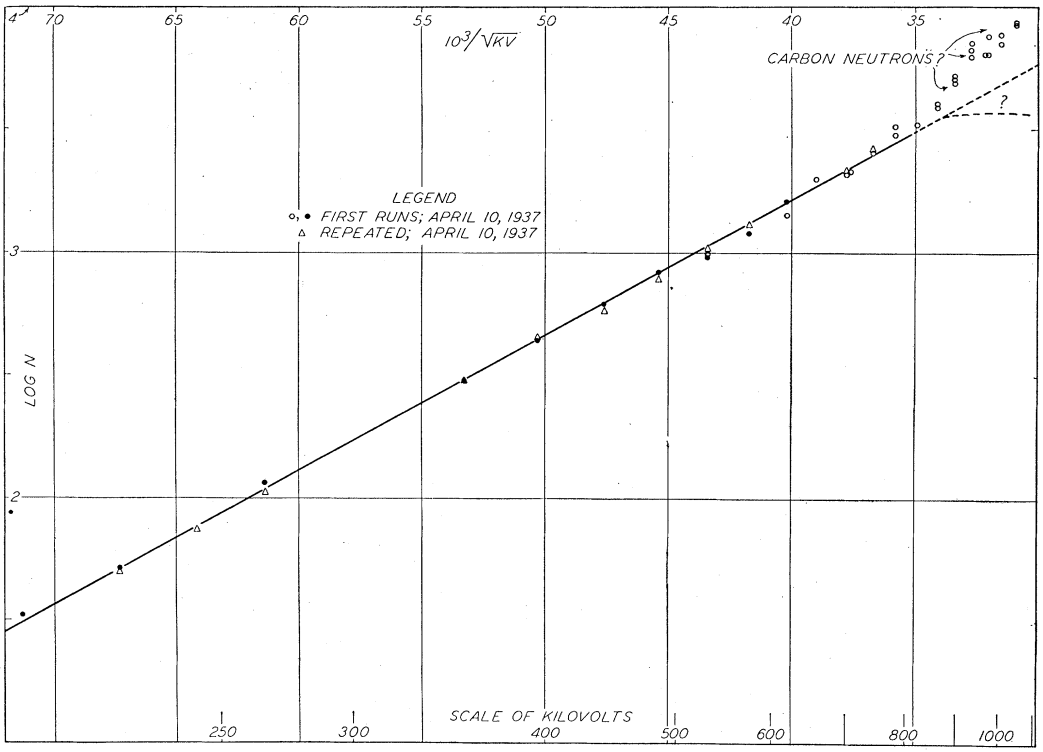

FIG. 3. Neutrons from Li^6+D^2.

C. YIELD CURVES

(1) Li^6+H^1

Figure 1 shows the yield curves for the proton produced reactions in lithium. The yield of the reaction $Li^6+H^1 \rightarrow He^3+He^4$ was measured with an ionization chamber by counting He^3 particles emitted from a thin isotopic target. It was impossible to carry the investigation to voltages higher than 600 kv because the range of the scattered protons there became sufficient to interfere with the counting of the He^3 particles. A thin isotopic target was used.

(2) Li^7+H^1

Thin targets of ordinary lithium, as well as thin Li^7 targets, were used in investigating the yield curve for the reaction $Li^7+H^1 \rightarrow He^4+He^4$. The alpha-particles from this reaction have a range of about eight cm, so all alpha-particles of ranges greater than six cm were counted with an ion chamber connected to a counting system set at medium bias.

The resulting yield curve, shown in Fig. 1, is in satisfactory agreement with the yield functions which have been published previously.[3, 4] The possibility that short-range alpha-particles are emitted in the reaction $Li^7+H^1 \rightarrow He^4+He^4+h\nu$ was investigated by the use of a Li^7 target to eliminate the short-range particles from Li^6+H^1. The range-number curve (Fig. 2) was taken at 475 kv, and the Li^7 target was sufficiently thick to insure the observation of any contribution from the 440 kev resonance. No evidence for a reaction emitting short-range alpha-particles was found.

The weak peak at 1.2 cm was ascribed to a Li^6 contamination of roughly one part in 500, since the cross section for the Li^6+H^1 reaction is approximately 30 times greater than for Li^7+H^1. While the contamination assumed is about 40 times greater than that measured for the thin isotopic lithium targets, it was expected to be large in the present instance because of the very high deposition current (0.6 milliampere) used in

[4] R. G. Herb, D. B. Parkinson, and D. W. Kerst, Phys. Rev. **48**, 118–124 (1935).

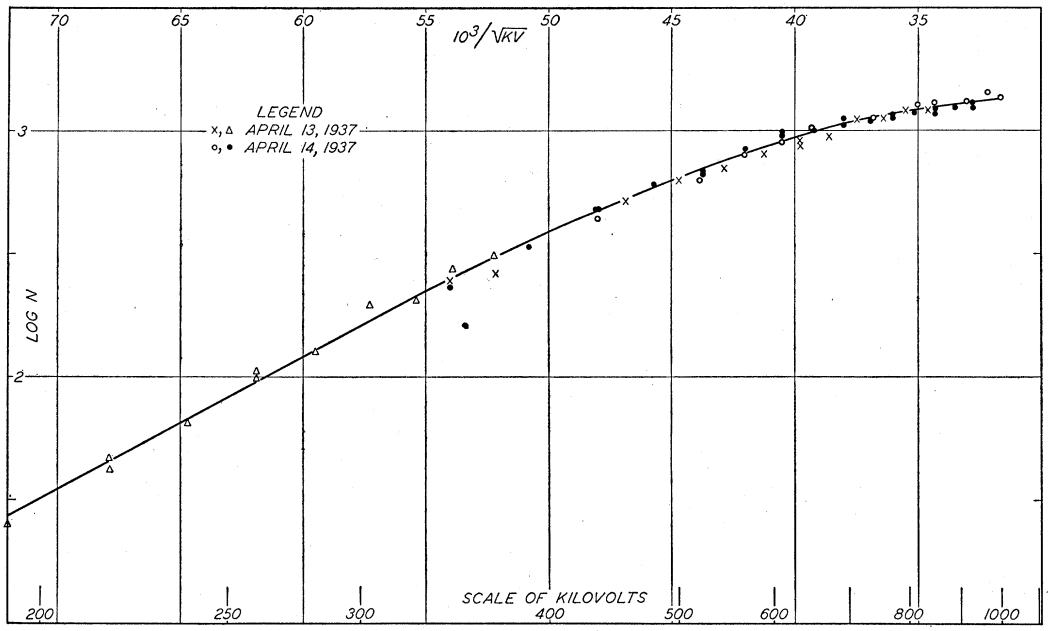

FIG. 4. Alpha-particles from Li^6+D^2.

collecting the thick Li^7 target in the mass spectrometer.

(3) Li^6+D^2

The yield of neutrons from a thin Li^6 target is shown in Fig. 3. The target was placed at the center of a large block of paraffin and the neutrons were counted in a boron lined ionization chamber. It was believed that neutrons from carbon deposited on the target made an appreciable contribution to the number of neutrons observed at the higher voltages (over 800 kv). The effect of the carbon contamination was negligible below 800 kv because of the very steep excitation curve for the production of carbon neutrons. A qualitative method of correcting for the carbon effect above 800 kv is indicated below, in the discussion of the neutrons from Li^7, where a similar carbon effect is estimated by recourse to the alpha-particles which are emitted in the same reaction as the Li^7 neutrons.

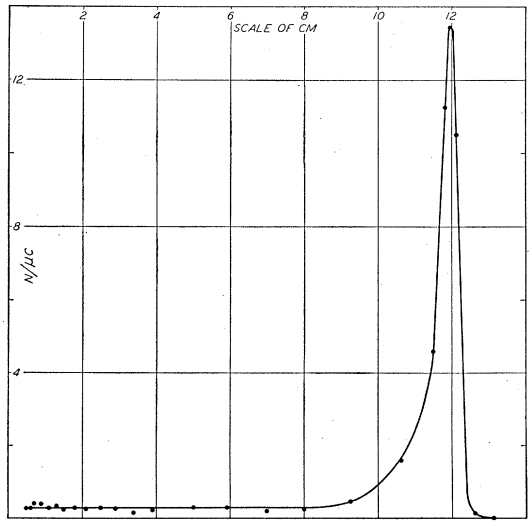

FIG. 5. Alpha-particles from Li^6+D^2, high counter bias.

Figure 4 shows the voltage-yield curve measured for the reaction $Li^6+D^2 \rightarrow He^4+He^4$. A thin isotopic target was used in order to minimize any contaminations, which otherwise would be more abundant by amounts corresponding to the proportional increase in useless Li^7. The alpha-particles were counted in an ionization chamber and the thyratron counter was biased to discriminate against protons. The same isotopic target was used in extending the number-range curve for Li^6+D^2 through the region ordinarily obscured by alpha-particles from Li^7+D^2. Fig. 5 indicates that reactions of the type $Li^6+D^2 \rightarrow He^4+He^4+h\nu$ are highly improbable.

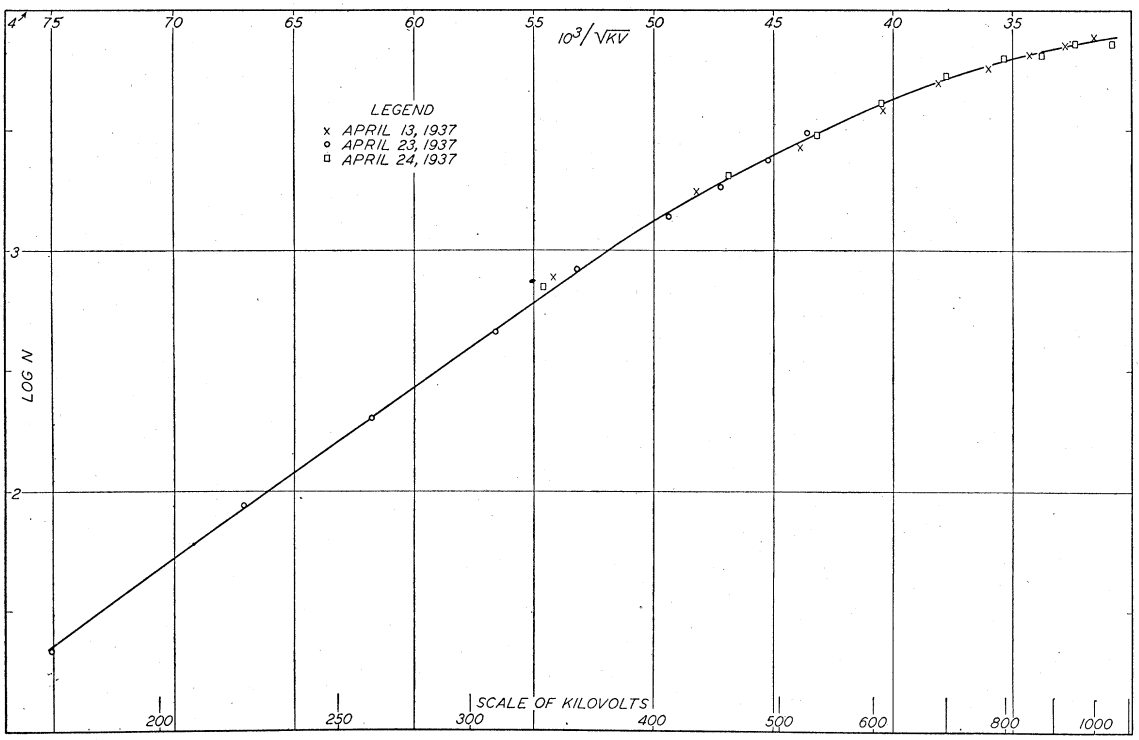

FIG. 6. Total protons from Li^6+D^2.

