Radiations from RaD and RaE†

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The electron and gamma-spectra from RaD and RaE have been thoroughly investigated. The gamma-radiation from RaD was studied with both krypton and argon proportional counters with brass and aluminum cathodes. The L_α, L_β, and L_γ radiations of Bi were observed and identified with a critical absorber. The intensity ratios of $L_\alpha:L_\beta:L_\gamma$ are 1:1:0.2. The previously reported 7.8-kev (10 percent) line was not found but could be strongly excited by copper backing. The weak 23-kev line (10^{-3} per disintegration in our measurements) could be contributed from the piling effect of the detecting system.

The conversion electrons of RaD were investigated in a solenoid magnetic spectrometer to obtain the L and $(M+N)$ conversion coefficients. The results are:

$$N e_L/N_\beta = 64 \text{ percent},\quad N e_{M+N}/N_\beta = 21 \text{ percent},\quad N e_{L+M+N}/N_\beta = (85\pm5) \text{ percent}.$$

The conversion electrons of RaD were again investigated with a 180° beta-spectrometer with a resolution of 0.8 percent and a counter window of $\sim 6\ \mu\text{g/cm}^2$. The L_{I}, L_{II}, and L_{III} conversion lines of the 46.5-kev gamma-ray were resolved. The ratio of $L_{\text{I}}:L_{\text{II}}:L_{\text{III}}:M_{\text{I-III}}:M_{\text{IV-V}}:N_{\text{I-V}}:N_{\text{VI-VIII}}+0$ are 1:0.075:0.007: 0.25:0.006:0.07:0.007. From the ratios of the L-subshell conversion electrons, the 46.5-kev transition is interpreted as an $M1$ type.

The upper limit of the intensity of the reported lines at 42 kev, at 37 kev, and at 31 kev must be less than 0.5 percent per disintegration if the same conversion coefficient is assumed.

The unconverted 46.5-kev gamma-radiation is 0.07 ± 0.02 per disintegration. Thus the excited state of 46.5 kev in RaE can account for (92 ± 5) percent of the disintegrations.

Neither internal conversion electrons nor nuclear gamma-radiations are found in RaE. A faint x-ray ($\sim$80 kev) of the order of 10^{-4} per disintegration due to the ionization effect was observed in RaE.

A brief discussion of the decay scheme of RaD and the possible spin assignments of various levels is included.

I. INTRODUCTION

THE radiations of RaD and RaE have probably been studied more than any others since radioactivity was discovered.[1] Until 1939 the mode of decay of RaD was thought to be well understood. It was assumed that RaD decays in a single mode to an excited state of RaE of 46.7 kev. The maximum energy of the β-particles is around 15–30 kev. The 46.7-kev gamma-ray is mostly internally converted. The internal conversion electrons from various atomic levels were analyzed in magnetic spectrometers.[2]

Since 1939 the picture concerning the decay of RaD has become perplexing and less satisfactory. It was pointed out that the sum of the conversion electrons and the unconverted gamma-ray of the 46.7-kev line can account for only 75 percent of the disintegrations.[3–5] Subsequently additional gamma-rays were reported. After another decade of investigation with cloud chambers,[6] curved crystal spectrometers,[7] proportional counters,[8] and NaI scintillation counters[9] seven gamma-rays were observed in RaD; and several modes of its complex decay were proposed, but none of them were certain. The energies and the intensities of the reported gamma-rays may be listed as follows:

kev	46.7	42.6	37 ± 0.5	31.3 ± 0.8	23.2 ± 0.6	$16.1+0.4$	7.3 ± 0.7
Int percent	2.8 ± 0.6	0.2 ± 0.1	0.2 ± 0.1	0.4 ± 0.2	$\sim$1	0	$\sim$10

It therefore seemed desirable to investigate systematically the gamma-ray spectrum of RaD, the internal conversion electron spectrum of RaD, and the β-spectrum of RaE to seek some simplification of the problem. Furthermore the forbidden shape of the beta-spectrum of RaE seems to require a 0−0 "yes" transition[10] in order to be in accord with the parity predictions of the nuclear shell model. A reinvestigation of the mode of decay of RaD may shed some light on the spin assignment involved.

II. EXPERIMENTAL ARRANGEMENTS

(a) Proportional Counter Spectrometer

Proportional counters of conventional design[11] were used. Three counters of different cathodes and different gas fillings were employed. One counter is made with an aluminum cathode and filled with one atmos of argon+7 cm Hg of methane. The other two are brass counters, filled with one or three atmos of krypton and 7 cm of methane. The pulses were amplified by a non-overloading amplifier designed by Chase and Higen-

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* Boese Postdoctoral Fellow of Columbia University, 1952.

[1] N. Feather, Nucleonics No. 7, 22 (1949).

[2] M. Danysz, Le Radium 9, 1 (1911); J. W. Ellis, Proc. Cambridge Phil. Soc. 21, 125 (1922); L. Meitner, Z. Physik 11, 35 (1922); D. H. Black, Proc. Roy. Soc. (London) A109, 166 (1925).

[3] D. D. Lee and W. F. Libby, Phys. Rev. 55, 252 (1939).

[4] L. Cranberg, Phys. Rev. 77, 155 (1950).

[5] B. Kinsey, Can. J. Research A26, 404 (1948).

[6] Tsien San-Tsiang, Phys. Rev. 69, 38 (1946).

[7] Frilley, Gokhale, and Valadares, Compt. rend. 232, 50 (1951).

[8] Curran, Angus, and Cockroft, Phil. Mag. 40, 36 (1949).

[9] R. C. Bannerman and S. C. Curran, Phys. Rev. 81, 143 (1951); Bannerman, Lewis, and Curran, Phil. Mag. 42, 1097 (1951).

