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Conversion Electrons of Radium D

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The conversion electrons of radium D have been studied with thin sources on thin backings in a beta-ray spectrograph using calibrated photographic emulsions. The number of conversion electrons due to the 47-kev gamma-ray has been measured to be 74±5 per hundred disintegrations. The $L:M:N$ ratio is 1:0.26:0.077. This implies a complex decay scheme for radium D, since earlier results give 3.5 unconverted 47-kev gamma-rays per hundred disintegrations.

INTRODUCTION

DESPITE numerous studies of the radiations of radium D, doubt remains regarding some of the main features of the decay because of the softness of these radiations.

Measurements on the primary beta-spectrum[1] indicates an endpoint less than 30 kev, but with large uncertainty. Most definite are the conversion lines associated with a 47-kev gamma-ray, for which the previous results are summarized in Table I. The gamma-ray is insufficiently energetic to convert in the K shell. Momentum is in gauss-cm. Intensities given in Table I are estimated from a photographic record, without correction for energy dependence of the emulsion sensitivity.

To determine the fraction of the decays associated with the 47-kev transition, several measurements have been made of the fraction of the decays associated with the unconverted gamma-ray, and the fraction associated with excited atomic levels of the radium E (bismuth^{210}) atom due to internal conversion of the gamma-ray. In addition to clarifying the decay scheme, these measurements give the conversion coefficient of the gamma-ray.

The results of various workers on the number of unconverted gamma-rays per hundred disintegrations are in good agreement. These have been given as follows:[3] Bramson, 3.1; Gray, 4.0; Von Droste, 3.5; Stahel, 3.8.

The results on the number of excited atomic levels caused by conversion of the 47-kev gamma-ray vary widely. Those obtained from measurements on the conversion electrons directly have given low results, of the order of 10 per hundred disintegrations,[4] implying that the 47-kev excited state is a relatively minor feature of the decay. Several measurements by an indirect method have given much higher results. This method involves determination of (a) the number of L x-rays produced due to internal conversion in the L shell, (b) the fraction of excited L levels which yields x-rays (the remainder decaying by Auger effect), and (c) the contribution of the L excited states to the total number of excited levels due to the 47-kev gamma-ray.

According to Stahel[3] (a) is 25.1 per hundred disintegrations, (b) is 42 percent, using a result of Lay,[5] and (c) is 60 percent from the results of Curtiss in Table I. This implies conversion occurs in about 99 percent of the disintegrations. Very recently Kinsey[6] obtained for (b) 47.5 percent, and estimated for (c) 75 percent from the known results for the 40-kev gamma-ray of thorium C. He notes Curtiss' figure must be too small because of the correction necessary for the dependence of photographic darkening on energy.

* This work was carried out in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

[1] H. O. W. Richardson and A. Leigh-Smith, Proc. Roy. Soc. **A160**, 454 (1937); W. F. Libby and D. D. Lee, Phys. Rev. **55**, 252 (1939); A. Saha, Proc. Nat. Acad. Sci., India **12**, 159 (1946).

[2] According to private communication from Dr. M. E. Rose theoretical calculations of the conversion coefficients for the L shells for different multipoles are in progress.

[3] S. Bramson, Zeits. f. Physik **66**, 721 (1930); J. Gray, Nature **130**, 738 (1932); G. Von Droste, Zeits. f. Physik **34**, 17 (1933); E. Stahel, Helv. Phys. Acta **8**, 651 (1935).

[4] T. San-Tsiang, Phys. Rev. **69**, 38 (1946).

[5] H. Lay, Zeits. f. Physik **90**, 533 (1934).

[6] B. Kinsey, Can. J. Research **26**, 421 (1948).

TABLE I. Previous results for the momenta of the β-ray conversion lines associated with the 47-kev gamma-ray from radium D.*

Conversion level	Danysz ρ	Ellis ρ	Ellis i	Meitner ρ	Black ρ	Curtiss ρ	Curtiss i
$L\,I$	602	600	1.00	602	600	594.3	1.00
$L\,II$	607	605	0.04	...	606	600.3	0.06
$L\,III$	...	628	0.01	...	...	...	
M	718	717	0.40	718	714	709.1	0.50
N	743	742	0.20	741	738	735.2	0.20
O	...	...		...	...	742.5	0.01

* J. Danysz, Le. Rad. **2**, 16 (1912); C. Ellis, Proc. Camb. Phil. Soc. **21**, 121 (1921–23); L. Meitner, Zeits. f. Physik **9**, 131 (1922); D. Black, Proc. Roy. Soc. **A109**, 166 (1925); L. Curtiss, Phys. Rev. **27**, 672 (1926).