The total yield of protons from the two processes $Li^6+D^2\rightarrow Li^7+H^1$ and $Li^6+D^2\rightarrow Li^{7*}+H^1$ is shown in Fig. 6 as a function of the deuteron energy. The thyratron counter was biased to count both fast and slow protons, and the ionization chamber was adjusted for ranges sufficiently short to include both Li^6 proton groups but sufficiently long to stop the alpha-particles from Li^6 and the proton groups from carbon and oxygen which comprised the only appreciable charged particle groups to be avoided.

The relative yields of the two Li^6 proton groups at various deuteron bombarding energies, as given in Fig. 7, indicate clearly that the relative probability of forming Li^7 in an excited state increases at higher bombarding voltages. The gradual increase in this probability observed by Williams, Shepherd, and Haxby[5] from 100 to 225 kv continues through the interval from 200 to 500 kv but becomes more rapid for deuteron energies above 500 kev. The mean energy separation of the two states of Li^7, as measured from the proton groups of Fig. 7, is 455 ± 15 kev.

[5] J. H. Williams, W. G. Shepherd, and R. O. Haxby, Phys. Rev. **52**, 390–396 (1937).

(4) Li^7+D^2

The yield of neutrons from deuteron bombardment of Li^7, shown in Fig. 8, was measured by counting neutrons and by counting alpha-particles produced in the same reaction, $Li^7+D^2\rightarrow He^4+He^4+n$. The alpha-particles were counted in the usual ionization chamber with the thyratron counter lightly biased to record all alpha-particles, both fast and slow. The alpha-particles produced in the radioactive disintegration of Li^8 were avoided by placing the ionization chamber at a range of four cm where the relative numbers of these alpha-particles become negligible. A thin Li^7 target was used in order to escape the uncertainty of corrections for the 13-cm group of alpha-particles emitted from Li^6. In counting the neutrons the target was surrounded with paraffin and the neutrons were detected in a boron lined ionization chamber.

These two independent methods of measurement gave identical results for bombarding energies between 200 and 770 kev, but at bombarding energies above 770 kev the alpha-particle yield ceased increasing and no longer corresponded to

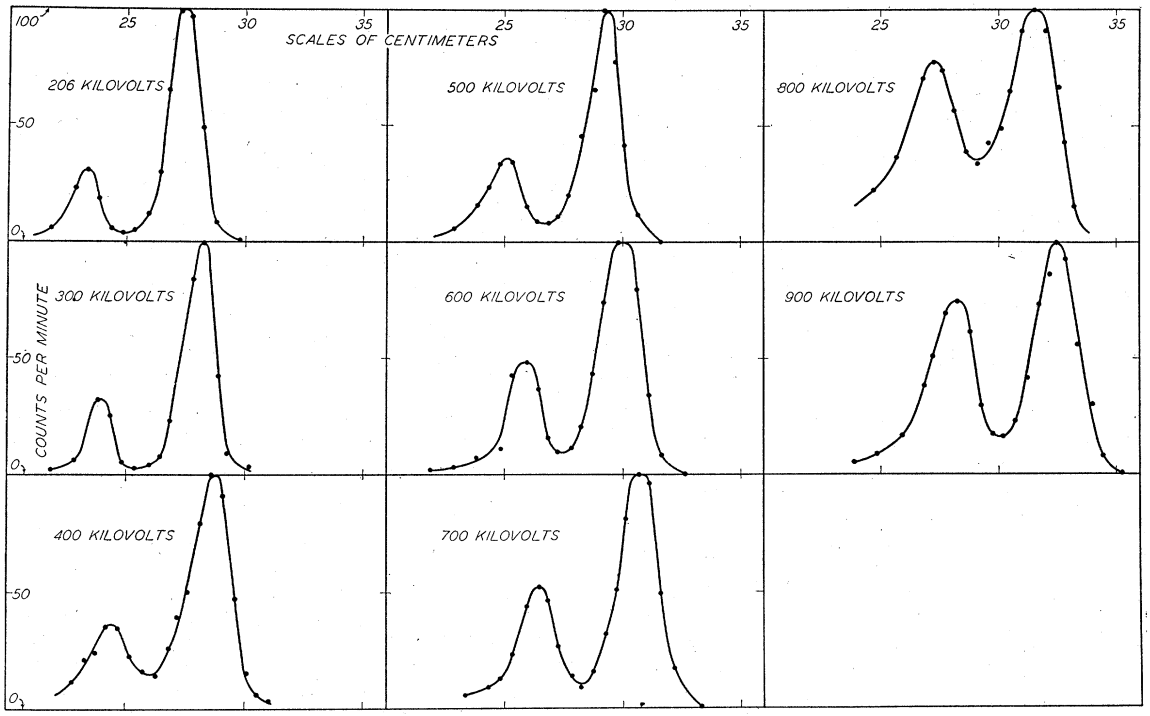

FIG. 7. Proton doublet from Li^6+D^2.

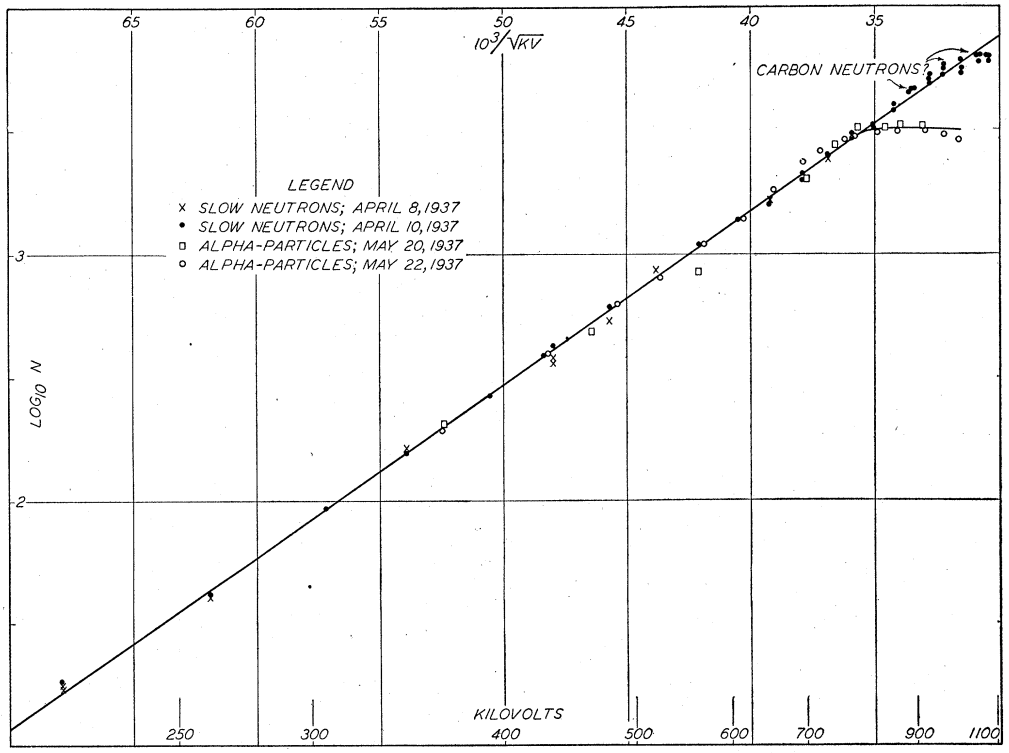

FIG. 8. Neutrons from $Li^7+D^2=He^4+He^4+n$.

the neutron yield. This discrepancy must be ascribed to the production of neutrons in some other process than the one mentioned above. One possible explanation is that the reaction $Li^7+D^2 \rightarrow Be^8+n$ only becomes relatively probable at deuteron energies above 770 kev since, according to Bonner and Brubaker,[6] this reaction contributes only a small fraction of the total lithium neutrons at voltages below 800 kv. A much more plausible explanation is that the excess observed above 800 kv was due to neutrons produced in carbon contamination on the target. The similar effect observed above 800 kv in the yield curve for neutrons from Li^6 indicates that the latter explanation is the correct one.

Figure 9 shows the yield curve for the production of radioactive Li^8 which was measured by counting the alpha-particles as well as the beta-particles released in the decay of Li^8. A double coincidence pair of Geiger-Müller counters was employed in counting the beta-particles. The counters were shielded with lead so that the background count remained very low even when the high voltage was applied to the tube. The counters viewed the lithium target through a small window in the lead covered with Dow metal to screen out the less energetic beta-particles from the carbon contamination on the target. The solid angle subtended by the counters was adjusted to give counting rates such that the correction for the recovery time of the counters (about 10^{-5} sec.) was negligible. The resolving time for coincident discharges was about 5×10^{-6} second. The effect of the carbon contamination was shown to be negligible by the fact that no increase in the background counting rate was observed after the target had been bombarded for some time. The effect of gamma-rays from carbon also was proved negligible by comparing continuous counting rates (that is, rates during bombardment) with the numbers of counts observed in five-second intervals after bombardment.

The alpha-particles emitted by Li^8 can be observed effectively only if some arrangement is made to separate them from the instantaneously emitted alpha-particles from the reaction $Li^7+D^2 \rightarrow He^4+He^4+n$. This separation was accomplished by using a target deposited on a wheel which was rotated during the bombardment. Only those alpha-particles were observed which were emitted after the wheel had rotated 90° from its position during bombardment. A thin target of ordinary lithium was used since a separate experiment proved Li^6 did not emit any delayed alpha-particles. The particles were counted with the usual ionization chamber. Counting rates also were taken with the wheel at rest to measure the background of neutron recoils.

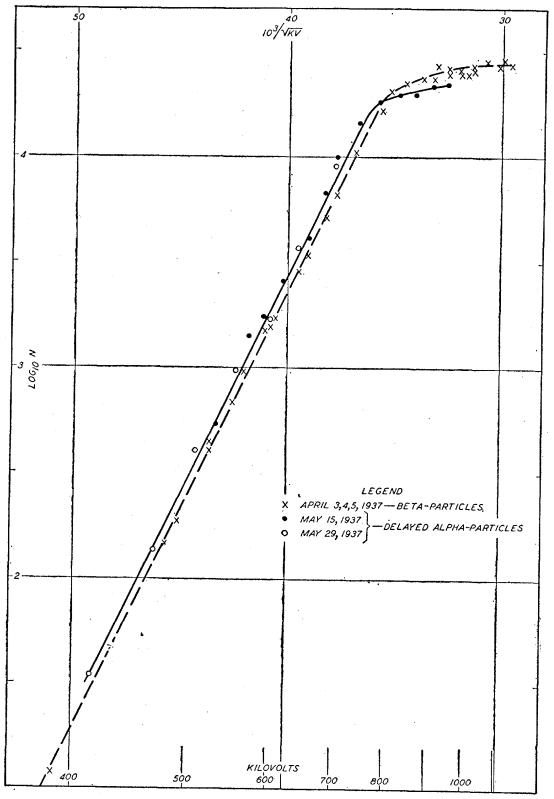

FIG. 9. Yield of delayed particles from Li^7+D^2.