[10] A. G. Petschek and R. E. Marshak, Phys. Rev. 85, 698 (1952).

[11] Bernstein, Brewer, and Rubinson, Nucleonics 6, 39 (1950).

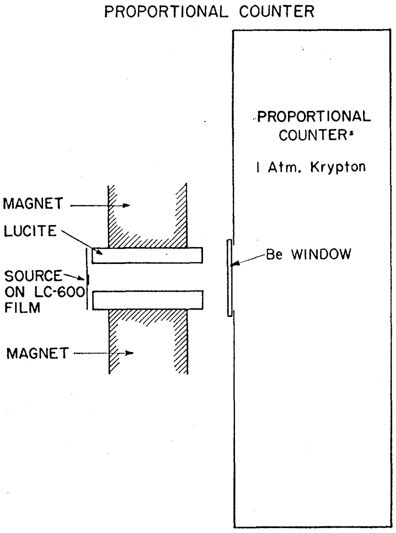

FIG. 1. Schematic diagram of the proportional counter arrangement.

botham[12] of Brookhaven National Laboratory. The pulse distribution was analyzed by a single channel pulse-height analyzer. The channel width used was small compared with the line width. All the counters gave a 10 percent resolution of the K x-ray line of Cd^{109} ($\sim$22 kev). The proportionality and stability were checked with x-rays and γ-rays from Cd^{109}, Cs^{137}, and RaD sources and found to be satisfactory.

To deflect the electrons out of the beam, a strong permanent magnet was placed between the window of the counter and the source. In order to minimize the external bremsstrahlung and to avoid the excited characteristic radiations of the pole faces of the magnet, the path of the electrons between the pole faces was lined with Lucite tubing (Fig. 1). The source used was always a thin and uniform chemical deposit of $\sim$50 μg/cm^2 on LC600 film.

The calibration of a proportional counter depends strongly on the counting rate, if the rate is high. This is caused by the change of the amplification factor as the space charge near the central wire increases. For reliable calibration this effect must be carefully considered.

(b) Magnetic β-Ray Spectrometers

Two magnetic β-ray spectrometers were used in this investigation. One is the high transmission solenoid spectrometer and the other is a 180° high resolution spectrometer. The latter has a resolution <0.8 percent and uses as a detector a Geiger counter with a window of 6 μg/cm^2 Formvar film.

(c) Sources

The RaD used in this experiment was procured through the U. S. Atomic Energy Commission from Atomic Energy of Canada, Ltd. The RaE was extracted by the dithizonate method. The RaD source for the conversion line spectrum in the 180° magnetic spectrometer was prepared by plating the RaD on a Pt wire by adding $Cu(NO_3)_2$ and NH_4NO_3 to the extracted lead solution.

III. RaD-γ-SPECTRUM

The RaD-γ-spectrum was investigated with a proportional counter spectrometer. The proportional counter used has a brass cathode and is filled with 1 atmos of krypton and 7 cm of methane. Figure 2 shows the complete γ-spectrum of RaD over the energy range of 5 kev−60 kev. Besides the pronounced and well-established 46.7 kev, its escape peak (34.2 kev) and the three L x-rays from Bi ($L_\gamma=15.7$, $L_\beta=13.0$ $L_\alpha=10.8$ kev),[13–15] the soft gamma-ray line of $\sim$8 kev can also be seen distinctly. The 23-kev line was extremely weak; nevertheless one could detect a slight hump in the inserted figure where the vertical scale is multiplied by a factor of 10. The upper limit of the intensity, however, is less than 10^{-3} per disintegration.

(a) 8-Kev Line

The uncertainty in the energy of the reported softest gamma-radiation from RaD has been large (7.3±0.7 kev). It is in the region of the characteristic K x-rays of Fe (6.4 kev), Co (6.9 kev), Ni (7.5 kev), and Cu (8.0 kev). Since the proportional counter used in our first investigation is made of brass, the origin of this line became rather doubtful. When a Tl^{204} source was investigated under the identical condition, the same line around 8 kev appeared adjacent to the three Hg L x-rays. This strongly suggested that at least part of that 8-kev line must be due to the fluorescence radiations of Cu as excited by the x- and γ-rays. To settle this point, a reinvestigation with a counter of a cathode other than brass is necessary. An aluminum counter was used for this purpose.[16] The thickness of the Be window on the Al counter is 15 mils as compared with 40 mils on the brass counter. Should it be a nuclear radiation, the 8-kev line should exhibit a more pronounced peak in the aluminum counter because of the smaller absorption in the window. The O curve of Fig. 3, taken with the Al counter, did not show the 8-kev line. Curran and his group[8] reported the strong 8-kev line from RaD in their proportional counter investigation, but no

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[12] R. L. Chase and W. A. Higenbotham, Rev. Sci. Instr. **23**, 34 (1952).

[13] The energies of these L x-rays are calculated using Frilley's (reference 14) values on the intensity distribution of the Bi L x-rays from RaD and the table of critical x-ray absorption energies prepared by Hill and his co-workers (reference 15).

[14] Frilley, Gokhale, and Valadares, Compt. rend. **232**, 157 (1951).

[15] Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).

[16] The authors are indebted to Mr. McKeown of Brookhaven National Laboratory for the Al proportional counter.

