with mine but they did not analyze their beam magnetically, so that no accurate comparison is possible.

The absolute yield of protons (1 proton for 6×10^6 deuterons) was calculated assuming an isotropic angular distribution of the protons. This yield is smaller than that reported by Oliphant, Harteck and Rutherford who estimated the yield to be of the order of magnitude of 1 proton for 10^6 deuterons when observed at the same angle as in my experiments.

These experiments were done under the direction of Professor Rudolf Ladenburg, to whom I am greatly indebted for his supervision and help. I also wish to express my appreciation to Dr. Giarratana and Mr. Beers for their valuable assistance, to my wife for her help in taking the readings, and to the staff of the Palmer Physical Laboratory for their work in preparing apparatus. Most of the equipment was purchased from a special grant to Professor Ladenburg from the Rockefeller Foundation.

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A Wilson Cloud Chamber Investigation of the Alpha-Particles from Uranium

W. M. RAYTON* AND T. R. WILKINS
Department of Physics, University of Rochester, Rochester, New York

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The *mean ranges* of the alpha-particles emitted by uranium have been determined in terms of the *mean range* of polonium alpha-particles. The ratios obtained for the *mean ranges* are, for U I/Po, 0.6904 ± 0.0007, and for U II/Po, 0.8357 ± 0.0008. These correspond to *mean air ranges* of 2.63 and 3.18 cm, for the particles from U I and U II, respectively, in dry air at normal pressure and at 15°C, on the basis of the present polonium value of 3.80 cm. The straggling coefficient was determined for the polonium alpha-particles used as the reference range group in the experiment and gave a value considerably closer to theoretical prediction than the usual experimental results. Possible evidence for the existence of a new alpha-emitting isotope of uranium is suggested in the data. The *mean air range* for the new group is believed to be in the vicinity of 2.9 cm. An identification with the actino-uranium isotope of approximately this range suggested by Wilkins as the parent of the actinium series and with Dempster's 235 uranium isotope is proposed.

PRECISE data on the characteristics of the alpha-particles from uranium have been sought for many years. It was of particular importance to the development of the general theory of the disintegration sequence in the uranium family when the alpha-emission from uranium was first definitely resolved by Laurence's[1] data into two range groups. The presence of two isotopes was thus indicated, but a still closer scrutiny of this subject seems worthwhile at the present time.

In the first place it seems advisable to establish the ranges of the alpha-particles from the known isotopes U I and U II with considerably higher precision than has so far been realized. This precision is deemed especially desirable because of the recent advances toward more precise theoretical formulation of the mechanics of alpha emission. Furthermore, this investigation was prompted by the fact that from several independent lines of reasoning the conclusion has been reached that uranium may be more complex than range data have so far indicated and that U I and U II may well have associated with them very small amounts of isotopes whose alpha-rays remain as yet undemonstrated in any direct fashion. These suspected isotopes are believed to be the missing long-lived parent, or parents, of the actinium series of radioelements; and their detection would consequently be a matter of some significance.

The presentation of the arguments which proceed to the conclusion that the parent of the actinium family will be found as an alpha-emitting isotope of uranium, present on earth today only in a small amount, would be too

* Present address, Massachusetts Institute of Technology, Cambridge, Mass.

[1] G. C. Laurence, Phil. Mag. **27**, 690 (1914).

lengthy for this paper, but a brief bibliography of the most important discussions of the topic is appended to this article.

All investigation of uranium alpha-ray ranges is seriously complicated by the extremely slow rate of decay of uranium. This has, in the past, necessitated the use of thick layers of source material to provide a suitable number of emitted particles. The unfortunate consequence was a broadening of the normal straggled distribution of path-lengths to a degree which nearly masked the fact that the radiation from uranium was complex. All work prior to 1910 gave uranium a single range group with a value between 2.9 and 3.4 cm, and subsequent work has realized resolving powers just sufficient to distinguish the two main range groups. The results of the most important range determinations are given in Table I.

