The Beta-Rays of Mesothorium 1 and Radium D

D. D. LEE AND W. F. LIBBY
Department of Chemistry, University of California, Berkeley, California

(Received September 6, 1938)

Mesothorium 1 has been prepared from an old sample of thorium sulfate whose purity was demonstrated by measuring the rate of decay of thorium B prepared therefrom. The mesothorium 1 was shown to be free from contaminations of other thorium decay products and isotopic radioactive elements by measuring the rate of growth of mesothorium 2, and proving the absence of alpha-emitters. Magnetic deflection and aluminum absorption experiments have shown that the particles emitted are electrons whose maximum energy is 53,000±4000 electron volts. Samples of RaD and RaE, pure to within 0.3 percent, have been prepared. The momentum of the most energetic conversion beta-ray of RaD has been determined as 765±32 gauss cm, and the upper limit of the RaE continuous spectrum was found to be 5150±250 gauss cm. The upper limit apparently to be assigned to the primary beta-ray spectrum of RaD was determined as 546±10 gauss cm, or 25,500±1000 ev, in agreement with the determination of Richardson and Leigh-Smith. An estimate of the observed number of secondary beta-rays arising from the conversion of the 47,2-kev gamma-rays of RaD indicates the emission of five to ten such electrons per 100 disintegrations, in rough agreement with the results of Kikuchi and the predictions of Fisk for the *L* conversion of dipole radiation, but in complete disagreement with the conclusions of Stahel and von Droste. Reasons for this disagreement are given.

AMONG the naturally occurring radioactive bodies about which there is very little, or only controversial, information available at present are mesothorium 1 and radium D.

The emission of ionizing radiation from the first of these has never been definitely established, although it was discovered in 1907, and it was for a long time considered to be "rayless." However, from the chemical properties of mesothorium 1[1] and its product, mesothorium 2 (which have been found to be isotopes of radium, and actinium, respectively) and the displacement law, it is assumed that a weak beta-particle is emitted in the decay of mesothorium 1.

The decay of radium D to form radium E is known to occur by the emission of primary beta-rays, gamma-rays, secondary beta-rays from the internal conversion of gamma-rays, x-rays, and perhaps tertiary, or Auger, electrons from the internal conversion of the x-rays.

The first definite information about the primary beta-particles of radium D was furnished by a very recent cloud-chamber investigation by Richardson and Leigh-Smith,[2] who found a large number of electrons having energies below four kv, and estimated the upper limit of the spectrum, by comparison with the distributions predicted in the beta-decay theories of Fermi, and of Konopinski and Uhlenbeck, as either 16, or 24, kv.

The energy of the gamma-radiation emitted has been determined by absorption measurements, and by analysis of the beta-ray line spectrum, the latter leading to the most accurate value, 47,200 ev.[3] The more recent of such determinations[4] have been done in such a manner as to determine not only the energy of this gamma-ray, but the energy of the accompanying x-radiation and to estimate the quantities of each emitted. The x-rays have been found to have energies corresponding rather closely to the *L* x-rays of bismuth, with which they have been identified, although determination of their ener-

[1] O. Hahn, Ber. **40**, 1462 (1907), Physik. Zeits. **8**, 277 (1907); B. Boltwood, Physik. Zeits. **8**, 556 (1907); H. N. McCoy and W. Ross, J. Am. Chem. Soc. **29**, 1709 (1907); L. Haitinger, K. Peters and St. Meyer, Vienna, Akad. Sitz.-Ber. **120** [*2a*], 1199 (1911); L. Meitner, Physik. Zeits. **19**, 257 (1918).

[2] H. O. W. Richardson and A. Leigh-Smith, Proc. Roy. Soc. **A160**, 454 (1937).