These results imply 71 percent of the decays give conversion electrons.

These conflicting results indicate the desirability of a direct measurement of the number of conversion electrons produced in the radium D disintegration, with special attention to minimizing self-absorption effects in the source, as these may have caused serious errors in the earlier work.

Still to be fitted into the decay scheme are the data on several weak, low energy gamma-rays recently reported recorded with a Cauchois spectrograph.[4]

EXPERIMENTAL PROCEDURE

The spectrum of radium D has been examined with thin uniform sources on thin backings by exposure in a conventional 180 degree beta-ray spectrograph using photographic detection. The dependence of photographic darkening on energy and exposure has been determined previously[7] for the emulsions used in this work.

To calculate the number of conversion electrons per disintegration from the spectrum, the geometry of the spectrograph and the disintegration rate of the source are measured. These data, together with the sensitivity

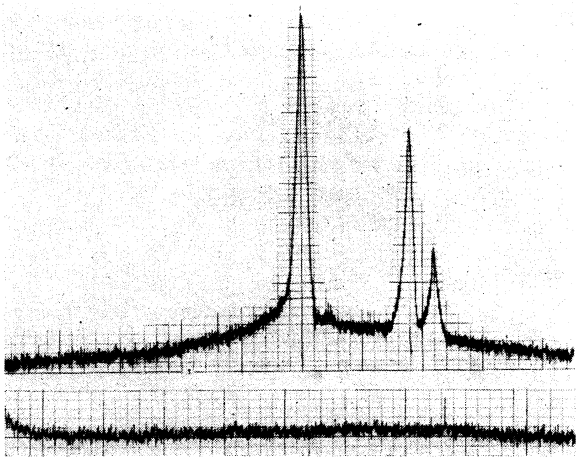

FIG. 1. Low energy spectrum of electrons from radium D and radium E in equilibrium.

[7] L. Cranberg and J. Halpern, Rev. Sci. Inst. **20**, 641 (1949).

TABLE II. Comparison of primary and secondary standards with progressively greater thickness of absorber next to the counter window.

Primary std.	Secondary std.	Ratio
6.78	252	37.0
1.47	59.5	40.5
1.10	46.0	41.8
0.85	34.2	41.6
0.49	20.5	41.9

of the emulsion used give C, the percentage of the disintegrations accompanied by the conversion electrons in a given line, according to the following equation:

$$C=100\cdot\frac{2\pi^2\rho}{\theta}\cdot S\int_{l_1}^{l_2} Ddl\cdot\frac{1}{\phi t}, \qquad (1)$$

where ρ is the radius of curvature to the center of the line in cm, θ is the semi-angle subtended by the width of the spectrograph slit at the source, S is the sensitivity of the emulsion in electrons/cm^2 per unit density for the electron energy involved, l_1, l_2 are the lower and upper endpoints of the line, D is the density above background at any point in the line, l is distance measured along the film in cm, ϕ is the disintegration rate of the source per second, t is the exposure time in seconds.

This calculation assumes (1) the lines are sharp, (2) the emission from the source is isotropic, (3) there is no loss of electrons due to self-absorption, (4) the photographic reciprocity law is obeyed, (5) the density-exposure relation is linear, and (6) fading is negligible. To satisfy the first three assumptions sources as thin and uniform as possible were prepared. Previous work[8] supports assumption (4). Most of the exposures were run so that densities were in the linear density-exposure range. For a few exposures the darkening due to the $L\,I$ line extended into the non-linear region. Appropriate corrections were then made based on density-exposure data taken with the electron gun at the energy of the $L\,I$ line. Fading was checked for nine-day exposure, which was the longest exposure time used. This was done by exposing a calibrating strip in the electron gun at the beginning and end of a nine-day period, during all of which time the strip was in vacuum, and comparing densities of the two sets of exposures. Fading was negligible.

The sensitivity S was determined each time a spectrograph exposure was processed by processing simultaneously, in the same container, a calibrating strip cut from the same sheet of film as the spectrograph strip. The calibrating strip was exposed to a known number of electrons in an electron gun, at the energy of the $L\,I$ line, with the apparatus and procedures described elsewhere.[7]

The spectrograph film and calibrating strips were run

[8] O. Klemperer, *Einfürung in der Elektronik* (M. S. Rosenberg, New York, 1944).

through a Leeds and Northrup recording microphotometer and the integral of Eq. (1) was evaluated graphically from the recording.

The spectrograph and magnet used have been previously described.[9] To minimize scattering all exposed surfaces of the spectrograph camera have been lined with polyethylene, and a shutter was installed to prevent exposure during the interval in which the camera is being evacuated.