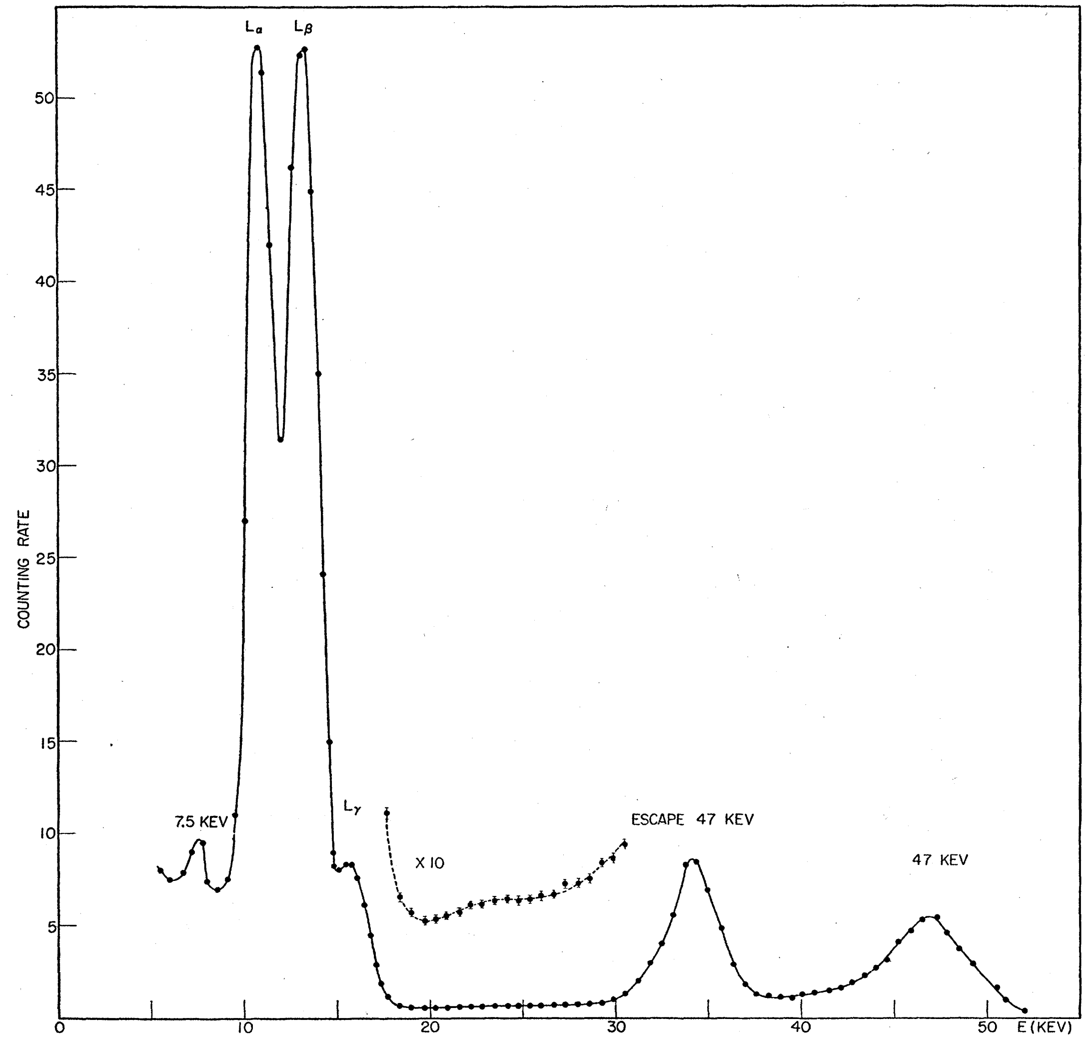

FIG. 2. Gamma-spectrum of RaD with a 1-atmos Kr proportional counter. (Corrected for detection efficiency and absorption of the window.)

mention was made on the material of their counter, nor of the source backing. Curran's results can be explained by assuming some copper or brass was near the source or counter or that the RaD source was prepared on a Ni or Cu wire. Our curve shows a strong peak at 8 kev when the RaD source is backed with a one-mm Cu sheet. Therefore, it is reasonable to interpret the reported 7.3±0.7 kev line which has been observed in both a cloud chamber and a proportional counter as a Cu or Ni fluorescence radiation.[17]

(b) 23-Kev Line

The upper limit of the intensity of the ~23-kev line is less than 10^{-3} per disintegration. The intensity reported by Tsien's group[6] is around one percent per disintegration. The intensity reported by Curran and his co-workers[8] on the 25.8-kev line which was assumed to be the known γ-ray of energy 23.4 kev, is 0.4 percent per disintegration. In some arrangements when the counting rate was high, we did detect a pronounced hump comparable to that previously reported. This suggested that the apparent hump might actually be due to a piling effect of the L_α and L_β radiation. That is to say, any two of the L x-rays may enter the counter within a certain time interval, short, compared with the resolving time of the detecting system, so that their additive pulse is registered as one of the sum of the energies. ($L_\alpha+L_\alpha$, $L_\alpha+L_\beta$, $L_\beta+L_\beta \sim$22–26 kev). There are various means one could use to distinguish between a piling effect or a real nuclear radiation. If it is due to a piling effect, then the intensity of the line should be proportional to the square of the intensity of the source instead of linearly as it should be for any nuclear

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[17] In a recent paper on RaD by A. A. Jaffe and S. G. Cohen [Phys. Rev. 89, 454 (1953)] it is mentioned that Mr. D. West (of Harwell, England) reached similar conclusions.