[3] J. Danysz, Le Rad. **2**, 1, 6 (1912); E. Rutherford and H. Richardson, Phil. Mag. **26**, 324, 937 (1913); C. D. Ellis, Proc. Camb. Phil. Soc. **21**, 121 (1921–23); L. Meitner, Zeits. f. Physik **9**, 131, 145 (1922); **11**, 35 (1922); I. Curie and G. Fournier, Comptes rendus **176**, 1301 (1923); D. H. Black, Proc. Roy. Soc. **A109**, 166 (1925); L. F. Curtiss, Phys. Rev. [*2*]**27**, 672 (1926).

[4] S. Kikuchi, Jap. J. of Phys. **4**, 143 (1925–27); L. H. Gray and O'Leary, Nature **123**, 568 (1929); L. H. Gray, Nature **130**, 738 (1932); J. Petrova, Zeits. f. Physik **55**, 628 (1929); S. Bramson, Zeits. f. Physik **66**, 721 (1930); E. Stahel and G. J. Sizoo, Zeits. f. Physik **66**, 741 (1930); H. O. W. Richardson, Proc. Roy. Soc. **A133**, 367 (1931); G. von Droste zu Vischering, Zeits. f. Physik **84**, 17 (1933); E. Stahel, Helv. Phys. Acta **8**, 651 (1935).

gies have given only average values. The number of such photons has been found to be 22 to 30 per 100 disintegrations, and the number of 47,000-ev gamma-rays, about 3.5 per 100.[5] We shall assume that the x-rays are excited by the conversion of 47,000-ev gamma-rays in L shells, (the K shell could not be excited because the energy of the gamma-ray is too low). The number observed, considered with the fluorescence yield of bismuth, has led to the conclusion that each primary beta emitted leaves the resulting RaE nucleus in an excited state 47 kv above the ground state of RaE, and that all such nuclei emit gamma-rays of this energy. Of these, 96 or 97 percent are internally converted in L, M and N shells of extranuclear electrons, the remaining three or four percent are emitted without conversion.

In a series of experiments, wherein were used a point counter covered with a collodion foil and a source of RaD, RaE and Po placed in an evacuated chamber, Stahel[6] measured the number of electrons from RaD. He distinguished these from the beta-rays of RaE, the alphas of Po, and the general background radiation by means of a magnetic field and various aluminum foils. The absorption of the RaD betas in one, two and three of his collodion foils led Stahel to the conclusion that he was observing, through the first permanent foil, only the secondary electrons due to the conversion of the 47,200-ev gamma-ray; but, as it was pointed out by Feather and Richardson,[7] the absorption which Stahel observed did not agree with the absorption which would be calculated from the known energies and intensities of these electrons, and the range-energy relations of Williams.[8] In fact, the observed absorption could be accounted for only by the assumption that Stahel was getting a relatively large number of low energy (4 to 20 kv) electrons through the first foil. From the work of Richardson and Leigh-Smith,[2] and the fact that Stahel's first foil could completely stop only electrons of less than 4500 ev, it is clear that more than half of these might have been primary beta-rays. From this fact alone and without estimating the number of observed electrons which might have been caused by Auger effect of the x-radiation, one is led to conclude that there are not 83 or more gamma-ray conversion electrons per 100 disintegrations as Stahel believed, but that this number must be considerably smaller, and that the number of 47,200-ev gamma-rays (emitted plus converted) is less than one per disintegration. It follows from this that since no beta-rays of RaD having energies corresponding to a transition from ground state to ground state have been observed, there may occur the emission of two or more gamma-rays of less than 47,200 ev energy (nuclear x-rays) in a "cascade" process involving more than the one known excited level of the RaE nucleus. It is not at all improbable, that such nuclear x-rays may have been observed by Kikuchi, Gray and O'Leary, von Droste and Stahel, and their corresponding conversion electrons may have been observed by Kikuchi, Petrova, Richardson, and Richardson and Leigh-Smith, only to be confused with the L rays and Auger electrons arising therefrom.