Experimental

In the present investigation improved results were hoped for as a consequence of the use of a large area of an extremely thin film of uranium which thereby minimized the disturbing effects of absorption in the source layer itself. Secondly, to accentuate the range differences, this source was used in a relatively large Wilson chamber filled with a low stopping power gas mixture.

The Wilson chamber used was similar in general design to that described by Dahl, Hafstad and Tuve,[2] being of the sylphon bellows type. The chamber proper consisted of a Pyrex annulus some 16 cm in diameter and 5 cm deep, to which was clamped a suitable disk of plate glass which formed the top and viewing window for the chamber. The present outfit was arranged for automatic operation and utilized a motor driven shaft and suitable cams. A useful flexibility in the cycle was obtained through the use of a time-delay relay in the motor circuit. This could be adjusted so as to give a proper rest interval just prior to the expansion. For the track photography, light was supplied by a high intensity arc lamp, located sufficiently distant to prevent serious thermal disturbance at the chamber. The beam was collimated to a rectangular cross section, adapted to the chamber dimensions, and was controlled by a shutter mechanism geared to the main drive shaft of the chamber.

The source was prepared by evaporation, in high vacuum, of metallic uranium[3] ribbon. The metallic vapor was condensed on a thin flat plate of rolled nickel, and produced a thin uniform deposit over the entire source area of approximately 30 square centimeters. The deposit weighed 0.09 mg per square centimeter, which is approximately one one-hundredth of the value used by Laurence, and one-twentieth of Kurie's.

In the chamber, a mixture of helium and air was adjusted to give a conveniently low stopping power, with resultant tracks which were approximately twice as long as their normal length in air under standard conditions. The use of helium was dictated by the ease of its retainment, compared with hydrogen, and by the fact that its stopping power relative to air is very close to a constant at all points along the range of a particle.

For several reasons it seemed advisable to refer the present range measurements to some standard range group. The use of polonium suggested itself, inasmuch as all investigations of its alpha-emission have consistently demonstrated its homogeneity. Consequently, a polonium source electrochemically prepared on the tip of a fine nickel wire was attached to one end of the uranium source support and provided one or more suitable reference tracks in every photograph.

Stereoscopic photographs of the tracks were obtained by the use of two Sept cameras with obliquely mounted objectives.[4] The cameras were fastened to a base plate of cast aluminum forming thereby a rigid unit which could be swung into a position such that the cameras looked downward through the top of the chamber. For reprojection,

Table I. *Summary of most important range determinations.*

Method	Range in Air @ 15°C 760 mm		Investigator	Date
	U I	U II		
Ionization	2.53 cm	2.91 cm	Geiger-Nuttall[1]	1912
Pleochroic Haloes	2.82	2.91	Gudden[2]	1924
Wilson Chamber	2.73	3.28	Laurence[3]	1927
Scintillations	—	3.23	Rutherford[4]	1927
Wilson Chamber	2.72	3.28	Kurie[5]	1932

[1] H. Geiger, J. Nuttall, Phil. Mag. **23**, 439 (1912).
[2] Gudden, Zeits. f. Physik **26**, 110 (1924).
[3] G. C. Laurence, Phil. Mag. **27**, 690 (1914).
[4] E. Rutherford, Phil. Mag. **4**, 580 (1927).
[5] F. Kurie, Phys. Rev. **41**, 701 (1932).

[2] Dahl, Hafstad, Tuve, Rev. Sci. Inst. **4**, 373 (1933).

[3] Purchased from Mackay Chemical Co., New York City.
[4] Glazebrook, *Dictionary of Applied Physics* No. 4, p. 400.