All points of the yield curves represent 1000 or more counts, except for a very few at the lowest voltages. Because of the great variations in the yields with incident particle energy, it was sometimes necessary to change the solid angle of the detector as well as to vary the current to maintain usable counting rates. The curves were fitted at all points at which it was necessary to change the solid angle. In most cases the two portions of the curve have a large overlap. Different portions of the curves, taken with different apertures, are

[6] T. W. Bonner and W. M. Brubaker, Phys. Rev. **48**, 742–746 (1935).

indicated on the curves by different symbols for the points.

D. Relative Reaction Cross Sections for the Lithium Reactions

The curves shown above give only the forms of the yield curves, since the observed number of particles is plotted against energy. For many theoretical considerations a knowledge of relative reaction cross sections of different reactions, as shown in Fig. 10, is of value, so a special effort was made to obtain dependable relative yields by a system of intercomparisons carefully chosen to avoid conflicting groups. In order to obtain the ratio

$$\frac{Li^7+H^1\rightarrow He^4+He^4}{Li^7+D^2\rightarrow He^4+He^4+n}$$

for instance, it was necessary to use an isotopic Li^7 target to avoid counting the 13-cm alpha-group from Li^6 and further to make the observation at a voltage so low that the contribution due to the delayed alpha-particles from the reaction $Li^7+D^2\rightarrow Li^8+H^1$ was negligible. For this reason a bombarding voltage of 380 kv was chosen. All alpha-particles of ranges greater than eight mm in the continuous distribution were counted. The ratios

$$\frac{Li^7+H^1\rightarrow 2He^4}{Li^6+H^1\rightarrow He^4+He^3}$$

$$\frac{Li^6+D^2\rightarrow 2He^4}{Li^6+D^2\rightarrow Li^7+H^1}$$

$$\frac{Li^7+H^1\rightarrow 2He^4}{Li^6+D^2\rightarrow 2He^4}$$

also were obtained at 380 kv from an ordinary Li target for which Brewer's value of 11.6 for the Li^7/Li^6 ratio was assumed. An isotopic Li^6 target bombarded at 690 kv checked the ratio

$$\frac{Li^6+D^2\rightarrow 2He^4}{Li^6+D^2\rightarrow Li^7+H^1}.$$

This ratio was related to the others by means of the yield curve for the 13-cm alpha-particles given in Fig. 4.

The ratio of the neutron yields

$$\frac{\begin{matrix}Li^7+D\rightarrow Be^8+n\\ \qquad\rightarrow 2He^4+n\end{matrix}}{\begin{matrix}Li^6+D^2\rightarrow Be^7+n\\ \qquad\rightarrow He^3+He^4+n\end{matrix}}$$

was obtained by bombardment at 690 kv of isotopic targets of nearly equal masses under otherwise identical experimental conditions, using paraffin and a boron lined ionization chamber for the detector.

The relative masses of the isotopic targets were determined in two ways, namely, (a) from the integrated collector current in the mass spectrometer in which they were deposited and (b) by taking the alpha-particle ratio

$$\frac{Li^7+D^2\rightarrow 2He^4+n}{Li^6+D^2\rightarrow 2He^4}$$

for the two targets. Then this ratio was compared with all those discussed above.

The number of neutrons from Li^7+D^2 in terms of numbers of Li^7+D^2 continuous alpha-particles was obtained from Bonner and Brubaker's estimate[6] that only five percent of total neutrons from ordinary $Li+D^2$ come from reaction Be^8+n^1. It follows that the rest of the Li^7+D^2 neutrons come from He^4+He^4+n. Though the voltage distribution of the deuterons used by Bonner and Brubaker was unknown and the Li^6 and Li^7 neutron excitation functions are not parallel, correction of their data for Li^6+D^2 neutrons gives no serious uncertainty since $Li^7/Li^6 = 11.6$ in nature.

The internal consistency of the above measurements and cross checks indicated the relative reaction cross sections given in Fig. 10 to be accurate to ± 20 percent between 300 and 750 kv with the exception of the radio-lithium yield.

It proved impossible to isolate the reaction leading to radio-lithium for an accurate determination of the relative yield because of the short life of Li^8 and the presence of continuous alpha-particles from Li^7+D^2. The estimate of the relative yield of radio-lithium given in Fig. 10 is taken from comparisons between the numbers of delayed and total alpha-particles from Li^7+D^2 and may be in error by a factor of two.

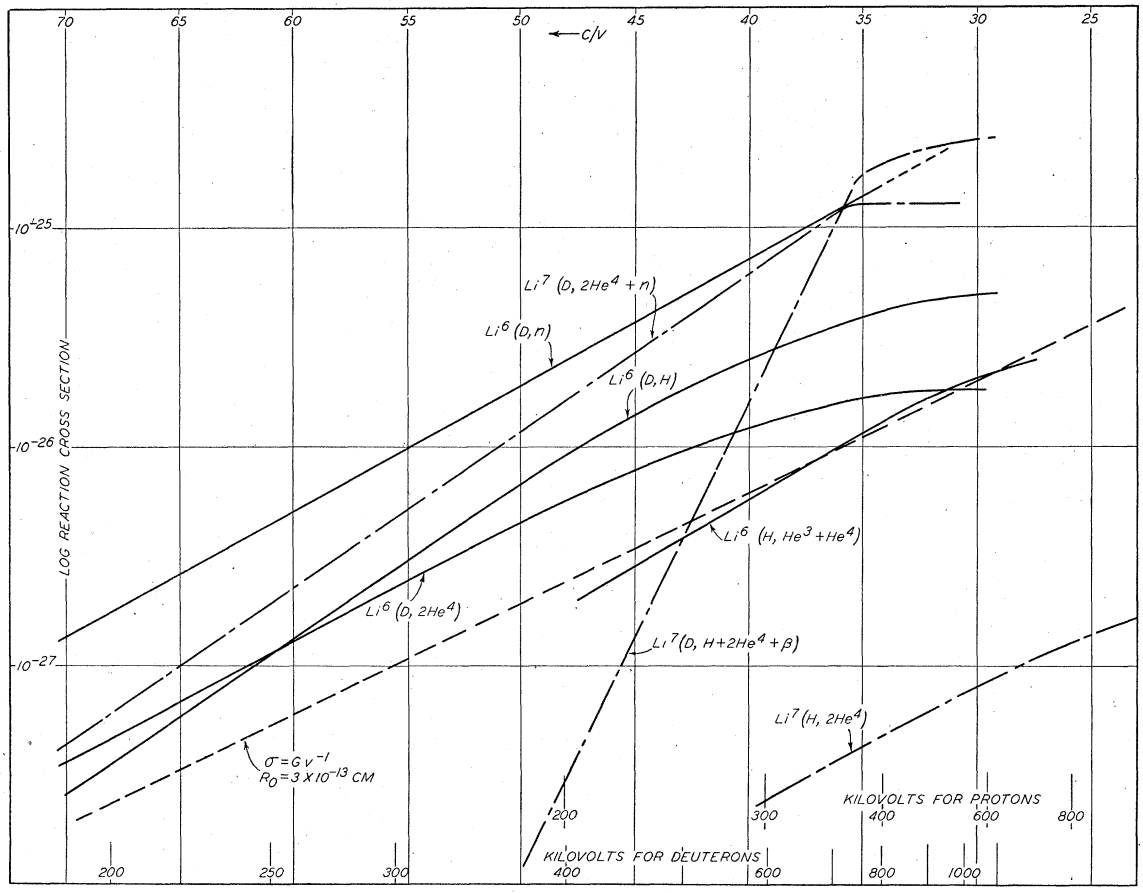

FIG. 10. Yield curves for lithium reactions.

E. ABSOLUTE CROSS SECTIONS FOR REACTIONS

The yield of He^4 from Li^6+D^2 is used to fix the scale of Fig. 8. The cross section scale adopted is the average computed from the masses of two LiOH thin targets analyzed by the National Bureau of Standards, through the kindness of Dr. C. J. Rodden, and from the mass of one Li^6 target measured in terms of collector currents during deposition in the mass spectrometer. Because of difficulty in obtaining absolutely uniform thin deposits, the various targets gave values for the absolute cross sections varying among themselves by a factor of nearly three.

It has been convenient in Fig. 10 to plot the logarithm of the reaction cross section in cm^2 against the velocity of light divided by the velocity of the bombarding particles in order to provide a common scale for the reactions resulting from both proton and deuteron bombardment. Several significant features are obvious in the collected cross section curves.

(1) The familiar exponential-like dependence of the reaction cross section upon the velocity of the incident particles appears in all the curves.

(2) The increase in the yield of Li^8 with incident deuteron velocity is exceptionally rapid. This increase will be discussed separately in connection with the radio-lithium problems of the next section.

(3) The remaining curves may be divided into two groups according to slope so that below 400 kv the rate of increase in cross section is approximately the same function of the incident particle velocity, v, for all the reactions within the same group. The rate for the first group, which consists of the two reactions $Li^6(D^2, H^1)$ and $Li^7(D^2, 2He^4+n)$, is greater than the rate for

the second group of reactions by approximately a factor $v^{\frac{1}{2}}$.

(4) The curves for the two reactions resulting from deuteron bombardment of Li^7 both break rather sharply near 770 kv and their cross sections become comparable.

(5) Deuteron bombardment of either lithium isotope leads to a high neutron yield, but the two neutron yield curves are different functions of v.

(6) The cross section for the reaction $Li^7(H^1, 2He^4)$ is much smaller than for any other reaction considered.

The exponential form of the yield functions may be explained in the usual way by assuming that the governing term resembles the Gamow factor for penetration through the potential barrier, $G \sim \exp(-2\pi\alpha Zzcv^{-1})$, where α is the fine structure constant, Z and z are the atomic numbers of the reacting particles, v is their relative velocity, and c is the velocity of light. The various reactions, excepting the case of radio-lithium, involve penetration of the potential barrier by the incident particle only, since the emitted particles receive energies considerably greater than the barrier heights. Theoretical discussions of reaction cross sections have been given by Gamow,[7] Breit and his colleagues,[8, 9] Bethe,[10, 11] and others. At bombarding energies that are small compared to the barrier heights, the various derived expressions generally are asymptotic to the simplified formula for the cross section, $\sigma \sim Gv^{-2}$, where v^{-2} has the significance of an absolute cross section proportional to the square of the wave-lengths of the incident particles. The simplified formula gives relative cross sections which increase too slowly by, roughly, a factor of v through the intermediate range of bombarding energies (200 to 400 kev) to agree with even the least steep excitation curves of Fig. 10.

An interpretation of the yield curve for the reaction $Li^7(H, 2He^4)$, as measured by Herb, Parkinson, and Kerst,[4] and by Hafstad, Heydenburg, and Tuve,[3] at bombarding energies overlapping those of the present experiments, has been given by Ostrofsky, Breit, and Johnson.[9] They calculated the reaction cross section as proportional to the density of incident protons within a potential well of chosen breadth and depth. Both the rate of increase with incident particle energy and the form of the calculated yield function then became sensitive to the positions of the resulting resonances. However, their expression for the cross section at low bombarding energies simplified to the formula $\sigma \sim Gv^{-2}$ when the density factor was included. On the other hand, a reaction cross section proportional to the flux of incident particles through the nucleus simplified to a prior expression, also given by Breit,[8] $\sigma \sim Gv^{-1}$. This oversimplified formula for the cross section is the one plotted in Fig. 10 for purposes of comparison, since the cross sections observed in the present experiments appear to agree more nearly with this expression.