From the work of Kikuchi one finds only about twenty electrons attributable to the conversion of 47,200-ev gamma-rays per 100 disintegrations. Since about thirteen of these are from the L orbits, the L conversion coefficient calculated from this value is 13/3.5, or about 3.7. A theoretical calculation by Fisk[9] gives the values 1.8 and 2.9 for the L conversion coefficients of dipole and quadrupole radiation, respectively. Only the assumption of multipole, or magnetic multipole radiation could have accounted for the coefficient 17.1 calculated by Stahel.

The subsequently described experiments with RaD were intended to furnish information regarding the upper limit of the primary beta-rays, and to permit an estimate of the number of secondary electrons produced by the conversion of 47,200-ev gamma-rays.

Preparation of Samples

All mesothorium preparations were made from a thirteen-year-old sample of Kahlbaum thorium sulfate, the purity of which was determined by

[5] H. Lay, Zeits. f. Physik **91**, 533 (1934).
[6] E. Stahel, Zeits. f. Physik **68**, 1 (1931).
[7] N. Feather and H. O. W. Richardson, Nature **129**, 314 (1932).
[8] E. J. Williams, Proc. Roy. Soc. **A130**, 310 (1931).
[9] J. B. Fisk, Proc. Roy. Soc. **143**, 674 (1934).

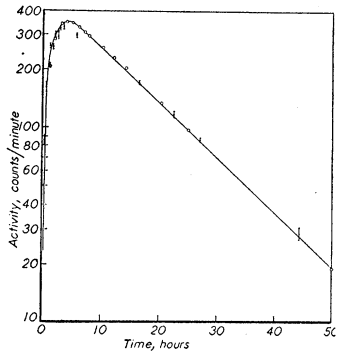

FIG. 1. Growth and decay curve for ThB precipitate to test purity of thorium sample.

following the growth and subsequent decay of gamma- and strong beta-ray activity in a sample of thorium B prepared from about one-half gram of thorium sulfate. The activity was measured with a solid-wall Geiger-Müller counter and increased in the first four hours after at exactly the rate to be expected for the growth of ThC from ThB preparation. After six hours, the activity decayed with the half-life of thorium B. This indicates the absence of more than three to five percent of any radium B, actinium B, or radium D contamination (see Fig. 1).

Mesothorium 1 and 2 were removed from a solution of thorium sulfate by precipitating barium sulfate. To remove from this precipitate any traces of occluded thorium or radiothorium, the $BaSO_4$ was converted to $BaCl_2$ by heating with carbon, digestion of the residue in concentrated sodium carbonate solution, and leaching of the resulting solid with dilute hydrochloric acid. To the resulting solution a trace of ferric chloride was added, and the ferric ion removed as ferric hydroxide by the addition of a slight excess of sodium hydroxide solution. This procedure removed thorium, radiothorium, mesothorium 2, and the elements of the thorium active deposit. As thorium X is isotopic with mesothorium 1, it cannot be removed by chemical separations but must be allowed to decay. About one month is necessary for the removal of 99.6 percent of ThX and its decay products.

From such a month-old sample of mesothorium 1 and 2, either of these two elements could be removed by simple chemical precipitations. The addition of ferric ion as the chloride and its removal as ferric hydroxide leaves a solution containing only about ten percent of its original content of mesothorium 2. Two further treatments of this kind suffice to remove all noticeable amounts of mesothorium 2 and the precipitation of the barium ion as barium carbonate removes mesothorium 1 for experimentation. The growth of activity of such a sample, as observed with a screen wall counter, is shown in Fig. 2. Comparison of the measured activities with the calculated solid curve shows the sample to have been pure to within at least two percent. The initial activity is not proved to be caused by MsTh 1 beta-rays since the same curve would have been obtained with a MsTh 1 sample containing a trace of MsTh 2. Counting of alphas possibly emitted by an equivalent sample of MsTh 1 and 2 showed certainly less than two percent contamination by radium, thorium X, or actinium X.