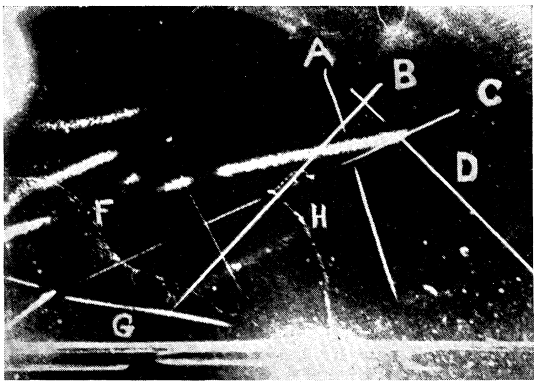

FIG. 1. Reproduction of typical chamber photograph, showing tracks of various types and ages.

the developed negatives were reintroduced into their respective cameras which were equipped with demountable projection illuminators, and the images were projected on a translucent viewing screen, arranged to be conveniently adjustable to any desired plane. Orienting the screen in such a manner that the images of a given track, as projected from the two units, were observed to coincide throughout, reestablished the original position of a track relative to the camera assembly, and a measurement of the true length could be made from the life-sized image so produced. Measurements were made directly by means of millimeter ruled coordinate paper which was waxed to a glass disk, forming thereby a translucent viewing screen which permitted setting and measurement from a position back of the screen. Two foot operated switches in the projection lamp circuits made it possible to flick rapidly from one projector to the other and facilitated accurate settings of the viewing plane, since even slightly incorrect positions were then made manifest by a pronounced flicker or image shifting as one switched rapidly back and forth between projectors. Refractive error, introduced by photography through the chamber top, was balanced out in the customary manner by the insertion of a compensatory glass plate in the projection beam.

With the apparatus as described about 3000 stereophotographs were obtained in the course of a few days, during which time conditions were maintained as constant as possible.

The large area of the source plate made impracticable the use of any shutter arrangements which sharply limited and defined the time during which alpha-particles were traversing the chamber. However, the tracks produced by particles which have passed a trifle prior to the optimum sensitive instant were found to be quite broad and diffuse, while those which came late in the period were characterized by incomplete condensation and a thin, stringy appearance. These characteristics have been shown, especially by the work of Kurie, to be sufficiently pronounced to serve as adequate criteria for the selection of tracks formed during the proper interval. Furthermore, in the present experiment, the duration of the illuminating flash was sufficient to reveal any appreciable falling motion and this provided another check on tracks of doubtful age, since in a track which had just been formed the droplets would be practically motionless. Tracks that showed motion were therefore rejected, as were also any which had passed through, or near, any regions which had been affected by condensations on older tracks in the vicinity. Such regions of prior condensations are locally heated and depleted in condensable vapor to the point that subsequent tracks fail to excite condensation. Moreover, for tracks traversing such regions there is a high probability that the alpha-particle will lose a great deal of energy in a chance journey through a single droplet and its apparent range would be completely altered.

Fig. 1 is a reproduction of a typical photograph and exhibits some of the effects described. The edge of uranium source appears as a white line across the bottom of the photograph, and the wire carrying the polonium is seen at the lower left corner. The broad, fuzzy segments are remnants of tracks of particles which passed so much prior to the sensitive interval that the ions had migrated under the influence of the electric field which was maintained in the chamber almost to the completion of the expansion stroke. Later tracks are seen to be quite sharp and well defined, except in the vicinity of previous condensations as seen in the tracks labeled *A*, *C*, and *D*. Track *C*, from the polonium, occurred quite late in the sensitive period and shows as a thin, threadlike structure. Track *G*, from polonium also, is a trifle old and furthermore ran in so close to the uranium source support that it entered the region of noncondensation which sheathes that object owing

to its high heat capacity. Track B is acceptable for measurement, since it had an uneventful and normal career, to all appearance. F and H are beta-trails which came presumably from the intermediate UX products present on the uranium source.