DETERMINATION OF SOURCE ACTIVITY

To determine the disintegration rate of a source, the following procedure was adopted. Aliquots of radium D salt solution were delivered at the same time to the spectrograph source support and to a silver disk furnished by the Bureau of Standards. After equilibrium was reached between the radium D and radium E (f an equilibrium mixture was not used to begin with) the activity on the disk was determined by comparison with a radium D–radium E equilibrium standard whose disintegration rate is furnished by the Bureau of Standards, with a 3 percent uncertainty.

The aliquot delivered to the disk was evaporated, giving a small ring-shaped deposit whose average thickness, estimated from a weighed aliquot, was always within a factor of 5 of the average thickness, 1 mg/cm^2, of the primary standard provided by the Bureau. To prevent loss of activity the secondary standards were covered with 5 μg/cm^2 zapon films.

To minimize the effect of differences in area of primary and secondary standards when making activity comparisons, the source-to-counter distance was made sufficiently large, 2.5 inches, so that counting rate was insensitive to source area. This was checked by varying the position of a small source. To minimize differences between the primary and secondary standards in regard to self-scattering and self-absorption, comparisons were made with progressively greater thickness of absorber placed next to the counter window. Table II shows the results of such a series of comparisons, the statistical uncertainty in the counting rates being 2 percent.

It is to be expected that the absorption curves should run parallel with greater absorber thickness, since then only the most energetic electrons are counted and these are the least affected by self-scattering and self-absorption. Hence the value taken[10] for the correct ratio from the data of Table II is 42.0±1.0.

SOURCE PREPARATION

The radium D used in these measurements was furnished in the form of lead nitrate by the Eldorado Mining and Refining, Ltd., of Canada. Evaporation of the material with excess hydrochloric acid converted this to the chloride. The radioactive purity of the material was checked by carrying out a separation of the radium E by the Erbacher[11] method. The activity of a separated sample was followed for forty days, the ratio of the final to initial activities being 20 to 1, the rate of growth corresponding to the half-life of radium E. These measurements were made with about 10 mg/cm^2 of absorber to eliminate the radium D radiations. Samples separated by a new method[12] gave a ratio of 70 to 1 on one test, implying a maximum contamination of 1.4 percent.

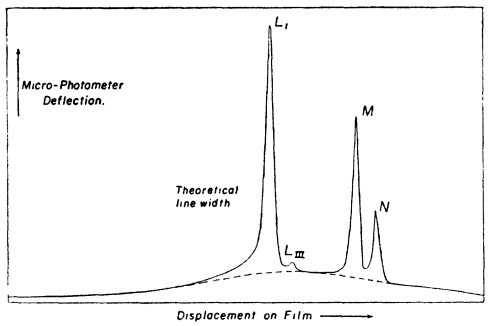

FIG. 2. Tracing of radium D–radium E low energy electron spectrum, showing estimated continuum.

The sources were prepared on zapon films of 5 μg/cm^2 by delivery of 10 to 15 microliters of radioactive solution, exposing to an atmosphere of ammonia for about 2 minutes to precipitate the lead as hydroxide, followed by evaporation in vacuum. The most uniform sources were obtained when the radioactive solution did not solidify in the vacuum, but evaporated from the liquid state. With the best technique, microscopic examination of a source after evaporation showed at least 90 percent of the nominal area of the source was actually covered with material in a uniform manner.

RESULTS AND CONCLUSIONS

Figure 1 is a reproduction of a photometric record of the low energy electron spectrum of radium D and radium E in equilibrium, taken with a source of average thickness of 43 μg/cm^2. Accompanying it is the record of the background density taken in the unexposed portion of the film. Figure 2 is a tracing obtained from this record by averaging the fluctuations due to granularity, above a base-line corresponding to the average background. Figure 3 is a replot of the region about the *L I* line in which account is taken of the density-exposure relation to show the true

[9] Plesset, Harnwell, and Seidl, Rev. Sci. Inst. **13**, 351 (1942).

[10] This procedure has been corroborated by another method by Dr. B. Burtt of the Brookhaven National Laboratories.

[11] O. Erbacher, Zeits. f. physik. Chemie **A156**, 142 (1931).

[12] Such efficient separation of radium D from radium E was obtained under the following circumstances. Radium D in the form of $PbCl_2$ solution in doubly distilled water was left undisturbed for about 24 hours. Samples of 10 to 20 microliters pipetted from the top of the solution were found on half a dozen tests to have less than 5 percent of the equilibrium amount of radium E, as determined from the growth of RaE activity, whereas the radioactive material was old enough for secular equilibrium to have been established in the mother solution. A one-cc glass beaker was used, half-filled with solution having a concentration of about 1 mc/cc.

relative intensity relationship between the line and the continuum on which it appears superimposed.