The final levels available for a given disintegration process include, of course, excited levels

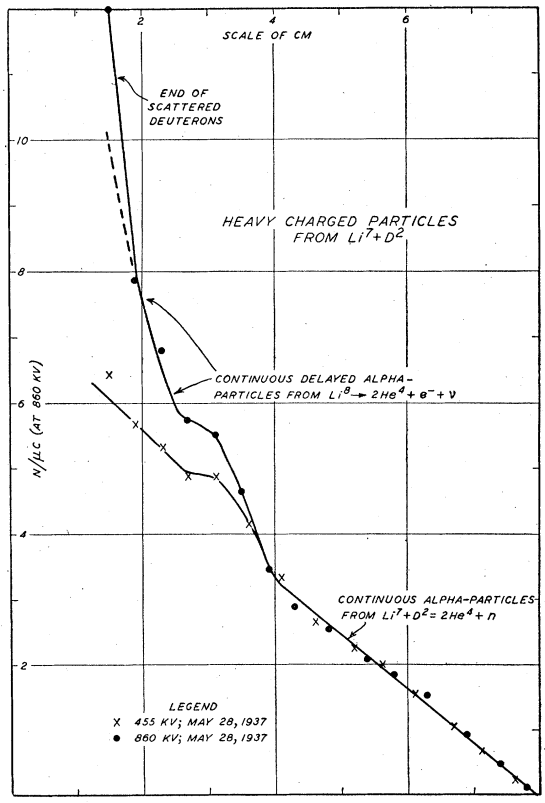

FIG. 11. Heavy charged particles from Li^7+D^2.

[7] G. Gamow, *Atomic nuclei and radioactivity* (Cambridge, 1937).
[8] G. Breit, Phys. Rev. **34**, 817 (1929).
[9] M. Ostrofsky, G. Breit, and D. P. Johnson, Phys. Rev. **49**, 22–34 (1936).
[10] H. A. Bethe, Rev. Mod. Phys. **9**, 186–219 (1937).
[11] E. J. Konopinski and H. A. Bethe, Phys. Rev. **54**, 130–138 (1938).

as well as the ground levels of the disintegration products. Consequently the cross section curves of Fig. 10 in some instances represent the measured sums of the cross sections for more than one final level, since such measurements were the experimentally practical ones. Two states of Li^7 are summed in the cross section curve for the reaction $Li^6(D^2, H^1)$, but the curve for either Li^7 state singly may be obtained by reference to Fig. 7. Only a small part of the steeper initial increase with relative velocity in the total cross section for the reaction $Li^6(D^2, H^1)$ may be attributed to the increased probability of forming Li^7 in the excited state, since the yield of this state relative to the ground state increases but slowly between 200 and 500 kv according to Fig. 7. However, at higher voltages the form of the curve is influenced considerably by the more rapid increase in the relative strength of the excited state. Likewise, in the reaction $Li^7(D^2, 2He^4+n)$ a part of the observed variation of the cross section might be related to accompanying changes in the distribution of the disintegration products among the numerous available final levels, since several known levels cover a range of about 12 Mev, and the bombarding energy was varied by nearly one Mev in obtaining the yield curve. However, there is no evidence of such changes in the distribution of available final states for this reaction since (1) the only discernible variation in the distribution of the associated alpha-particles which occurred when the bombarding energy was varied by 400 kev is actually due to the delayed alpha-particles from Li^8 (see Fig. 11), and (2) the cross-section curve from neutron measurements and that from alpha-particles of ranges greater than four cm appear identical in shape except for the effect ascribed to carbon neutrons at high bombarding voltages (Fig. 8). The final levels for the reaction $Li^6(D^2, n)$ are unknown except that they are perhaps about equally divided between $Li^6+D^2 \rightarrow He^3+He^4+n$ and $Li^6+D^2 \rightarrow Be^7+n$. The number of final levels is probably small because the energy released in the disintegrations is not large. The disintegrations $Li^7+H^1 \rightarrow 2He^4$ and $Li^6+D^2 \rightarrow 2He^4$ seem able to occur only to the ground levels according to Fig. 2 and Fig. 5. Likewise, the reaction $Li^7+D^2 \rightarrow Li^8+H^1$ seems to involve only the ground state of Li^8 since the energy release is small and no gamma-radiation has been detected (see Section F). However, the apparent absence of gamma-radiation is not very conclusive evidence by itself due to the experimental difficulties of detecting it.

The break in the cross-section curves for Li^7 under deuteron bombardment at 770 kv cannot be due solely to the effect of competing reactions in the compound nucleus since both curves break at apparently identical deuteron energies. According to the views of Breit and his colleagues,[9] a high resonance in Be^9 might account for either or both breaks. Such a resonance would need to be located in the neighborhood of $(Li^7+D^2-Be^9)+(7/9)(0.77)$ Mev ≈ 17.2 Mev above the ground state of Be^9, and at the same time would account, at least qualitatively, for the observation that the cross section for the reaction $Li^7(D^2, 2He^4+n)$ increases faster with voltage than do the cross sections for most lithium reactions.

Above 800 kv the cross section for the formation of radio-lithium appears to be somewhat larger than the cross section for neutron emission in Li^7, though this has not been established definitely due to the uncertainty in the beta-ray yield, as described in Section D. Various investigators have identified a considerable number of final levels associated with neutron emission in the reaction $Li^7(D^2, 2He^4+n)$,[6, 12] but radio-lithium appears to be formed only in the ground state (no gamma-radiation). Consequently, it may be inferred that in light nuclei other factors can be more important in determining relative yields of competing reactions than is Bethe's rule[10] that at emission energies above the potential barrier the relative yields of competing reactions are approximately proportional to the relative numbers of final levels available for the reactions. On the other hand, this rule more successfully approximates the relative yields of the competing processes in Li^6 under deuteron bombardment.

It should be noted that at the lower bombarding energies the cross section for neutron emission from Li^6 is sufficiently larger than that from Li^7 to account for an appreciable fraction of all the neutrons observed during deuteron bombardment of ordinary lithium, and such neutrons must be taken into account in studies of the distribution in energy of ordinary Li neutrons.

The comparatively low yield of the reaction

[12] W. E. Stephens, Phys. Rev. **53**, 223–226 (1938).

$Li^7(H^1, 2He^4)$ relative to other lithium reactions has been discussed by Goldhaber,[13] by Breit and his colleagues,[9] and by Konopinski and Bethe[11] in connection with the influence of the orbital momentum of the incident particle upon the probability that it will penetrate the barrier.

F. Radio-Lithium

The reaction usually assumed for the production of radio-lithium, $Li^7+D^2\rightarrow Li^8+H^1+Q_1$, seems the only possible one fitting the experimental facts, although the proton group from the reaction never has been found. During the previous work with isotopic targets, the upper limit of the range of this group had been set at eight cm.[1] Consequently, in the present investigation, a special effort was made to find the proton group superimposed upon the continuous distribution of alpha-particles ending at eight cm from the reaction $Li^7+D^2\rightarrow He^4+He^4+n$.

It is obvious, according to the reaction cross sections of Fig. 10, that such a proton group should be especially conspicuous at bombarding energies of 800 kev or more but should decrease at lower voltages much more rapidly than do the yields of other lithium reactions. Accordingly, range-number curves were obtained at 860 kv and at 455 kv, using a bias on the detector such as to count protons near the ends of their range. These curves are shown in Fig. 11 with ordinates adjusted to bring the two curves into juxtaposition for easy comparison. There is no proton group of range greater than 1.7 cm associable with Li^8, since the two curves apparently are identical except for the effect at shorter ranges on the 860-kv curve due to the delayed alpha-particles from short life radioactive disintegration of Li^8. This effect readily could be estimated, either from the results of Fig. 10 and Fig. 13, or, independently, by observing the number of delayed alpha-particles after interrupting the bombarding beam of deuterons. The proton group sought should appear in numbers large compared to the delayed alpha-particles of Fig. 11, being in equilibrium with the delayed alpha-particles of all energies after a few seconds' bombardment. The group at 3.5 cm clearly is not due to the formation of Li^8 for (1) it is far too weak, (2) it still persists at 455 kv whereas the yield of Li^8 has decreased by a factor of 100 according to Fig. 10, and (3) it appears to be a broad alpha-particle group belonging to the reaction $Li^7+D^2\rightarrow He^4+He^4+n$.

[13] M. Goldhaber, Proc. Camb. Phil. Soc. **30**, 561–566 (1934).

Mass of Li^8

The 860-kv curve of Fig. 11 can be used to fix an upper limit for the energy release, Q_1, in the formation of Li^8. Since the energy of the bombarding deuterons was 0.860 Mev and since 1.7 cm protons have an energy of 0.830 Mev, it follows that Q_1 is less than $(1+\frac{1}{8})(0.830)-(1-2/8)(0.860)=0.290$ Mev. A lower limit on Q_1 can be set by the fact that disintegrations of radio-lithium were observed for bombarding energies as low as 360 kev. The kinetic energy of the incident deuteron available to the center of mass system is, at the latter bombarding voltage, $(\frac{7}{8})(0.360)=0.280$ Mev, so Q_1 is greater than -0.280 Mev since the escaping proton must have positive kinetic energy. In fact, the exceedingly low yield of the reaction at 360 kev suggests that Q_1 is considerably nearer to -0.280 Mev than to $+0.290$ Mev.

A better value for Q_1, and consequently for the mass of Li^8, can be obtained as follows. Assuming that the proton is emitted with very low energy, it can escape the system only by penetrating out through the potential barrier. Hence, two penetration factors must be included in computing the cross section for the reaction. Using the approximate formula due to Breit[8] we have

$$\sigma \cong K v_D^{-1} \exp(-2\pi 3\alpha c v_D^{-1}) \exp(-2\pi 3\alpha c v_p^{-1}),$$

where v_D and v_p are the relative velocities for the deuteron and for the proton, and c and α are the velocity of light and the fine structure constant, respectively. The quantity v_p is unknown but is related to v_D by the momentum and energy relations which give

$$E_p=9/8Q_1+7/8E_D,$$

where E_p and E_D are the kinetic energies of the proton and deuteron. By means of these equations, σ can be calculated as a function of E_D for various values of Q_1 and compared with the cross-section curve of Fig. 9 in the range 400 to 600 kv. In this way, the best fitting value of Q_1 was found to be -0.200 ± 0.010 Mev. It is not certain