These standards for track selection reduced the number of usable uranium tracks to about 900. The length distribution obtained is presented in Fig. 2 where the experimental points are shown as small circles; the smooth full-line curves representing Gauss exponential functions which have been fitted to these points.[5] On the theory of probability, it is evident that some alpha-particles will lose more energy than others in traversing a given distance. Consequently there will be differences in the observed ranges of the particles, a phenomenon referred to as "straggling," even though the particles are all ejected with identical speeds for a given radioactive substance. These remarks serve to explain why the observed ranges are to be found grouped around some mean value according to a Gaussian error function, and thus the problem of determining the mean value for a range group involves fitting a curve of this type to the data. This exponential is of the form

$$y=\frac{1}{\alpha_1\sqrt{\pi}}e^{-(x-l)^2/\alpha_1^2}\cdot dx,$$

where y is that fraction of the total number of tracks whose length lies between x and $x+dx$, α_1 is the shape parameter of the distribution, and l the mean path length, or mean range of the group.

To find l and α_1 the method here followed was to determine first an error function which, to the eye, appeared to provide the best fit to the most significant experimental points in the distribution. The fitting of this exponential was carried out by projecting a plot of the experimental points on a screen, and then superposing on this the silhouette of a cardboard error function pattern, projected by means of a point light source. The dimensions of this shadow were thus conveniently adjustable through changes in projection distance, and by rotation of the pattern about its axis of symmetry. By this means it was possible to estimate the approximate shape and extent of a distribution group, and the weighed mean of that distribution gave l, to a zero-order

[5] A. King and W. Rayton, Phys. Rev. **51**, 826 (1937).

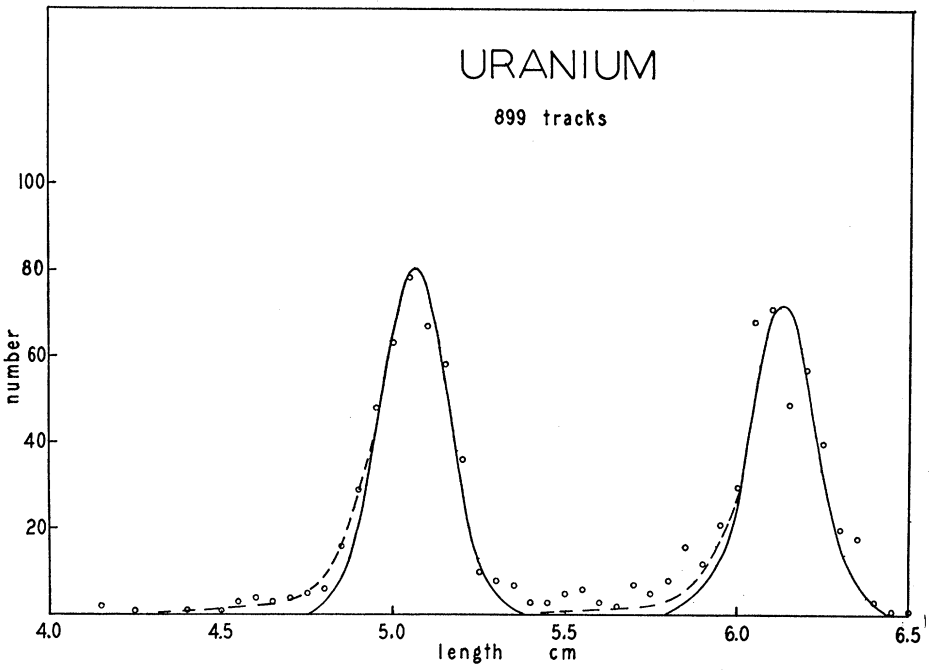

FIG. 2. Track length distribution for uranium alpha-particles.