The ferric hydroxide precipitates containing mesothorium 2 contain also appreciable quantities of mesothorium 1. About eight to ten percent has been found in all such precipitates examined in this work by following the decay of activity with an electroscope. However, if such a sample be dissolved in dilute hydrochloric acid, the hydrogen ion concentration adjusted to about 10^{-4} M by the addition of sodium acetate, and a large excess of sodium dichromate with a trace of barium chloride added, the resulting precipitate removes enough mesothorium 1 so that the ferric hydroxide precipitate which can be prepared from the filtrate will contain mesothorium 2 pure to within 98 percent (see Fig. 3).

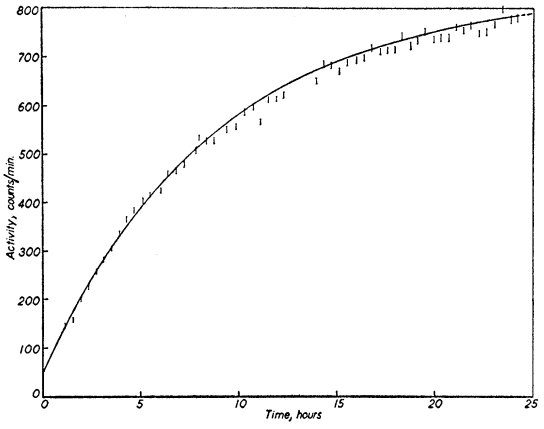

FIG. 2. Growth of mesothorium 1 precipitate.

Samples of radium D and radium E were prepared from the contents of an old radon tube, which was washed in concentrated nitric acid and water, and crushed under about five cc of concentrated HNO_3. After heating and allowing the glass particles to settle, the solution was decanted and diluted by the addition of about three cc of distilled water. Individual preparations were made from one-cc portions of this final solution by the usual methods, i.e., the solution was evaporated twice with excess concentrated HCl, the residue dissolved in about five cc of 0.1*N* HCl, and heated to 90° or 100°C. Then 99 percent of the polonium content was removed by rotating in the solution at about 200 r.p.m. a silver foil (of approximately one cm^2 area) for one hour, and the remaining polonium removed by the same treatment with another silver foil. Radium E was removed similarly on a nickel foil. Such a sample of RaE can be shown by following the decay of activity for 35 days to have been originally free from Po to at least 99.9 percent and to have contained no more than 0.25 percent of RaD. This was done by using the screen wall electroscope to be described later and various aluminum foils in order to distinguish between the beta-rays of RaE and the alphas of Po.

After the above-mentioned removal of RaE, 0.1 cc of a solution containing 10^{-4} gram/cc each of Pb and Bi, as chlorides, were added to prevent the loss of RaE or RaD by occlusion on dust particles, etc. the solution was allowed to stand over night. Then the RaE left by the previous treatment and that produced subsequently by the decay of RaD was removed by rotating in the hot solution a nickel foil of the same size as that used in the previous removal of RaE. The resulting residue was evaporated once again with excess HNO_3, then twice with distilled water, and the final residue digested with five cc of a two percent solution of NH_4NO_3 to insure the solution of the lead content. The $BiONO_3$ residue was filtered off to remove the last traces of RaE. Radium D was removed from the filtrate by precipitating the lead as PbS and found to be radioactively pure to at least 99.8 percent.

Apparatus and Experimental Technique

The counter tubes, voltage supply, amplifier, recorder and magnet used in these magnetic deflection experiments were the same as those described in the preceding article.

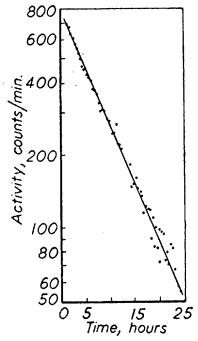

FIG. 3. Decay of mesothorium 2 precipitate.

In determining the absorption of the mesothorium 1 and 2 beta-rays by aluminum, measurements were made with a Lauritsen electroscope having a portion of its cylindrical case cut out and replaced by a piece of copper gauze such as is used for the screen of a screen-wall counter. In order to determine how far an ionizing particle must travel into the space enclosed by this modified case to be detected, measurements were made of the rate of discharge of the electroscope due to the alpha-particles from a Po source placed at various distances from the screened portion. The results showed that any alpha-particle, and presumably any other ionizing radiation which passes into the screen-enclosed space produces ions leading to the discharge of the electroscope.