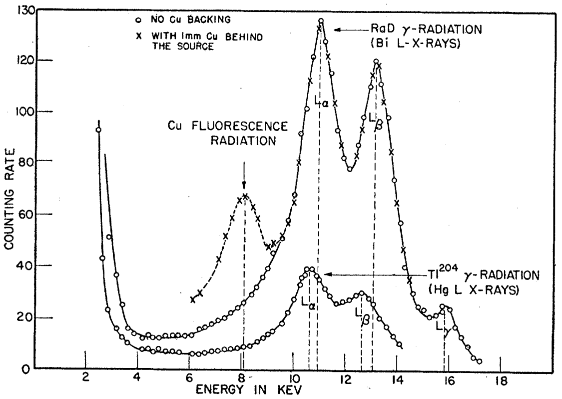

FIG. 3. Low energy part of the RaD gamma-spectrum in the Al-counter showing disappearance of the 8-kev line.

gamma-rays. Secondly, if the 23-kev line is due to the piling effect of the L x-rays, the absorption of this 23-kev peak will be greatly affected by a thin foil of Ni, as Ni strongly absorbs in the Bi L x-ray region. Figure 4 shows two curves representing the 23-kev region of the RaD gamma-spectrum from a weak and a strong source. The escape peak (34.2 kev) of the 46.7-kev line was always included for comparison. The curve of the stronger source exhibited a very pronounced hump while the weak source showed only a slight one. Two curves representing the γ-spectrum of RaD with and without a Ni absorber are shown in Fig. 5. The Ni foil was 13 mg/cm^2 in thickness. It should reduce the intensity of the 23-kev line to 0.73. However, the intensities of L_α and L_β lines should be reduced to 0.10 and 0.22, respectively. If the observed hump at 23-kev region is due to the piling effect of L_α and L_β rays, then the intensity of this piling effect should be reduced to 0.01 to 0.04. Therefore, it should completely disappear. The flat region on curve B with Ni foil demonstrates this effect very clearly.

This piling effect should, of course, be observable for a single gamma-radiation if the counting rate is high enough. Figure 6 shows the characteristic K-radiation of Cd^{109} at 22 kev, together with a small hump at the region of twice its energy (44 kev). Particularly, this piling effect may very easily be mistaken as the weak cross-over transitions when two gamma-rays are in succession.

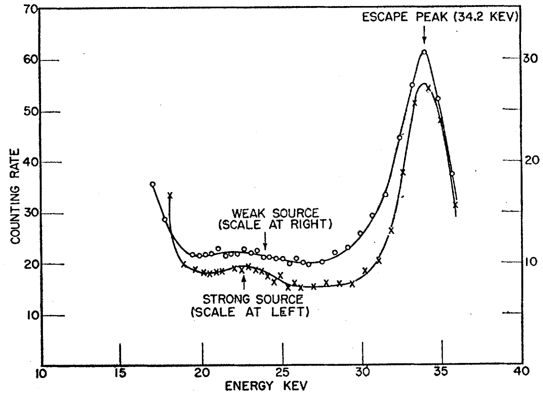

FIG. 4. Piling effect in the 23-kev region with a weak and a strong RaD source (1 atmos Kr counter).

(c) Bi L X-Rays

The three lines occurring at 15.7, 13.0, and 10.8 kev are the Bi L x-rays emitted when the excited Bi atoms from L-conversion processes return to the ground state. The identification of these lines as Bi L x-rays was made both through the energy calibration and also through the characteristic absorption. The Se K absorption edge is at 12.653 kev, while the L_α and L_β radiations of Pb and Bi are

$$\text{Bi } L_{\alpha1}=10.839\text{ kev},\ L_{\alpha2}=10.729\text{ kev},$$
$$L_{\beta1}=13.021\text{ kev},\ L_{\beta2}=12.977\text{ kev},\ L_{\beta4}=12.691\text{ kev},$$
$$\text{Pb } L_{\alpha1}=10.551\text{ kev},\ L_{\alpha2}=10.449\text{ kev},$$
$$L_{\beta1}=12.616\text{ kev},\ L_{\beta2}=12.619\text{ kev},\ L_{\beta4}=12.303\text{ kev}.$$

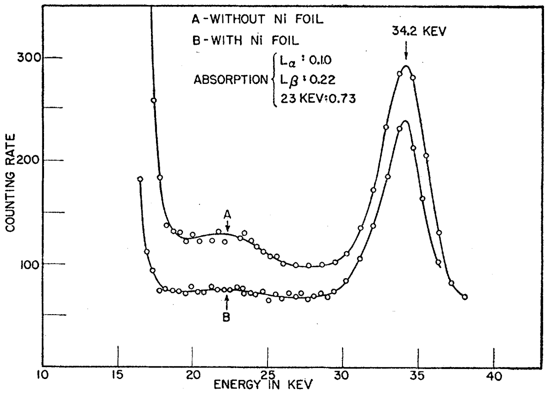

FIG. 5. Piling effect with and without the Ni absorber.

It is obvious that Pb L_α and L_β rays will not be critically absorbed by the thin Se absorber because their energy is not sufficient to excite the Se K-levels while the L_β rays of Bi will be critically absorbed by Se as the energies of $L_{\beta1}$, $L_{\beta2}$, and $L_{\beta4}$ are all above the K-edge of Se. Figure 7 shows the two curves with and without a Se absorber of $\sim$30 mg/cm^2. The identification of Bi L x-rays is established.