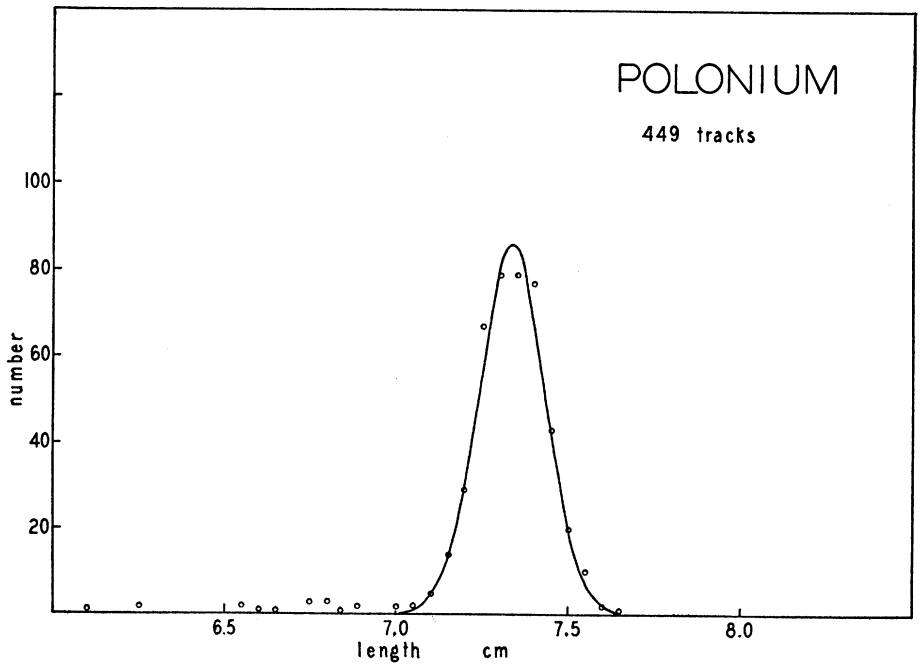

FIG. 3. Track length distribution for polonium alpha-particles.

approximation. The shape parameter α_1 was determined, likewise approximately, from the analytical relation that its square is equal to twice the mean square deviation of the distribution. Having thus found approximate values for the parameters, we proceeded by the method described in the accompanying related paper[5] to the calculation of successively closer approximations as determined by the condition of a least square ordinate deviation between experimental points and the smooth curve.

In the case of the polonium data, the fit of the resulting exponential is quite good even at considerable distances from the mean, as seen in Fig. 3. With the uranium data, however, the situation is complicated by the presence of an appreciable number of tracks whose lengths are such as to place them in the interval between the two main distribution groups. The precision of the results from these data is therefore somewhat inferior to that attained in the polonium case because of the greater uncertainty in determining the proper approximate forms for the component distribution groups. This uncertainty was not thought to be of serious magnitude however, since the long-ranged side of the uranium II group, and the short-ranged side of the uranium I group, provided us with templates which could be applied to the complicated portions of the curves.

The figures thus obtained for the mean values of the distributions came out as in Table II.

The above-mentioned figures cannot be directly compared to establish the ranges of particles from uranium I and uranium II relative to the polonium reference, since the stopping power of the chamber mixture is not quite constant for particles of different energies. However, a comparison of the mean polonium range (7.336 cm) observed in the chamber, with the present accepted value of 3.805 cm[6] for the mean range in air at 15°C and 760 mm pressure sufficed to determine the composition of the chamber mixture,

TABLE II. *Main values of range from distribution curves.*

	Mean values, cm
Polonium	7.336±0.003
Uranium I	5.061±0.003
Uranium II	6.123±0.003

[6] Lewis, Wynn-Williams, Proc. Roy. Soc. **A136**, 349 (1932).