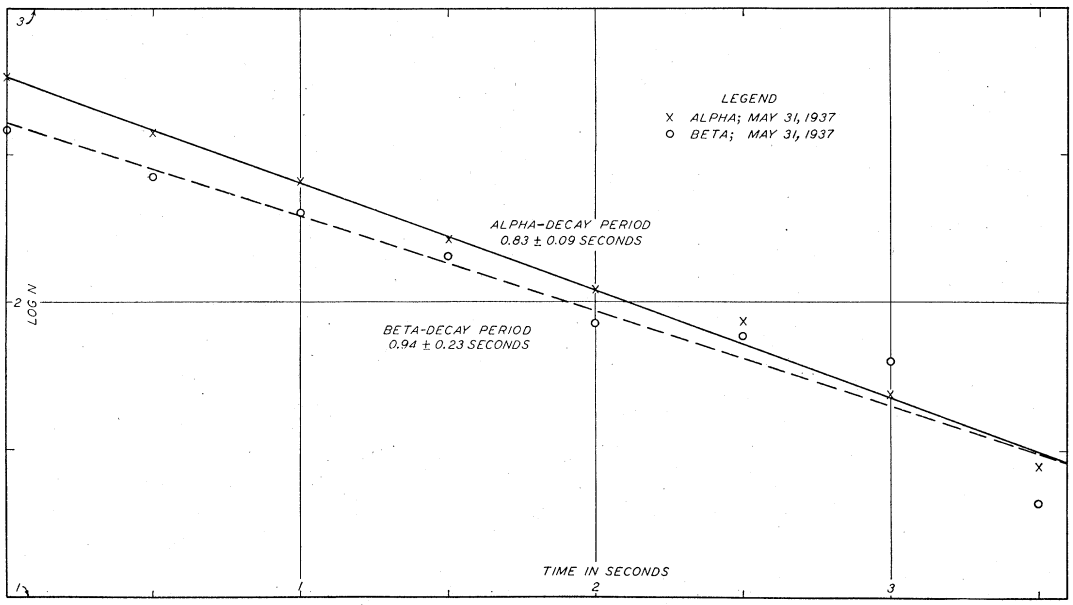

FIG. 12. Periods of alpha- and beta-particles from Li^8.

that the factor $1/v_D$ is correct in this application and the observed form of the cross-section curve probably is influenced by resonances, as discussed in the preceding section. In spite of this uncertainty, it is very probable that Q_1 is -0.200 ± 0.030 Mev, because the double penetration factor is by far the more important in the formula. When Livingston and Bethe's masses for Li^7, D, and H are used, this value of Q_1 gives a mass of 8.02499 for Li^8. The error in this mass principally depends upon the error in the mass of Li^7.

Disintegration of Li^8

An effort to detect gamma-rays from Li^7 bombarded by deuterons was unsuccessful, and a similar result has been reported by Bayley and Crane.[14] Hence it may be assumed that, of the possible reactions[1] in the radioactive decay of Li^8, only the reaction $Li^8 \rightarrow 2He^4 + e^- + Q$ is of importance. This conclusion is supported by the fact that the number of (delayed) alpha-particles of range greater than six mm is slightly greater than (1.1 times) the total number of beta-particles observed. Corrections for alpha-particles of shorter range and for the geometry of the beta-counter[15] would tend to make this ratio closer to two, which is the factor necessary if Li^8 always disintegrates with alpha-emission. Furthermore, the facts that the two yield curves taken with beta-particles and with alpha-particles are identical (Fig. 9) and that the decay periods, determined for both kinds of particles by least-square adjustment, are the same within the experimental error (Fig. 12) also suggest that the alpha-particles and the beta-particles are emitted in the same reaction. As a final check numerous cloud chamber photographs were obtained of the simultaneous emission of two charged heavy particles of equal ranges in approximately opposite directions from a very thin film of lithium previously bombarded by deuterons. Since the heavy particles therefore have equal masses and equal charges and were observed to behave like doubly

[14] D. S. Bayley and H. R. Crane, Phys. Rev. **52**, 604–609 (1937).

[15] The relative numbers of alpha- and beta-particles were obtained by counting the delayed particles of each type where they emerged through a one-mm hole in a six-mm thick lead plate at the end of a tube about eight cm distant from a thin target. Even the use of turned lead baffle plates, carefully spaced and aligned, apparently did not eliminate excessive counts due to the "piping" of beta-particles by small angle scattering. The beta-particle yields obtained in this manner did not agree with those obtained with more distant beta-counters viewing a wide-windowed target chamber through air when no alpha-particles were being counted.

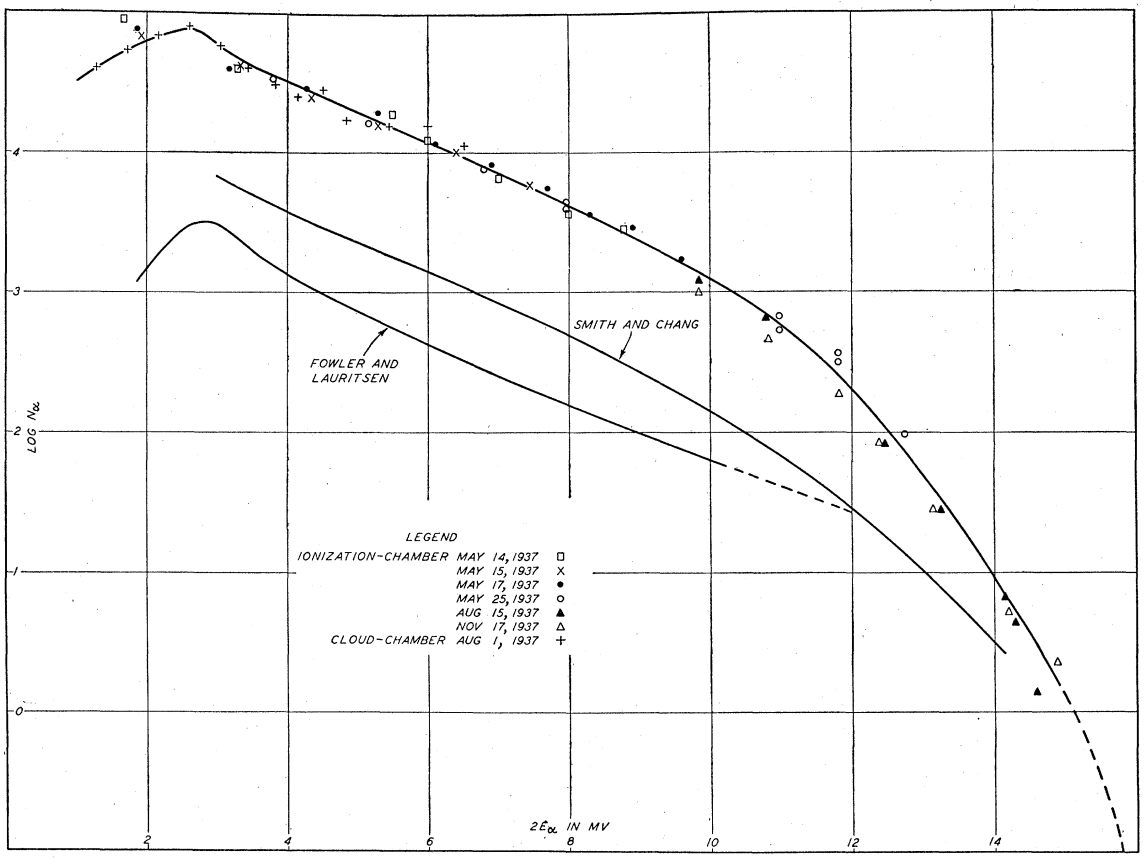

FIG. 13. Li^8 alpha-particle distribution curve.

charged particles in an ion chamber, it follows that they can be only He^4.[16]

Alpha-particles from Li^8

Breit and Wigner[17] have suggested that the radioactive disintegration of Li^8 is associated with the emission of alpha-particles from a rather broad excited state in the neighborhood of three to five Mev in Be^8, and a somewhat similar explanation has been advanced by Kronig.[18] The most crucial test of such a hypothesis should be given by the form of the energy distribution curve for the delayed alpha-particles. Three different methods were found necessary to observe these particles effectively at short, intermediate, and long ranges because of their wide variations in number and range.

An ion chamber in combination with the thin lithium target deposited on a rotating wheel (described above) was suitable for observing alpha-particles of intermediate range. The number-energy distribution at constant bombarding voltage was readily measured for ranges between one and four cm with the aid of this device and showed a decrease in intensity by a factor of over 100 as four cm was approached. At greater ranges the rotating wheel proved to be too inefficient since only a small fraction of the delayed alpha-particles was emitted in front of the detector.

At the longer ranges, an intermittent rotator, devised by our colleague R. C. Meyer, was used to measure the alpha-particles. In this apparatus two targets fixed on opposite ends of a rod about 25 cm long alternately were rotated into position in the bombarding beam and in front of the detector. The magnetic coupling to the driving motor was so arranged that the targets were in

[16] See also J. C. Bower and D. P. R. Petrie, Proc. Camb. Phil. Soc. **33**, 534–539 (1937).
[17] G. Breit and E. Wigner, Phys. Rev. **51**, 593 (1937).
[18] R. de L. Kronig, Physica **4**, 171–174 (1937).

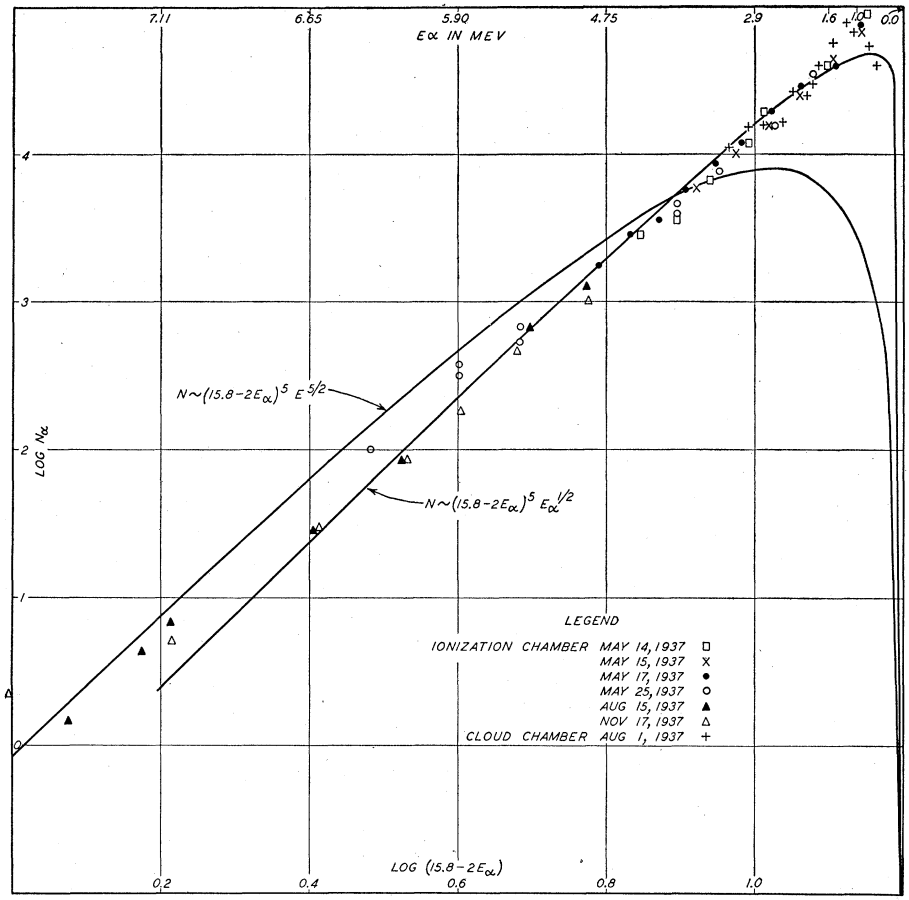

FIG. 14. Li^8 alpha-particle distribution curve, comparison with Gamow-Teller theory.

position for roughly one second and then rapidly exchanged in a fraction of a second. By means of this apparatus it was possible to fix the end point of the distribution of delayed alpha-particles at 15.8 ± 0.5 Mev, the large error mainly being due to the manner in which the alpha-particle intensity approaches zero.