The ratio of $L_\alpha:L_\beta:L_\gamma$ is around 1:1:0.2, which is in reasonable agreement with Frilley's[14] results from curved crystal and again a strong demonstration of the presence of Auger transitions of the Coster-Kronig type.[14,18]

(d) Conclusions on Gamma-Rays

From the investigation of the γ-spectrum of RaD in a proportional counter, one might conclude that the 46.7-kev line is present. The two relatively strong lines

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[18] D. Coster and R. Del Kronig, Physica 2, 13 (1935); E. H. S. Burhop, *The Auger Effect* (Cambridge University Press, Cambridge, 1952).

at 8 kev and 23 kev reported in literature were detected, but the 8-kev line is the excited fluorescence radiation of Cu, and the 23-kev line is from the piling effect of Bi L x-rays. The 16.1-kev line is probably the L_γ line. The 42.6-, 37-, and 31-kev lines were not resolved in the γ-spectrum as they are too close to the 46.7-kev line and its escape peak of the 34.2-kev line. However, from the conversion electron spectrum which is shown in Fig. 8, one could put an upper limit of 0.5 percent for all these lines. Inconclusive or negative results[19,4] on the γ-lines of RaD from conversion electron spectrum have been reported from other laboratories. However, recent investigation of γ-rays of RaD on curved crystal by Ewan and Ross[20] has put an even smaller upper limit on these lines.

Energy of γ-ray (kev)	46.5	42.5	37	31	23.2	16.1
Relative intensity	1	≤ 0.015	≤ 0.016	≤ 0.015	≤ 0.025	≤ 0.06

If one uses the intensity of the unconverted 46.5 gamma-ray as 0.07 per disintegration as determined in a later section, then the intensities of the 42.6-, 37-, and 31-kev lines must be less than 0.10 per 100 disintegration. Therefore, the 46.5-kev line is the only gamma-ray in RaD of any significant intensity. The energy of this line is now 46.52 kev in agreement with previous determinations but recalculated with recent physical constants.[20]

IV. NUMBER OF CONVERSION ELECTRONS

The number of conversion electrons from the 46.5-kev line can be determined either directly from measuring the area of the conversion lines or indirectly by determining the number of L x-rays due to the internal conversions. Because of the softness of the conversion electrons, the smearing and self-absorption effect inside the source usually causes the number of conversion electrons determined directly to be too low. For instance, a value of the order of 10 conversion electrons per hundred disintegrations was obtained by Tsien.[6] Later, Cranberg[4] redetermined the conversion electron spectrum from RaD by using an extremely thin and uniform source and also calibrated the photosensitivity of the film *vs* the energies of the electrons. From the measured photometric reading of the lines, the number of photoelectrons was calculated with the knowledge of the calibrated photosensitivities and the calculated transmission of the magnetic spectrometer. The source strength was determined separately on a counter by comparing with a standard source. The total number of conversion electrons thus obtained was 74±5 per hundred disintegrations. This value is very close to the one determined indirectly from L x-rays. In applying the indirect method, one not only has to measure the number of L x-rays produced; the L-fluorescence yield, (the fraction of excited L levels yielding x-rays, the remainder decaying by Auger effect) and the contribution of the L excited states to the total number of excited levels must be used in the calculation. The number of L x-rays in RaD as determined by Stahel[21] is 25.1 per hundred disintegrations. Kinsey[5] used his calculated fluorescence yield of L x-rays of 47.5 percent and a contribution of 75 percent from the L levels to the total number of excited levels as observed in the case of ThC. His number of conversion electrons thus estimated is about 71 percent of the disintegration.

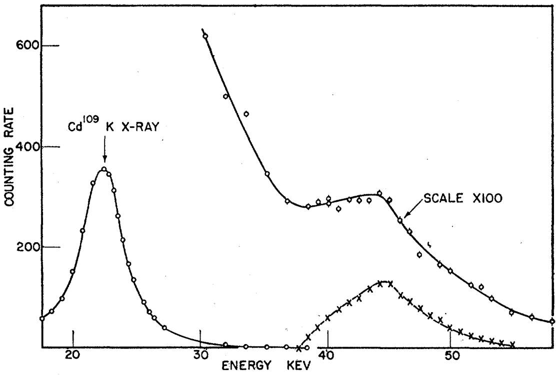

FIG. 6. Piling effect detected in Cd^{109}, K x-rays.

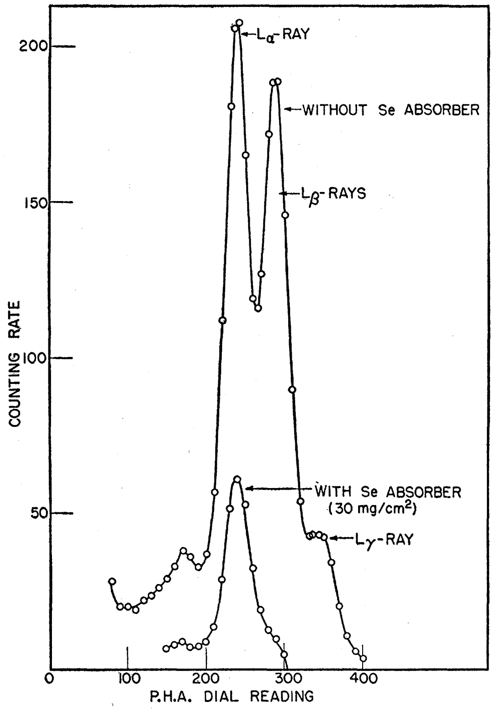

FIG. 7. Se critical absorption curve of RaD L x-rays.

To determine the number of conversion electrons per disintegration directly, a vacuum evaporated Ra(D+E) in equilibrium source on a Formvar film (~15 $\mu g/cm^2$) was prepared. The β^- spectrum of RaE and the conversion electron lines of RaD were investigated simul-

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[19] J. M. Cork *et al.*, Phys. Rev. **83**, 681 (1951).

[20] G. T. Ewan and M. A. S. Ross, Nature **170**, 760 (1952).

[21] E. Stahel, Helv. Phys. Acta **8**, 651 (1935).