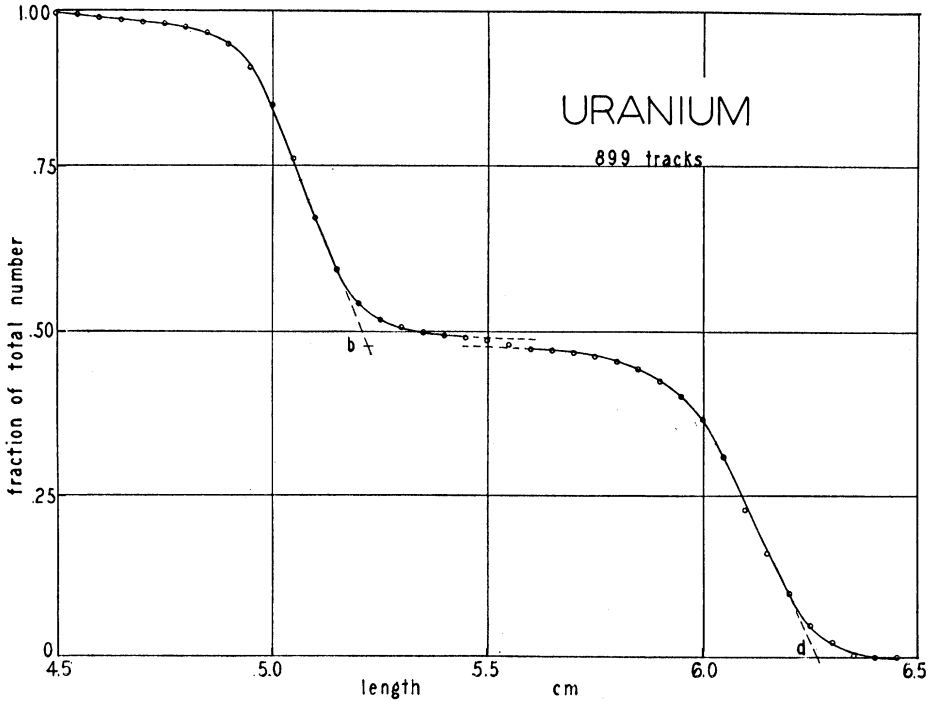

FIG. 4. Integrated number-distance distribution of uranium data.

since Gurney[7] has determined the stopping power of pure helium for polonium alpha-particles. Knowing the chamber mixture, its stopping powers for particles of air range 2.6, 3.2, and 3.8 cm were calculated on the basis of Gurney's data. These stopping powers turned out to be 0.5191, 0.5189, and 0.5187, respectively, and when applied to the chamber data, the mean ranges of uranium I and uranium II, referred to polonium, reduce to the following values:

Uranium I/Polonium $0.6904 \pm 0.0007(5)$
Uranium II/Polonium $0.8357 \pm 0.0008(2)$.

Taking the mean air range for polonium as 3.805 cm leads to mean air ranges for uraniums I and II of 2.63 and 3.18 cm, respectively. The estimated uncertainty in the absolute magnitude of the figure for polonium is stated as being in the vicinity of 0.01, consequently there is small point to quoting further than to the second decimal place in the uranium values here given.

Comparison of the results of this and previous experiments can be made only on a basis of the so-called *extrapolated number-distance*[5] range values. Graphically, these are obtained from a plot in which the number of tracks whose lengths exceed a given value is graphed against length values. An extrapolation of the descending linear portion is commonly used to specify this type of range, by the point of intersection of the line of extrapolation and the horizontal axis. This method of range specification is especially ill-suited to nonhomogeneous cases, as in Fig. 4, owing to uncertainty as to the location of the plateaus which thus renders uncertain the extrapolated values for all except the longest range group.

For purposes of comparison with the earlier results, one may easily compute these extrapolated values, once the mean ranges are known, since the former exceed the latter by the amount $\alpha\sqrt{\pi}/2$, as may be easily shown analytically to be the case for Gaussian functions. In the present instance, the computation yields these values:

Uranium I 2.71 cm @15°C and normal pressure.
Uranium II 3.27 cm

Gurney, Proc. Roy. Soc. **A107**, 340 (1925).