In all cases the lines observed are superimposed on a continuum, the height of which, relative to the peak intensities of the lines, differs greatly for thick and thin sources. For three thin sources, however, the results are very similar to those shown in Fig. 1. There is a rise in density approaching the *L I* line, an approximately constant level between the *L* and *M* lines, then a slow decline to the end of the film, which corresponds to a momentum of 850 gauss cm, or an energy of 60 kev.

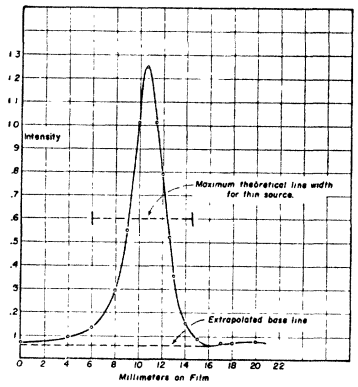

FIG. 3. True relative intensities of line *L I* and continuum.

Reference is made below to possible interpretation of this continuum. Its outline is indicated by the dashed lines of Figs. 2 and 3. Also shown on these figures are the maximum line widths calculated from the geometries of the source and spectrograph. Low energy tails are evident due, undoubtedly, to source-thickness effects. The continuum is distinguishable from these tails on the assumption that the former may be estimated by extrapolation from the high energy sides of the lines as shown in the figures.

A. Relative Line Intensities

The weak lines *L II* and *O* are not resolved in Figs. 1 and 2 because of the unfavorable combination of wide source and fast scanning of the record. To obtain these lines special runs were taken with a narrow source, and much slower speeds of scanning with the microphotometer. Line intensities were taken relative to the *L I* line, using the three thinnest and most uniform sources.

TABLE III. Intensities of the conversion lines relative to the intensity of *L I*.

Line	Density ratio, weighted average	Resolution correction	Film correction	Relative intensity
L I	1.0	1.0	1.0	1.0
L II	0.09 ±0.015	1.01	0.98	0.090±0.015
L III	0.019±0.004	1.05	0.93	0.019±0.004
M	0.38 ±0.02	1.19	0.65	0.29 ±0.02
N	0.115±0.015	1.24	0.60	0.085±0.01
O	0.029±0.01	1.25	0.58	0.021±0.007

The results are summarized in Table III. The "film correction" factor, which takes account of the energy dependence of the film sensitivity, was obtained with the procedure and apparatus described before.[7] With some sources more than one exposure was obtained. The weighted averages represent the results of three or more exposures for the strong lines. The uncertainties are due almost entirely to uncertainty in estimating the position of the continuum.

From Table III the number of conversions in the *L* shells is 73±2 percent of the total, in good agreement with Kinsey's estimate based on the thorium *C* 40-kev gamma-ray.

B. Absolute Intensities of Lines *L I* Plus *L II*

Because of the difficulty of resolving lines *L I* and *L II* with the dimensions of source it was desired to use for this phase of the work, measurements were made from records on which the lines were not resolved. From the preceding results the separate intensities may be calculated.

In Table IV are given the values of the measured quantities from which the quantity *C* is computed for lines *L I* plus *L II* according to Eq. (1). The results are given for four sources exposed a total of 6 times. All runs were taken with No-screen x-ray film, except that marked with an asterisk, for which Ilford B-2 plates of 50 micron emulsion thickness were used. The transmission of the spectrograph was the same in all cases, amounting to 0.0715 percent for the *L I* line. This is the reciprocal of the geometric factor in Eq. (1). The activity of source 12 is given as an upper limit because contamination of the apparatus was evident after the first run, and the source had visibly deteriorated; the calculated value of *C* is consequently a lower limit. The estimated uncertainties for the individual measurements are 5 percent in *S*, 4 percent in ϕ, and 5 to 10 percent in the integral, being greatest for the smallest values of the integral. The consistency of the results for a range of source thicknesses is to be particularly noted.

Taking account of the *L III* line, 54 percent of the disintegrations are accompanied by conversion in the *L* shell. Taking a value of 3.5 percent for the unconverted 47-kev gamma-ray, the ratio of the number of

TABLE IV. Values of the measured quantities from which the quantity *C* is computed for lines *L I* plus *L II*.