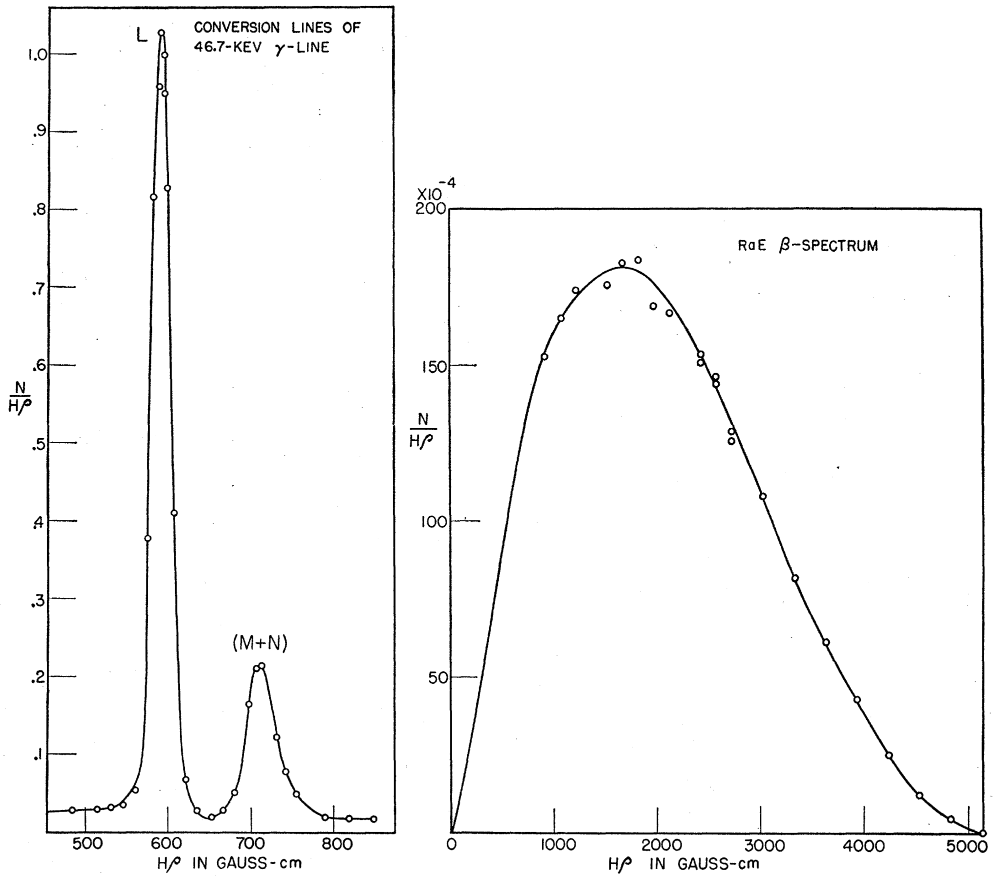

FIG. 8. Beta-spectrum and conversion electrons of Ra(D+E) in equilibrium from solenoid spectrometer.

taneously on the solenoid spectrometer. Figure 8 shows the results. The left side exhibits the L and $(M+N)$ conversion lines of the 46.5-kev gamma-ray of RaD. The right side shows the β^- spectrum of RaE. The practically zero background between the L and $(M+N)$ lines insures the extreme uniformity of the source as has been the case for most of the vacuum evaporated sources. Since the conversion lines and the β^- spectrum are taken under identical conditions, a direct comparison between the areas under each curve gives the ratio of their numbers. The ratio evaluated under the areas gives

$$N_{eL}/N_{\beta^-}=64 \text{ percent}, \quad N_{eM+N}/N_{\beta^-}=21 \text{ percent},$$
$$N_{eL+M+N}/N_{\beta^-}=85\pm5 \text{ percent}.$$

The ratio $N_{eL}/N_{eM+N}=3.05\pm0.05$ is in good agreement with Cranberg[4] and also Kinsey's value on ThC.[5]

V. THE NUMBER OF UNCONVERTED 46.5-KEV γ-RAYS

Various workers[21–24] are in good agreement on the number of unconverted 46.5-kev γ-rays. It is around 3–4 photons per 100 disintegrations. All of these results were determined by resolving the absorption curve taken by ionization chambers or Geiger counters into two components (one for the L x-ray and the other for 46.5 kev). By assuming the number of L x-rays as 25.1 per 100 disintegrations as determined by Stahel,[21] the unconverted gamma-rays can be estimated from the ratio of these two components. In most of the cases, the RaD was electroplated on Pt wire.

The gamma-spectrum which we obtained on the Kr-filled counter comprises an energy region from ~5 kev to 80 kev. All the lines are well resolved. The window and air path absorption and the efficiency of the counter have been calculated *versus* the energies. After applying all these corrections, we found the ratio between the area under the L x-rays and the area under the 46.5-kev γ-rays and its escape peak to be around 4.4±0.7. Since the fraction of excited L levels which yields x-rays is only 47.5 percent as calculated by Kinsey,[5] the ratio of atoms excited in the L levels to the number of unconverted gamma-rays must be around 9.3. The number of atoms excited in the L levels as determined directly from the conversion spectrum is 64 percent per disintegration. The number of the unconverted 46.5-kev gamma-rays therefore is around 7 percent per disintegration. Because of a much larger value of the unconverted gamma-ray as compared with previous de-

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[22] S. Bramson, Z. Physik **66**, 721 (1930).
[23] J. A. Gray, Nature **130**, 738 (1932).
[24] G. F. Van Droste, Z. Physik **84**, 17 (1933).