Corrections for the growth of mesothorium 2 in samples of mesothorium 1 which were used to determine the aluminum absorption and the upper energy limit of the beta-ray spectrum were made in the following manner. The activity observed on the electroscope for the uncovered sample in a standard position was measured first. Then absorbers were added in increasing thicknesses, as measurements of the penetration were made until the absorber thickness reached 7 or 12 mg/cm^2. Following this procedure the absorber thickness was reduced by the same increments until the sample was uncovered. The average of the two readings taken at each absorber thickness automatically gave a value of the activity corrected for growth of mesothorium. This average is nearly the activity which would have been obtained had it been possible to make all measurements simultaneously at the time of the

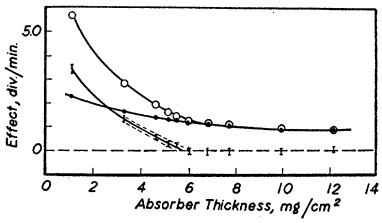

FIG. 4. Aluminum absorption curve for mesothorium 1 and mesothorium 2.

maximum thickness measurement, since the growth of mesothorium 2 is a nearly linear function of time within any period of two hours or less, and the two readings at each thickness are time-symmetrical with respect to the maximum thickness reading. To find the activities caused by mesothorium 1 alone, this curve was compared with the absorption curve for pure mesothorium 2 and the portion of the former curve which was parallel to the latter was extrapolated back. Subtracting the activities on this extrapolated curve from those on the measured curve gives a third curve (see Fig. 4) which is the absorption of the beta-rays of mesothorium 1 in aluminum.

The same technique was used in magnetic deflection experiments on mesothorium. The activities from a pure mesothorium 2 sample were measured at various field strengths and compared with the curve obtained when a sample originally containing only mesothorium 1 was measured at various fields. The field was increased to some maximum strength by predetermined increments, then decreased by the same steps. The difference between the measured curve and the extrapolated mesothorium 2 curve shows the effect of field on the beta-rays of mesothorium 1 alone (see Fig. 5).

In the case of the magnetic deflection experiments on radium D no such corrections were necessary provided the measurements were made within a few hours after preparation of the samples because of the slower growth of radium E.

EXPERIMENTAL RESULTS

The average of two determinations of the mesothorium 1 upper limit by absorption (see Fig. 5) is 57,000±5000 ev, while five magnetic deflection experiments gave an average of 771 ±45 $H\rho$, or 51,000±4700 ev (see Fig. 6). Because of the greater accuracy inherent in the second type of measurement, these values are taken as indicating that the upper limit of the mesothorium 1 spectrum is 53,000±4000 ev. That this is the upper limit of the primary beta-ray spectrum does not necessarily follow, for although no gamma-rays have been observed in the disintegration of mesothorium 1, the present

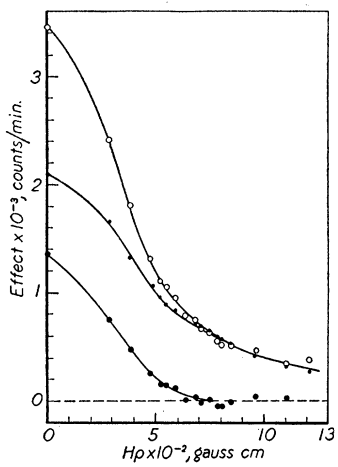

FIG. 5. Magnetic determination of mesothorium 1. Upper limit.

work does not exclude the possibility that soft gamma-rays might exist and their conversion give rise to secondary electrons having energies up to the observed maximum. This might make the observed upper limit somewhat higher than the true value. However, since the total conversion of soft gammas is relatively improbable because it must occur in the L shell in this case, it seems that most of the electrons are of nuclear origin. This point is illustrated in the work on RaD.