In the low energy region the determination of range-number distributions by electrical counting methods is unsatisfactory because of the short ranges of the emitted particles. If the counter is set to differentiate sharply the effective range is reduced, and if differentiation is not used a maximum in a distribution appears only as an inflection. Though an inflection at short ranges was observed during the present experiments by using a counter without electrical differentiation, not enough points could be obtained on the low energy side of the inflection to establish its position with any certainty. These observations merely check qualitatively a better determination made with a cloud chamber. The use of windows in the cloud chamber was avoided by mounting a thin target on an arm which could be moved from the outside. The target proper was a lithium film deposited on a foil of one mm of air equivalent thickness. This target could be swung into a side tube of the chamber where it was bombarded by deuterons entering through a metal foil. The arm was moved to the center of the cloud chamber before expansion, thus permitting the tracks of all charged particles taking part in a disintegration to be photographed, without any intervening windows or wall effects to mar observation of the shortest tracks. The cloud chamber observations showed a definite maximum at about seven mm air equivalent in the range distribution but this maximum was less

sharp on the low energy side than that found by Fowler and Lauritsen.[19] The observations of Smith and Chang[20] do not show this maximum, but it would not be expected to appear in their experiments since they used a counter technique.

The energy distribution resulting from observations covering an intensity range of nearly 10^5 is plotted in Fig. 13 with the Pasadena and the Cavendish results shown for comparison. This distribution curve indicates that the excited level in question lies between 2.5 and 3 Mev rather than at the higher value previously reported from the Cavendish.[21] The lower value agrees well with the excited level of Be^8 found by Dee and Gilbert[22] in the disintegration of boron.

Discussion

Gamow and Teller[2] have pointed out that the main features of the distribution curve can be explained as a four-body disintegration by considerations based on the Fermi theory of beta-neutrino decay. According to this view the equation for the disintegration is

$$Li^8 \rightarrow He^4 + He^4 + e^- + v + Q_2,$$

where v signifies the neutrino. The disintegration energy, Q_2, is divided between the energy of the light particles, $E_L = E_e + E_v$, and the energy of the two alpha-particles which is $2E_\alpha$ since the two heavy particles must receive nearly equal energies if momentum is to be conserved. Then

$$Q_2 = 2E_\alpha + E_e + E_v = 2E_\alpha + E_L.$$

The probability of beta-decay, $N(E_L)$, is proportional to E_L^5 in the Fermi theory and therefore to $(Q_2 - 2E_\alpha)^5$, but in the present case there is an additional factor $E_\alpha{}^n$ describing the separation into two alpha-particles, where n is $\frac{1}{2}$ or $5/2$ corresponding to alpha-particles of angular momenta zero or 2. Consequently, on the basis of the Fermi theory, the number-energy distribution function for the alpha-particles is given by $N(E_\alpha) \sim (Q_2 - 2E_\alpha)^5 E_\alpha{}^n$, since $N(E_L)$ must be proportional to $N(E_\alpha)$ if charge is to be conserved.

The observed number-energy distribution of the delayed alpha-particles has been replotted in Fig. 14 using as abscissa the logarithm of the energy $(Q_2 - 2E_\alpha)$ available to the light particles. It is seen that the Gamow-Teller formula with $n = \frac{1}{2}$ agrees fairly well with the experimental points except in the region of low alpha-particle energies, which indicates that the effect of the potential barrier must be included before a calculation of the above type can be made exact. The Konopinski-Uhlenbeck beta-neutrino field would predict a seventh power dependence on the energies of the light particles, in marked disagreement with the observed distribution.

Consequently, it appears that the Fermi beta-theory should account satisfactorily for most features of the observed distribution of alpha-particles from the radioactive decay of Li^8. However, it is important to emphasize that in the region of the high energy alpha-particles (where the energy available to the light particles becomes small) the definiteness of agreement with the Gamow-Teller calculations becomes quite sensitive to the values of $(Q_2 - 2E_\alpha)$. $Q_2 = 15.8$ Mev, the observed end point of the distribution of delayed alpha-particles, is used in Fig. 14.

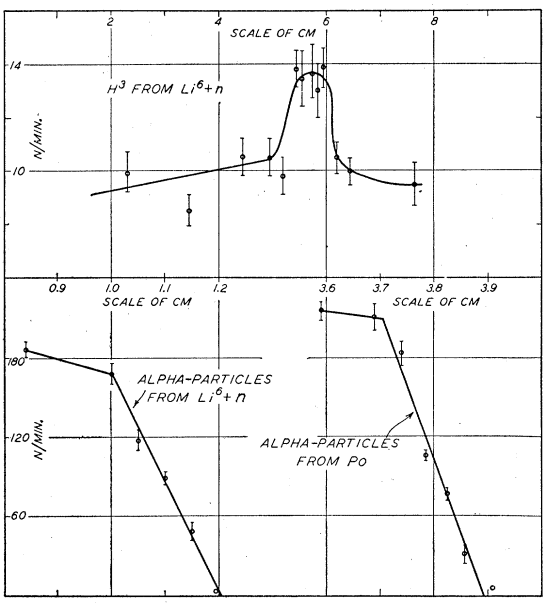

FIG. 15. Range curves for particles from $Li^6 + n$.

[19] W. A. Fowler and C. C. Lauritsen, Phys. Rev. **51**, 1103 (1937).
[20] C. L. Smith and W. Y. Chang, Proc. Roy. Soc. **166**, 415–424 (1938).
[21] W. B. Lewis, W. E. Burcham and W. Y. Chang, Nature **139**, 24 (1937).
[22] P. I. Dee and C. W. Gilbert, Proc. Roy. Soc. **154**, 279–296 (1936).

The value of Q_2 may be computed on the basis of the observation that Li^8 disintegrates into two alpha-particles without gamma-radiation, if negligible rest mass for the neutrino is assumed. Direct computation from the mass of Li^8, as determined above, and from the mass of He^4, gives $Q_2=16.03$ Mev to which the uncertainty in the mass of Li^7 alone contributes about 0.170 Mev in probable error. It seems better to compute Q_2 by taking the mass of Li^7 as equivalent to $2He^4-H^1+17.13$ Mev. Subtracting the equation $Li^7+H^1=He^4+He^4+17.13\pm0.06$ Mev from $Li^7+D^2=Li^8+H^1-0.20\pm0.03$ Mev, gives $Li^8=D^2-2H+2He^4-17.33$ Mev. Then, since $Li^8=2He^4+Q_2$, $Q_2=D^2-2H^1+17.33$ Mev$=15.90$ Mev, where Bainbridge and Jordan's[23] value for the separation of the mass-spectrographic doublet ($H_2{}^1-D^2$) is used. It is, of course, pointless to attempt to use the less accurate end point of the alpha-particle distribution from Li^8 as a check on any of the above values.

G. Disintegration of Lithium by Neutrons

Two thin, square isotopic deposits, 2.5 cm on a side, were prepared upon flat silver plates especially for study of the disintegration of lithium by slow neutrons. One target contained 400 micrograms of Li^6 and the other 4600 micrograms of Li^7. These targets were interchangeable in a target holder located close to a neutron source at the center of a large block of paraffin in order to obtain a maximum flux of slow neutrons through the lithium.

The neutron produced disintegration particles from the lithium were observed by means of a linear amplifier connected to a broad but shallow ion chamber whose face was parallel to the lithium target and target holder. The face of the chamber carried a collimator made from a plate five mm thick drilled with $\frac{1}{2}$-mm diameter holes closely spaced in hexagonal pattern, so that only those disintegration particles whose paths lay within 6° of the normal to the target could enter the ion chamber. The chamber could be moved with respect to the target by means of an attached screw and each setting was read with a micrometer.

[23] K. T. Bainbridge and E. B. Jordan, Phys. Rev. **49**, 883 (1936).

Li^6+n

The alpha-particles and the H^3 particles emitted in the disintegration of Li^6 by neutrons, according to the reaction $Li^6+n\rightarrow He^4+H^3$, were observed with this apparatus. The neutron source, equivalent in strength to 7000 millicuries of (Rn − Be), was produced by bombarding a graphite plate with ten microamperes of 1-Mev deuterons.[24] Carbon neutrons produced in this manner are of relatively low energies and, through the consequent elimination of high energy recoil particles, give only a fraction of the background effects of other neutron sources. When the electrical counter was biased to count H^3 particles near the end of their range, the background was about equal to the effect produced by the H^3 particles, but the H^3 peak was still clearly defined; when the counter was biased to count only alpha-particles, the background was negligible. The range-number curves for both types of particles are plotted in Fig. 15. The alpha-particles are more suitable for an accurate determination of the energy released in the disintegration, since their energy-range relation is well established in the range interval under consideration. The less accurately measurable range and energy of H^3 are chiefly useful for determining the disintegration which occurs and for checking the experimental value of the reaction energy obtained from the alpha-particle measurements.

Immediately after the range-number curve was taken for the alpha-particles from Li^6, their range was compared directly with the range of polonium alpha-particles under identical conditions of geometry and counter bias by replacing the Li^6 target with a clean polonium source prepared on a silver square similar to that carrying the lithium. This comparison gave 2.13 Mev as the energy of the alpha-particles from Li^6 exposed to slow neutrons, when the Cornell range curves for alpha-particles (1938) were used and 3.842 cm was assumed as the mean range of the polonium alpha-particles.[25] The corresponding reaction energy is then $(7/3)(2.13)=4.97$ Mev, in sufficient agreement with the

[24] E. Amaldi, L. R. Hafstad and M. A. Tuve, Phys. Rev. **51**, 896–912 (1937).

[25] M. G. Holloway and M. S. Livingston, Phys. Rev. **54**, 18–38 (1938).

value of 4.86 Mev observed by Livingston and Hoffman,[26] but definitely in disagreement with the energy release of 4.56 Mev calculated from commonly accepted values for the masses involved. The value for the energy release found in the present experiments requires an energy of $(4/7)(4.97)=2.84$ Mev for the H^3 particle. According to the Cornell proton range curve (1937 revised), the corresponding mean range expected for the H^3 particles is 6.15 cm, in good agreement with the value of 6.1 cm obtained from the range-number curve of Fig. 15.