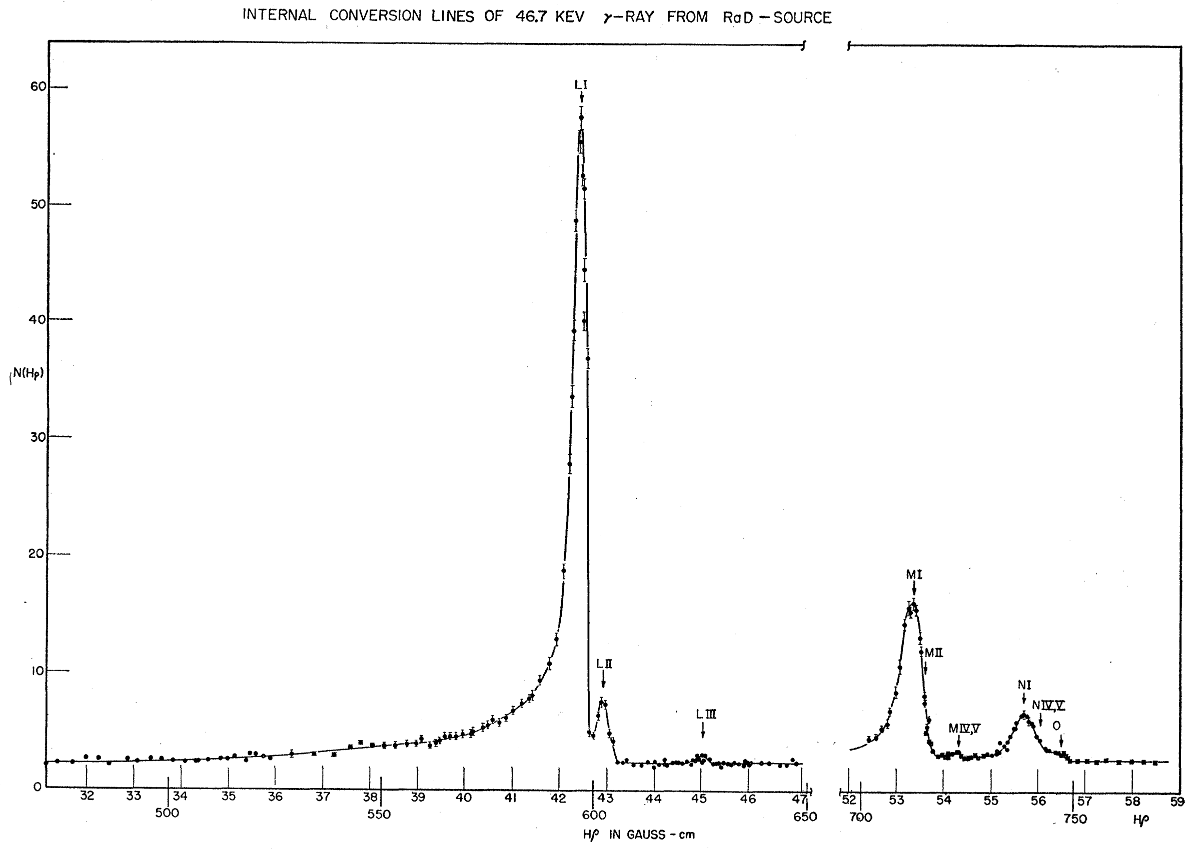

FIG. 9. Internal conversion electron spectrum from 46.5-kev gamma-ray of RaD source.

terminations, the measurements of the γ-spectrum of RaD were repeated in three different counters (1 atmos argon, 1 atmos krypton, and 3 atmos krypton), and with 4 RaD sources varying in strength by a factor of 8. The ratio between the L x-rays and the unconverted 46.5-kev γ-rays remained around 4.4. Adopting 7 percent per disintegration as the number of unconverted 46.5-kev gamma-rays, one obtains the total intensity of the 46.5-kev line as 92 ± 5 percent per disintegration.

It is quite reasonable that there are 8 ± 5 percent of the disintegrations undergoing ground-to-ground transitions. Inconclusive evidence of the presence of this group of β-particles has been reported,[25] but no details were given.

VI. L MULTIPLICITIES AND THE MULTIPLE ORDER OF THE 46.5-KEV LINE

The K conversion coefficient and the ratio of K to L conversion electrons have been most useful in assigning the multipolarities of the gamma-rays. But for a low energy gamma-ray emerging froma high Z atom as in this case, the K-level conversion is energetically impossible. However, to determine the multipolarities of the gamma-ray, one could investigate the fine structure of the L lines. Figure 9 shows the internal conversion lines of 46.5-kev γ-ray in a high resolution spectrometer. Here, one sees that the L_{I}, L_{II}, L_{III}, M, N, and O lines are well separated. The relative intensities of these lines are tabulated in Table I and compared with those of Cranberg,[4] of Curtiss,[26] and of Ellis.[27] The agreement between ours and those of Cranberg is good. The theoretical calculations on the L-conversion coefficients have been done by Gellman[28] and his group. In his calculations, the electron screening factor was not taken into consideration. Although the agreement between the theoretical conversion coefficient ($\sim$30) and that experimentally determined (9.1) is not good, the ratios of $L_{\mathrm{I}}/L_{\mathrm{II}}$ are in excellent agreement. It is quite certain that the 46.5-kev gamma-ray must be a magnetic dipole radiation (*MI*).

VII. INTERNAL CONVERSION ELECTRONS AND GAMMA-RADIATION OF RaE

RaE has 83 protons and 127 neutrons. The shell model predicts that the extra proton is in one of the states $h_{9/2}$, $f_{7/2}$, or $p_{3/2}$ and the extra neutron in $i_{11/2}$, $g_{9/2}$, or $d_{5/2}$. Therefore, from the shell model, the parity

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[25] Insch, Balfour, and Curran, Phys. Rev. **85**, 805 (1952); A. A. Jaffe and S. G. Cohen, Phys. Rev. **89**, 454 (1953).