Straggling of Alpha-Particle Ranges

It is clear that the shape of a track length distribution curve will depend upon the precision of the individual measurements of length and thus the constant α_1, previously mentioned as the shape parameter of the distribution, will depend not only on the actual straggling but upon the distribution of errors inherent in the technique of measurement. This dependency can be shown[5] to be of the form,

$$\alpha_1^2=\alpha_0^2+\alpha^2,$$

where α_0 is the modulus of precision related to the measurement process itself, and α is the parameter of the true straggling distribution. In the present case an effort has been made to evaluate α_0 separately, by means of a set of measurements taken on a test object, and thus from the observed α_1, it was possible to determine the α representative of the actual straggling. In the present instance, α_1, was observed as 0.141 cm in the case of the polonium distribution, and α_0 was determined as 0.101 cm, leading to a corrected value of 0.098 cm for α.

Dividing α by l, the mean value of the length distribution, defines a constant known as the *straggling coefficient* and denoted by the letter ρ. As incidental to the present experiment ρ has been determined from the polonium data to have the value 1.34×10^{-2}; a value that is in considerably closer agreement with the results of the Bohr theory[8] of alpha-particle straggling than has usually been observed. The present result is 1.21 times the Bohr value, as contrasted with the more usual discrepancies amounting to factors between 1.4 and 1.5.[9–12]

The inconsistencies between theory and experiment have been large and uniformly in the same direction, but it seems to us probable that they are in large measure to be attributed to the difference between α_1 and α, a distinction which appears to have been overlooked at least in previous Wilson chamber work. To a smaller extent the difficulty is presumably inherent in the limitations of the simple Bohr theory with its simplifying assumptions. The Bethe-Williams[13, 14] wave mechanical treatment is at present the most rigorous discussion of this subject, and the results of this theory are in general about 10 to 15 percent larger than the corresponding Bohr figures. For the present experiment a calculation of the Bethe-Williams straggling coefficient was made, on a basis of mean excitation potential data used by Mano[15] in similar calculations. The result obtained ($\rho=1.19\times10^{-2}$) is exceeded by our experimental value only to the extent of 13 percent. A similar new and close approach to theoretical values has been recently reported by Bennett[16] who used an electrical method to investigate straggling in micas. The results range between 1.1 and 1.2 times the Bohr figure, and lend further strength to the belief that previous determinations have been in error.[17]

Isotopic Composition of Uranium

A comparison of the relative areas included beneath the smooth distributions of the uranium I and uranium II data shown in Fig. 1 is readily made in terms of their definite integrals. The result shows that there are 90.5 percent as many tracks from uranium II as from uranium I. On the basis of the radioactive equilibrium one would expect equal numbers in the two cases, but a definitely smaller angular aperture was available to the longer tracks in the space limitations of the Wilson chamber.

Of particular interest is the appearance of an unexpectedly large number of tracks in the region between the two main groups. (Fig. 2). The dashed portion shown appended to the left side of the uranium II distribution is a transposition, proportionately reduced, of the left side of the uranium I group (observed points—dashed line). This makes possible a rough estimate of the number of "extra" tracks in the vicinity. Plotting this excess leads to the suspicion that there is a

[8] Bohr, Phil. Mag. **25**, 10 (1913); **30**, 581 (1915).
[9] I. Curie, Ann. d. Physik **3**, 320 (1925).
[10] Meitner-Freitag, Zeits. f. Physik **37**, 481 (1926).
[11] Curie, Mercier, J. de phys. **7**, 289 (1926).
[12] Briggs, Proc. Roy. Soc. **A114**, 313 (1927).
[13] H. Bethe, Ann. d. Physik **5**, 325 (1930); Zeits. f. Physik **76**, 293 (1932).
[14] E. J. Williams, Proc. Roy. Soc. **A135**, 108 (1932).
[15] G. Mano, Ann. d. Physik **1**, 407 (1934).
[16] Bennett, Proc. Roy. Soc. **A155**, 419 (1936).
[17] The calculated values of our straggling coefficients are not corrected for the effects due to variation of charge on the alpha-particle during its flight, nor for the influence of the alcohol vapor present in the gas mixture. These two effects are of opposite natures regarding their influence on ρ, and would be of very small magnitude.