Figure 7 shows the activities observed at various magnetic field strengths with a sample of RaD. Curve (I) gives activities due to about one-half of the sample measured at various fields up to 1500 gauss; curve (II) gives the corresponding values for a smaller portion of the same sample. (Electroscope measurements made 19 days later to determine the relative activities of these portions of the sample due to accumulated RaE in each indicated a ratio of 21 to 1.

As the activity of the small portion at zero field is observed to be 1200 counts per minute, that of the large portion must have been over 25,000, a value too large to be recorded by the equipment used.)

Examination of curve (I) reveals an activity which falls off very rapidly with increasing field strength up to an $H\rho$ value of 570 gauss cm, a more gradual slope from that value to 770 gauss cm, and a very gradual slope from 770 to 1500 gauss cm. Curve (II), on the other hand, shows only the steep portion ending at 530±20 gauss cm, which is apparently the upper limit of the primary beta-ray spectrum of RaD, since all the betas from the conversion of the 47,200-ev gamma-rays have larger values of $H\rho$. The gradual slope in curve (I) is, therefore, presumably due to conversion betas, and gives an upper limit of 770±40 gauss cm which is not far from the accepted value,[10] 738 gauss cm for the highest conversion line. The remaining activity in the large sample is undoubtedly due to RaE betas, and perhaps Po alphas, as well as RaD gammas. Similar curves obtained with

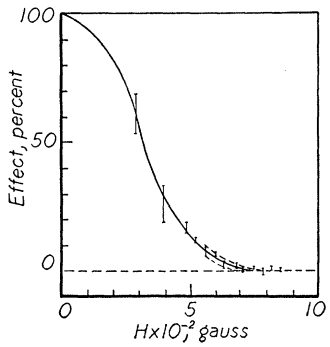

FIG. 6. Average curve for magnetic determination of mesothorium 1. Upper limit.

RaD samples prepared in the same or a slightly different manner, gave the values collected in Table I.

To obtain an independent check on any possible systematic errors in the magnetic depth determinations of such upper limits, measurements were made with a sample of mesothorium 2 and with one of RaE. The observed upper limit for mesothorium 2 was 6880±250 $H\rho$, in excellent agreement with the magnetic spectrographic determination of Yovanovitch and d'Espine.[11] For RaE, the upper limit was 5150±250 $H\rho$, a value entirely concordant with the latest determinations.

[10] L. Meitner, reference 3.

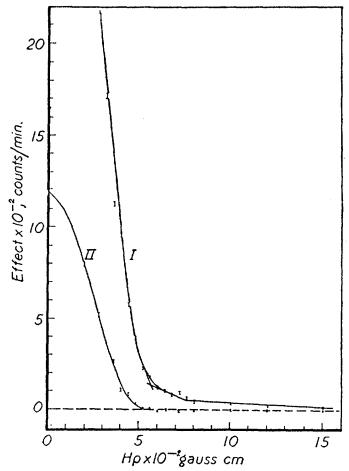

FIG. 7. Magnetic analysis of RaD.

DISCUSSION OF RESULTS

From the data of Table I one is led to conclude that the end point observed with RaD at 546±10 gauss cm, or 25,500±1000 ev, is that of the continuous primary beta-ray spectrum, although it is possible that the conversion of nuclear x-rays, if they exist in this disintegration, might give rise to electrons of energies up to this limit.

The electrons found at $H\rho$ values of 530 to

TABLE I. *Data on* RaD.

TOTAL INTENSITY OF SAMPLE (COUNTS/MIN)	UPPER LIMIT (STEEP PORTION OF CURVE) (GAUSS CM)	HIGHEST CONVERSION LINE (GAUSS CM)
1200	530	Not observed
1500	540	Not observed
1600	540	Not observed
Av. 1433	Av. 537	
15,000	540	Not determined
20,000	570	Not determined
25,000	570	770
35,000	530	760
Av. 23,750	Av. 553	Av. 765

[11] D. K. Yovanovitch and J. d'Espine, Comptes rendus **179**, 1162 (1924); J. phys. radium, [6]**8**, 276 (1927).