Mass difference of H^3 and He^3

The direct measurement of the energy released in the disintegration of Li^6 by neutrons is of special interest in view of its application in establishing a value for the mass difference between He^3 and H^3. A value for this mass difference previously has been obtained by eliminating the mass of deuterium between the equations

$$D^2+D^2=H^1+H^3+Q_1, \quad (1)$$

$$D^2+D^2=n+He^3+Q_2, \quad (2)$$

which gives $(He^3-H^3)+(\overset{\smile}{n}-\overset{\smile}{H^1})=Q_1-Q_2=0.69$ Mev, where Q_1 is taken as 3.98 Mev, according to Livingston and Bethe's[27] revision of data by Oliphant, Kempton, and Rutherford,[28] and Q_2 is 3.29 Mev according to Bonner.[29]

On the other hand, the masses of Li^6 and He^4 may be eliminated between the equations

$$Li^6+n=He^4+H^3+Q_3, \quad (3)$$

$$Li^6+H^1=He^4+He^3+Q_4, \quad (4)$$

giving $(He^3-H^3)+(n-H^1)=Q_3-Q_4=1.20$ Mev, if Q_3 is taken as 4.92 Mev (the average of the value determined in the present experiment and that of Livingston and Hoffman) and Q_4 is taken as 3.72 Mev.[27, 30]

Since the value of (Q_1-Q_2) fails to agree with the value of (Q_3-Q_4) by 0.5 Mev, there either must be considerable error in the determination of one or more of the four reaction energies or gamma-radiation must accompany at least one of the disintegrations. In the deuterium reactions, for example, if Q_1 were assigned too small a value or Q_2 were too large, or if gamma-radiation were emitted in reaction 1, the value for (Q_1-Q_2) given above would be too small. Likewise, the value of (Q_3-Q_4) given by the lithium reactions might be assumed to be too large. However, it appears difficult to explain an appreciable part of the 0.5-Mev discrepancy by the latter assumption. The gamma-radiation from lithium under proton bombardment has been investigated extensively in a number of laboratories but no gamma-radiation assignable to reaction 4 has been found, since the observed gamma-radiation can be produced from Li^7 but not from Li^6 targets and exhibits very strong resonance characteristics.[1, 2] A 0.5-Mev error would occur in the determination of Q_3 if the measured range of He^4 were 1.5 mm in error in the present investigation of reaction 3, while a like error in Q_4 would be introduced by an error of two mm in the range of He^3 as determined by Neuert[30] for reaction 4, assuming the Cornell range-energy relation to be correct. However, possible errors in the range-energy relation should have but small effect on the energy difference, (Q_3-Q_4), since the ranges of He^4 in the lithium-neutron reaction and He^3 in the lithium-proton reaction are very nearly equal. Furthermore, it seems unlikely that the value used for Q_3 is seriously in error in view of the manner in which it was obtained and the close agreement between the present experiment and that of Livingston and Hoffman.

Since $(n-H^1)=0.74$ Mev according to Bethe's adjusted mass values,[31] the evidence obtained from the Li^6 reactions predicts that He^3 is unstable by 0.46 Mev and may be transformed to H^3 by K-electron capture. Consequently, it seems important to look for possible gamma-radiation accompanying the (D+D) reactions and to reinvestigate the energy liberated in the disintegration of Li^6 by protons because of the relation of these reactions to the problem of proton-proton and neutron-neutron interactions.

[26] M. S. Livingston and J. G. Hoffman, Phys. Rev. **53**, 227–233 (1938).
[27] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 371 (1937).
[28] M. L. E. Oliphant, A. R. Kempton, and Lord Rutherford, Proc. Roy. Soc. **149**, 406–416 (1935).
[29] T. W. Bonner, Phys. Rev. **53**, 711–713 (1938).
[30] H. Neuert, Physik. Zeits. **36**, 629–642 (1935).
[31] H. A. Bethe, Phys. Rev. **53**, 313–314 (1938).

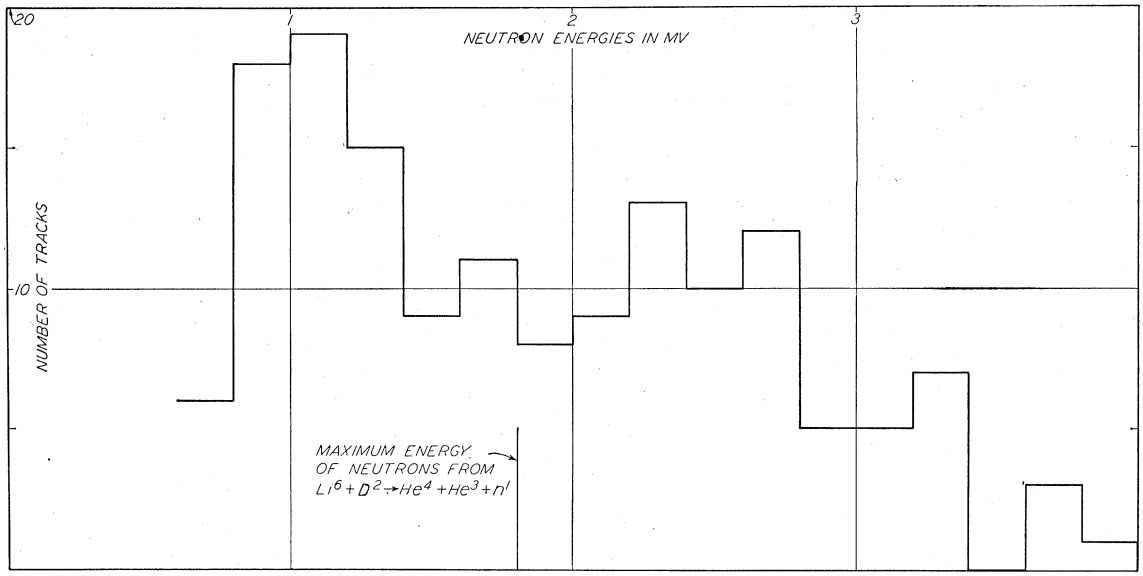

FIG. 16. Energy distribution of neutrons from Li^6+D^2.

Li^7+n

The apparatus described above also was used in a search for disintegrations produced in Li^7 by neutron bombardment. The presence of the 4600 microgram Li^7 target in front of the ion chamber under exposure to 7000 millicuries (Rn−Be) equivalent of carbon-deuteron neutrons caused no observable increase over the background counting rate at any counter bias, indicating that no appreciable number of charged heavy particles is emitted by Li^7 under these conditions. Veldkamp and Knol[32] have observed a faint beta-activity, which they attribute to Li^8, by bombarding lithium with (Rn−Be) neutrons. The observations of delayed alpha-particles from the disintegration of radio-lithium is necessary and sufficient to confirm the presence of Li^8, as discussed in Section F, but since the radioactivity observed by Veldkamp and Knol is of very low intensity, it is scarcely to be expected that such alpha-particles would be detected in the arrangement described above. Consequently, a small ionization chamber lined with ordinary lithium was placed at the center of a large block of paraffin and close to a (D−Be) neutron source equivalent to 70,000 millicuries of (Rn−Be).[24] Repeated observations failed to detect the emission of any alpha-particles after the neutron source was removed by interrupting the deuteron beam, though the ion chamber counted 4.6 Li^6 disintegrations per minute per millicurie (Rn−Be) equivalent of (D−Be) neutrons when the beam was on the target, as determined by direct count with a weak (D−Be) neutron source. With the Li^7/Li^6 ratio taken as 11.6, it was calculated that the maximum cross section for production of Li^8 from Li^7 by neutrons is less than 5×10^{-6} times the neutron disintegration cross section of Li^6.

It is difficult to compare the sensitivity of the above method with that used by Veldkamp and Knol, but it seems unlikely that their method should have been sufficiently more sensitive to compensate for the much larger number of neutrons (700 to 1) employed in the present observations. It should be pointed out that the present apparatus was sensitive only to heavy particles and therefore strictly limited to the detection of Li^8, whereas the method of Veldkamp and Knol would detect the formation of any other beta-active elements as well. Since the half-period of He^6, a beta-emitter,[33] is nearly identical with that of Li^8, it follows that they probably detected the reaction $Li^6+n\rightarrow He^6+H^1$ −2.8 Mev. The endothermic nature of this

[32] J. Veldkamp and K. S. Knol, Physica **4**, 166–170 (1936).

[33] T. Bjerge, Nature **138**, 400 (1936).

reaction would account for the low intensity of the radioactivity which they observed, since only a fraction of their total (Rn−Be) neutrons would possess sufficient energy to form He^6.

H. Neutrons from Li^6

The energy distribution of the neutrons produced by deuteron bombardment of Li^6 was investigated by measuring proton recoil tracks in a cloud chamber placed to receive neutrons emitted roughly at right angles to a deuteron beam incident on a Li^6 target deposited upon silver. A bombarding energy of 800 kev was chosen in accordance with the yield curve for this process (Section C) in order to obtain the highest possible intensity of Li^6 neutrons without introducing a serious number of neutrons from carbon. Under these conditions, the maximum kinetic energy of the Li^6 neutrons was $[(\frac{7}{8})Q+\frac{5}{8}(0.8)]$ Mev, where Q is the energy released in the reaction. Only recoil proton tracks beginning and ending in the chamber and projected within 30° of the direction of the incident neutrons were measured. About 200 such recoil tracks were obtained in a preliminary survey. The distribution of the corresponding neutron energies, plotted in Fig. 16, had a range of over three Mev. Roughly one-half of the measurable tracks were produced by neutrons of more than 2.0-Mev energy, so that the corresponding values of Q were greater than 1.7 Mev, provided these neutrons originated in Li^6 reactions. A series of tests indicated that the presence of any contamination in amounts sufficient to have produced a considerable fraction of the total neutrons could have been detected easily by the accompanying characteristic groups of charged particles. Carbon on the target doubtless contributed a small part of the low energy neutrons, but the effect of deuterium upon the observed neutron distribution at higher energies apparently was inappreciable.

Deuteron bombardment of Li^6 may produce neutrons by either of two reactions

$$Li^6+D^2\rightarrow Be^7+n+Q_1, \quad (1)$$

$$Li^6+D^2\rightarrow He^4+He^3+n+Q_2. \quad (2)$$

Strong evidence for the occurrence of the first reaction has been obtained from the energies of recoil particles observed in an ion chamber during the previous work with lithium isotopes,[1] while the second reaction is to be expected, in analogy with the reaction $Li^7+D^2\rightarrow He^4+He^4+n$, since it is exothermic.

The energy balance, Q_1, for the first case can be predicted only in so far as the mass of Be^7 can be estimated. Since the previous observations indicated that the formation of Be^7 leads to no radioactivity of short life,[1] it may be predicted that Be^7 should be roughly one Mev heavier than Li^7, either by estimating the Coulomb energy excess of Be^7 over Li^7 or by interpolating between the relative masses of neighboring pairs of nearly stable isobars. The corresponding value of Q_1 is 3.1 Mev. In the second reaction, Q_2 can be calculated from accepted values for the masses involved and is 1.56 Mev, where the chief uncertainty is introduced by a probable error of 0.2 Mev in the mass of Li^6 according to Livingston and Bethe.[34] However, the results of experiments discussed in the preceding section indicate that the accepted mass of He^3 possibly is too small by 0.5 Mev so that Q_2 might be as low as one Mev. When the uncertainty in the mass of Li^6 is taken into account it follows that an upper limit of 1.7 Mev appears reasonable for Q_2, so neutrons of energies greater than $(\frac{7}{8})(1.7)+(\frac{5}{8})(0.8)=2.0$ Mev would not be expected to originate in reaction 2. Consequently, the neutron energy distribution of Fig. 16 constitutes strong but qualitative evidence that Be^7 is formed according to reaction 1. A greater number of recoil tracks must be measured before the mass of Be^7 can be fixed accurately or any of its energy levels determined in case Be^7 is not always left in the ground state. However, the observed distribution of neutron energies is not in disagreement with the estimate of the mass of Be^7 given above.