small distribution group in the vicinity of 5.55 cm. The same effect appears to be reflected in the integral plot of Fig. 4 as a small but perceptible break in the middle section of the curve. This distribution, if real, corresponds to an alpha-ray group of about 2.9 cm mean air range at 15°C, or an extrapolated range close to 3.0 cm. It may be significant that this value for a range group is in fair agreement with that required by Wilkins[18] in explanation of the observed time shift in the inner rings of the pleochroic haloes in old micas. The magnitude of the observed hump, about 3 percent of the uranium I group, is in accord with most current hypotheses regarding an actinouranium isotope, and it seems especially significant in view of Dempster's[19] recent detection of a uranium isotope of atomic mass 235. It is tempting to identify the source of the alpha-particles here observed with the 235 isotope, which is in all probability the radioactive parent of the actinium family.

Regardless of the existence or nonexistence of a range group at precisely the point suggested, there is definitely an abnormally large number of tracks whose lengths exceed 5.4 cm in the chamber medium, which figure very probably defines the upper limit of the region which can receive contributions from the spread of the uranium I group. Of the total number of measured tracks, 49.7 percent are longer than this in spite of the definite discrimination imposed by the available space in the chamber against the longer tracks. Such an observed result thus seems indicative of the presence of another long range group other than uranium II. Further, since no tracks were observed to be of lengths so great as definitely to place them beyond the normal straggled distribution of uranium II, it can be assumed with some certainty that the mean range of the new group is less than the mean value of that distribution. Further speculation on the point is probably pointless until more data have been secured.

[18] Wilkins, Phys. Rev. **29**, 352 (1927).
[19] Dempster, Proc. Am. Phil. Soc. **75**, 755 (1935).

Conclusion

The mean ranges of the alpha-particles emitted by uranium have been determined in terms of the mean range of polonium alpha-particles. The ratios of these mean ranges are, for uranium I/polonium, 0.6904 ± 0.0007, and for uranium II/polonium, 0.8357 ± 0.0008. These correspond to mean ranges of 2.63 and 3.18 cm in dry air at normal pressure at 15°C for uraniums I and II, respectively. These figures are based on the present polonium value of 3.805 cm, which is believed to be correct in absolute magnitude to within less than 0.01 cm.

The straggling coefficient has been determined for the polonium alpha-particles used as the reference range group in the experiment. The result is in markedly better agreement with theoretical prediction than has been customarily observed.

The distribution of track lengths observed among the alpha-particles emitted from an extremely thin layer of uranium suggests the possibility of a new alpha-emitting isotope of uranium, whose mean range in air has been tentatively placed in the vicinity of 2.9 cm. It seems possible that this may be alpha-particles from Dempster's uranium isotope of mass 235.

Supplementary bibliography on the problem of the radioactive parentage of the actinium series

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Johnstone-Boltwood, Phil. Mag. **40**, 50 (1920).
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A. v. Grosse, Phys. Rev. **42**, 565 (1932); Proc. Roy. Soc. **A150**, 363 (1935).

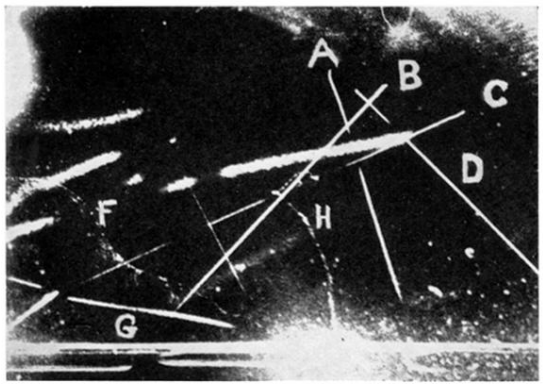

FIG. 1. Reproduction of typical chamber photograph, showing tracks of various types and ages.