Source	t sec. 10^4	S electrons / den. cm 10^7	ϕ sec.$^{-1}$ 10^5	$\int Ddl$ den. cm	*C*%	Average source thickness μg/cm²
11	9.40	2.15	1.84	0.31	54	101
12	8.65	2.45	<1.73	0.16	>37	207
	14.7	2.16	<1.73	0.30	>36	
17	*17.9	11.6	1.26	0.068	49	68
	7.39	2.20	1.26	0.16	53	
18	78.5	2.50	0.37	0.44	53	43
			Weighted average		53±3	

internal conversion electrons from the L shell to the number of unconverted gamma-rays is 15.5.

The total number of conversion electrons is 74 ± 5. Adding to this the number of unconverted gammas gives 77.5 ± 5 for the percentage of the decays associated with the 47-kev excited state of radium E. Thus there must be an alternative decay scheme accounting for approximately 20 percent of the decays. These results are in good agreement with the results of Kinsey based on the indirect method, involving measurement of the L x-rays. The results of Kinsey and Stahel taken together with those reported here comprise a set of mutually consistent data which may be interpreted as lending support to whichever of the following three quantities is least certain: the number of L conversion electrons, the number of L x-rays, or the yield of L x-rays caused by internal conversion (fluorescence yield).

C. Possible Additional Conversion Line

Figure 4 shows the record obtained with a source on which only one exposure was run, after which the zapon film deteriorated seriously. There is indication here of a weak line at 470 ± 10 gauss cm, corresponding to an energy of 19 ± 1 kev. The same indication was found on three scans over different portions of the exposure. An upper limit to the intensity of this line relative to the $L\,I$ line is about 2.5 percent. The uncertainty in the intensity is of the same order, however. A similar upper limit may be placed on the strengths of any other lines, down to an energy of about 15 kev. Below this energy the sensitivity of the emulsions used was very low. Assuming a line at 19 kev is due to conversion in the $L\,I$ shell, the energy of the gamma-ray is 35.4 ± 1 kev, in agreement with one of the new gamma-rays reported by San-Tsiang.[4]

D. The Continuum

Comparison was made of the continua obtained with a source in which radium D and radium E were in equilibrium, and a source in which the radium E had on the average only 20 percent of its equilibrium value during the exposure. In both cases the continua bore approximately the same intensity relation to the lines,

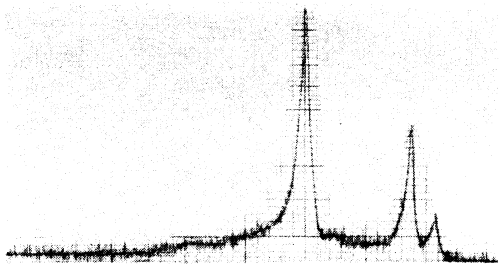

FIG. 4. Radium D electron spectrum with indication of additional line.

the electrons recorded between the centers of the $L\,I$ and M lines accounting for about 25 percent of the disintegrations. This eliminates the possibility that the radium E contamination is responsible for any appreciable portion of the continuum. The continuum can be interpreted as evidence of a more energetic primary beta-spectrum than has been hitherto been reported but also it may be due to scattering of conversion electrons, probably from the side-walls of the spectrograph.

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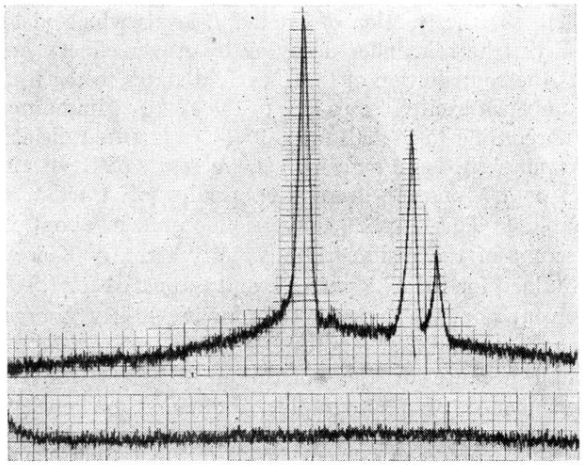

FIG. 1. Low energy spectrum of electrons from radium D and radium E in equilibrium.

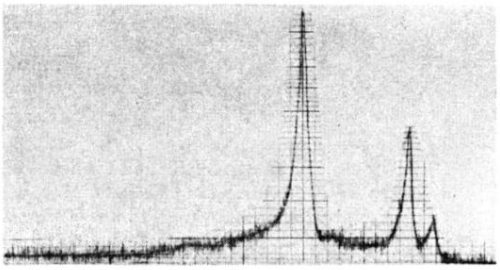

FIG. 4. Radium D electron spectrum with indication of additional line.