The relative probabilities of reactions 1 and 2 can be estimated to be very roughly equal at 800 kev if all neutrons of energies greater than 2.0 Mev are assigned to reaction 1 and all others to reaction 2. However, such an estimate is subject to considerable error, chiefly because of the uncertainty in the value of Q_2 and the large statistical variations which are probable when

[34] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. 9, 373 (1937).

only 200 tracks are measured. In addition, the usual observational uncertainties for cloud chambers are present. The measurement of tracks tends to discriminate against very low energy neutrons because the recoil tracks are short, and against high energy neutrons because the longer recoil tracks are less apt to end within the cloud chamber. Likewise, there is the usual background from scattered neutrons and target contaminations. When all these factors are weighted the formation of Be^7 appears to be the more probable reaction, but the ratio 1 : 1 for the relative probabilities of the two reactions, as estimated by the more direct method, is believed to be within a factor of three of the true value.

I. Radioactivity of Be^7

Be^7 may be expected to be unstable since this isotope does not occur in beryllium minerals. Furthermore, the evidence outlined in the preceding section indicates that Be^7 is heavier than Li^7 and consequently should be transformed to Li^7 either through positron emission or absorption of an orbital electron by the nucleus. A previous investigation of isotopic targets bombarded by deuterons had shown that no short period activity existed which could be associated with Li^6 or the formation of Be^7.[1] Consequently, the possibility of a long half-life was investigated. Through the cooperation of Dr. G. L. Locher, a target of ordinary LiF which had been bombarded a month earlier with 35 microampere hours of 1000-kev deuterons was examined and found to exhibit radioactivity with a half-value period of 43 ± 6 days. Likewise, a thick target of lithium metal bombarded with 20 microampere hours of 1000-kev deuterons showed an activity corresponding to the increased proportion of lithium in the target. The surface of the latter target was scraped off and was found to carry the activity, which demonstrated that deuterons and not neutrons had produced the activation.

A chemical separation, carried out by Dr. M. H. Van Horn of George Washington University, concentrated the active element in a final precipitation of beryllium from 100 micrograms of beryllium carbonate added as a carrier. However, the chemical separation would not remove iron, which is a common impurity in metallic lithium. Consequently, an iron target was bombarded with 1000-kev deuterons and tested for radioactivity, but it was found that an iron impurity of more than 100 percent would have been required to account for an activity equivalent in intensity to that observed in the lithium target. It was apparent, therefore, that the radioactive element produced was a beryllium isotope and almost certainly Be^7, in view of the evidence from the Li^6 neutrons and the fact that the known isotopes Be^8, Be^9, and Be^{10} do not possess a 43-day half-period.

Proton bombardment of boron

A second reaction which should lead to the production of Be^7 is $B^{10}+H^1\rightarrow Be^7+He^4+Q$. A boron carbide target which had been bombarded with 11 microampere hours of 950-kev protons exhibited radioactive properties equivalent to those observed in the lithium targets. The intensity of this radioactivity was 75 percent as great as that produced in the lithium metal target, which indicates that transmutations producing Be^7 constitute an appreciable fraction of the disintegration processes for B^{10} under proton bombardment.

The range of the alpha-particle group from the reaction $B^{10}+H^1\rightarrow Be^7+He^4+Q$ could be used to determine the mass of Be^7. An attempt was made to observe this group superimposed upon the intense continuous distribution of alpha-particles from the four times more abundant isotope B^{11} but the experimental difficulties proved to be too great. If Be^7 is assumed to be one Mev heavier than Li^7, the expected range of the alpha-particle group from B^{10} is only about five mm for the negligible bombarding energies which are required in order to avoid protons scattered from the target. A reasonably intense superimposed group usually can be resolved by using counters with strong electrical differentiation, but differential counters cannot be used for very short range particles. Consequently, in the present experiments it was necessary to use a shallow ion chamber connected to an amplifier and counter without differentiation, but this arrangement did not reveal any significant departure from the expected continuous distribution of alpha-particles from B^{11} down to ranges as short as three mm, where scattered 200-kev protons from the target began to enter the ion chamber.

Nature of radiation

The 43-day half-life radioactivity observed in the lithium and boron targets was shown to be due to gamma-ray emission. When covered targets were alternated with uncovered targets within the chamber of a Lauritsen electroscope, the comparative discharge rates were those to be expected from gamma-ray ionization only. Likewise, no charged particles could be detected with Geiger-Müller counters having wall thicknesses equivalent to only a few centimeters of air, although the counting rates obtained with shielded counters indicated the emission of more than 100 gamma-ray quanta per second.

This unusual type of radioactivity might originate in the decay of Be^7 to form Li^7 by either of two reactions

$$\left.\begin{aligned} &Be^7 \rightarrow Li^7 + e^+ + v, \\ &e^+ + e^- \rightarrow 2h\nu_1, \end{aligned}\right\} \quad (1)$$

$$\left.\begin{aligned} &Be^7 + e^- \rightarrow Li^7 + v, \\ &Be^7 + e^- \rightarrow Li^{7*} + v, \\ &Li^{7*} \rightarrow Li^7 + h\nu_2, \end{aligned}\right\} \quad (2)$$

where v signifies a neutrino. In the first case, Be^7 may emit a positron with so little energy that it does not ionize appreciably but can be detected by the resulting radiation of two simultaneous 0.5-Mev annihilation quanta. In the second case, an orbital electron may be absorbed by Be^7 so that the resulting Li^7 nucleus is formed, at least part of the time, in an excited state from which it then passes to the ground level with gamma-ray emission. In the second reaction, the expected number of gamma-ray quanta per transmutation is less than unity because part of the Li^7 nuclei may be formed in the ground state and emit no gamma-radiation.

It is evident from the above discussion that several experimental tests (involving energy, simultaneity, and number of gamma-ray quanta emitted by Be^7) may be used to discriminate between transmutations of Be^7 by positron emission and by K-electron capture. Geiger-Müller counters were used in making three such tests and it was found efficacious to compare the Be^7 radiation directly with the 0.5-Mev annihilation radiation from the positron emitter N^{13}, produced in ample quantities according to the reaction $C^{12} + D^2 \rightarrow N^{13} + n$ by bombarding a graphite target with 1000-kev deuterons. N^{13} probably decays entirely by the transmutation $N^{13} \rightarrow C^{13} + e^+ + v$ so that two 0.5-Mev annihilation quanta per N^{13} atom result. In addition, comparatively faint 0.2-Mev gamma-radiation (presumably corresponding to occasional formation of C^{13} in an excited state) has been reported.[35] In the present experiments, monochromatic 0.5-Mev radiation in equilibrium with its secondaries was assured after the rays had passed through $\frac{1}{8}$ inch of lead, since the absorption curve observed with greater thicknesses of lead was a simple exponential.

Absorption coefficient

The comparative absorption coefficients for the gamma-radiation from Be^7 and for the 0.5-Mev radiation from N^{13}, measured in lead under identical conditions, were found to be 0.169 ± 0.008 and 0.130 ± 0.002 g cm^{-1}, respectively, by a least-square determination. The corresponding energy for the Be^7 gamma-radiation is 425 ± 25 kev, in reasonable agreement with the energy to be expected if a K electron were absorbed by Be^7 to form Li^7 in its known excited state at 450 kev (Section C).

Simultaneity

Another test to determine the decay process was made by placing Be^7 between two Geiger-Müller counters arranged to count double coincidence discharges. No statistically significant departure from the cosmic-ray background rate of 0.6 counts per minute was observed with the Be^7 in position. If Be^7 decayed only by positron emission, the resultant simultaneous emission of two annihilation quanta in opposite directions should have increased the counting rate by a factor of three over the background rate.

Number of quanta

A third experimental test, indicative of the process by which Be^7 decays, was made by determining approximately the average number of quanta emitted per Be^7 atom. The gamma-ray counting rates from lithium targets containing Be^7 and from graphite targets containing N^{13} were measured under identical conditions with a Geiger-Müller counter shielded with $\frac{1}{8}$ inch of lead. If μ radioactive atoms having a decay constant λ $sec.^{-1}$ and emitting n gamma-rays per disintegration are presented to a counter shielded to insure equilibrium between gamma-rays and

[35] J. R. Richardson, Phys. Rev. **53**, 610 (1938).

their secondaries, the observed counting rate, C, is given by

$$C=\lambda\mu n\epsilon \text{ sec.}^{-1},$$

where ϵ is the over-all efficiency of the detecting apparatus. In the present experiment, ϵ was considered to be the same for both Be^7 and N^{13} gamma-ray quanta since their energies are practically alike and all other conditions were held constant throughout the observations. Then, the ratio of the gamma-ray counting rates for Be^7 and N^{13} is given by

$$C_1/C_2=(\lambda_1\mu_1 n_1)/(\lambda_2\mu_2 n_2),$$

where the subscripts 1 and 2 refer to Be^7 and N^{13}, respectively. The decay constants, λ_1 and λ_2, were known with considerable accuracy and n_2 was assumed to be two annihilation quanta per N^{13} atom. The ratio μ_1/μ_2 was calculated, with appropriate corrections for radioactive decay, from available information on neutron yields for the reactions in which Be^7 and N^{13} were formed. The numbers of neutrons from deuteron bombardment of lithium and carbon targets had been measured by Amaldi, Hafstad, and Tuve,[24] the relative neutron yields from Li^6 and Li^7 were obtained from Fig. 10 of the present experiments and the known isotope ratio, and the approximate fraction of Li^6 neutrons corresponding to the formation of Be^7 was estimated within a factor of three from Fig. 16. Calculations based on the above data gave n_1 as 0.1 gamma-ray quantum per Be^7 atom within limits $0.03<n_1<0.3$, which is consistent with the hypothesis of K-electron capture because transitions to the ground state of Li^7, which produce no gamma-radiation, should be more probable than the less energetic transitions to excited levels.

Possibility of positron emission

The experiments enumerated above did not preclude the possibility of the emission of low energy positrons by a small fraction of the Be^7 atoms. Accordingly, a bare LiF target which had been bombarded some weeks previously was placed inside a cloud chamber. Repeated expansions failed to reveal any particle track of any length whatsoever which definitely originated in the target, nor was there any evidence of ionization in the immediate neighborhood of the target. However, the gamma-ray emission was sufficiently intense that occasional Compton electron tracks were observed in various parts of the chamber. The total number of Be^7 atoms decaying during the sensitive periods of the cloud chamber could be estimated, since the decay constant and the approximate number of Be^7 atoms present in the target were known, as outlined above. Thus, it was possible to estimate that, provided positron emission occurs at all, probably not more than one Be^7 atom per thousand decays with the emission of a positron of more than a few kilovolts' energy.

Discussion

There are four separate lines of experimental evidence in all, which strongly indicate that positron emission does not occur and that Be^7 decays only through the absorption of an orbital electron to form Li^7 sometimes in the ground state and sometimes in the 450-kev excited level. It follows that the upper and lower limits for the mass difference (Be^7-Li^7) may be set at 1.0 Mev and 0.45 Mev, respectively, since for greater differences positron emission would become probable and for lesser differences Li^7 could not be formed in the 450-kev state. Moreover, mass differences near the lower limit are not to be expected, since the transition probability for the 450-kev state should decrease more rapidly than Be^7-Li^7-450 kev, the energy available for this transition, in analogy with all other known radioactive processes. For example, if the relative transition probabilities for the two Li^7 states, as given by the number of gamma-rays observed per Be^7 atom, arbitrarily are taken to be proportional to the fourth power of the relative transition energies,[36] then the mass difference (Be^7-Li^7) is approximately one Mev for 0.1 gamma-ray quantum per Be^7 atom, and 0.8 Mev for 0.03 quantum per Be^7 atom.

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[36] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 197 (1936).