770 gauss cm are then to be considered the only observed beta-particles from the conversion of the 47,200 ev gamma-ray; and by extrapolating this portion of the curves to zero magnetic field (under the assumption of the theoretical appearance of the curve which would be expected with this apparatus for a source emitting only the gamma-ray conversion-betas of RaD) one finds about five such conversion electrons per 100 disintegrations. Since this extrapolation is somewhat difficult, it seems entirely possible that there might be as many as ten such conversion betas per 100 disintegrations in which case the calculated L conversion coefficient would be 2.1, very nearly the value calculated by Fisk for the case of dipole radiation.

FEBRUARY 1, 1939 PHYSICAL REVIEW VOLUME 55

The Absorption Coefficient of 5.8-Mev Gamma-Radiation in Aluminum

J. HALPERN AND H. R. CRANE
University of Michigan, Ann Arbor, Michigan
(Received December 7, 1938)

The absorption coefficient of monochromatic 5.8-Mev radiation in aluminum is found to be 0.075 ± 0.009 cm^{-1}. By deducting the theoretical value of the pair absorption coefficient, the Klein-Nishina absorption coefficient is found, which is 0.062 ± 0.009 cm^{-1}. This value is in good agreement with the theoretical value. The experimental method used is such that all quanta which are scattered more than 10 degrees in the aluminum are excluded completely from the data.

SEVERAL measurements[1] of the absorption coefficient of the F+H[1] gamma-radiation have been made by means of the ionization chamber. The experiment to be described gives a result by a different method, which does not involve the assumption that the radiation consists of a single line and which reduces the effect of scattering and geometrical errors to a very small amount. The method was first used by Delsasso, Fowler and Lauritsen[2] for the measurement of the absorption coefficient of the Li+H′ and the F+H[1] radiation in lead.

Our experimental arrangement is shown in Fig. 1. On alternate expansions of the cloud chamber a block of aluminum 10.2 cm thick was placed in the path of the gamma-rays, between the target and the lead collimator. A plot of the number *versus* energy of the recoil electrons ejected from the carbon sheet was made for each of the two cases: with and without the aluminum block in the gamma-ray path. These are shown in Fig. 2. If, in both curves, we consider only tracks in the shaded area (above 4.6 Mev) we can be sure that the gamma-ray quanta responsible for these have not been scattered more than 10 degrees before reaching the carbon sheet. It is calculated from the angle-energy relations in the Compton effect that if a quantum of initial energy 5.8 Mev were scattered more than that amount in the aluminum, it could not produce a recoil electron in the carbon sheet with an energy greater than 4.6 Mev, and hence could not fall within the shaded area of the curve. Similarly, gamma-ray quanta of initial energy less than 4.85 Mev are automatically excluded, because they cannot give rise to recoil electrons of more than 4.6 Mev.

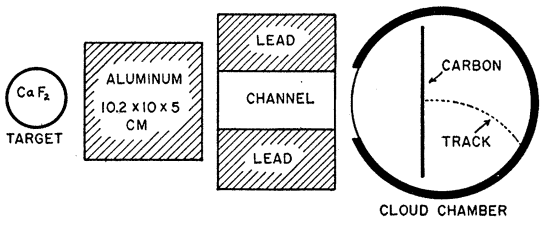

FIG. 1. Experimental arrangement.

[1] E. McMillan, Phys. Rev. **46**, 325 (1934); H. R. Crane, L. A. Delsasso, W. A. Fowler and C. C. Lauritsen, Phys. Rev. **46**, 1109 (1934).
[2] L. A. Delsasso, W. A. Fowler and C. C. Lauritsen, Phys. Rev. **51**, 391 (1937); Phys. Rev. **51**, 527 (1937).