[26] L. F. Curtiss, Phys. Rev. **27**, 672 (1926).

[27] C. D. Ellis, Proc. Cambridge Phil. Soc. **21**, 121 (1922–23).

[28] Gellman, Griffin, and Stanley, Phys. Rev. **85**, 944 (1952).

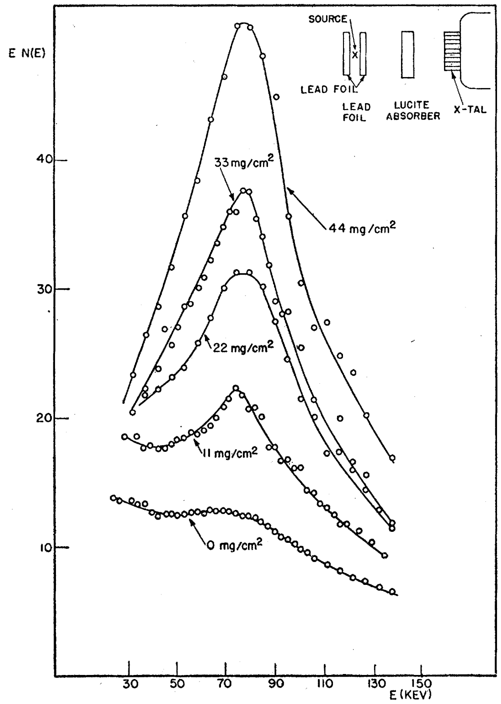

FIG. 10. Excitation of characteristic radiation with RaE source sandwiched between lead foils of various thicknesses.

is most certainly odd. The beta-transition must involve a parity change, as the parity of the final even-even Po nucleus must be even. The previous fitting[29] of RaE to second forbidden tensor interaction must therefor be discarded.

However, for first forbidden transitions, all other spectra yield the allowed shape, especially for high Z elements, (except where $\Delta I=2$ "yes," which gives a unique forbidden spectrum). Thus the forbidden shape observed in the case of RaE can be explained only by resorting to linear combinations of interaction forms, unless the RaE spectrum is complex instead of simple. A weak gamma-ray at 85 kev with an intensity of 10^{-3} per disintegration and its internal conversions electron of 66-kev energy were reported.[30]

To search for the weak gamma-ray from RaE at about 80 kev, a NaI scintillation spectrometer (Fig. 1) was used. The strong RaE source was prepared using the dithizonate method. The complete separation of RaE from RaD was assumed by the complete disappearance of the 46.5-kev line. In Fig. 10, the bottom curve shows the energy distribution of the gamma-spectrum from RaE as observed on the NaI spectrometer. The continuous background is due to the inner bremsstrahlung. A small bump superposed on the background around 80 kev has an intensity of 10^{-4} per disintegration. This is caused by the ionization of the atom accompanying beta-decay.[31] The series of curves above the bottom one were obtained by sandwiching the RaE source between lead foils of various thicknesses. It is obvious that the pronounced peak observed under those conditions is due to the excited fluorescence radiation of lead. This shows that the fluorescence radiation from a thick source of RaE (a few mg/cm²) could be quite considerable and therefore may easily be taken as nuclear radiation.

The low energy region (15–80 kev) of the RaE beta-spectrum was carefully searched for any internal conversion lines using the solenoid beta-spectrometer. The presence of a line of the intensity of 10^{-4} per disintegration should have been detected (Fig. 11).

In conclusion, no gamma-ray or internal conversions of an intensity larger than 10^{-4} could be found in RaE.

VIII. DISCUSSION AND CONCLUSIONS

With the removal of the reported gamma-rays, except the 46.5-kev line, the mode of decay of RaD may once again be regarded as simple. The transition to the excited level could account for 92±5 percent of the disintegrations. The fraction of the transition from ground state still remains to be determined. This could be 8±5 percent of the disintegrations. One might tenta-

TABLE I. Relative conversion coefficient for the 46.7-kev γ-ray of RaD source.

		L_{I}	L_{II}	L_{III}	$M_{\mathrm{I-III}}$	$M_{\mathrm{IV,V}}$	$N_{\mathrm{I-V}}$	$N_{\mathrm{VI-VIII,O}}$
Wu and Boehm		100	7.5±0.5	0.7±0.3	25±2	0.6±0.2	7±1	0.7±0.3
Cranberg		100	9±1.5	1,9±0.4	29±2		20	1
Curtiss		100	6	···	50		20	···
Ellis		100	4	1	40			···
Gellman	M1	100	8.5	0.11	···			···
(Theory)	E1	100	84.8	112	···			···
	E2	100	3450	3450	···			···

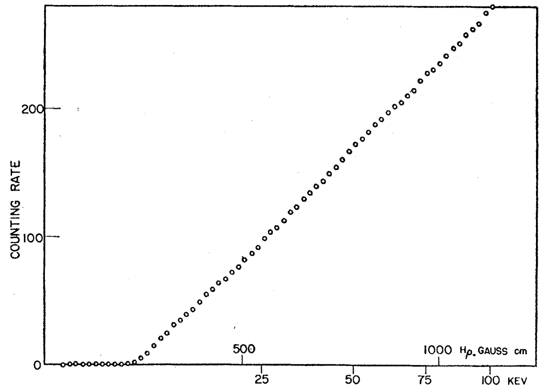

FIG. 11. Search for internal conversion lines in RaE between 15 and 80 kev.

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[29] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **60**, 308 (1941).

[30] A. S. Zavelski *et al.*, J. Exptl. Theor. Phys. (U.S.S.R.) **19**, 1136 (1949).

[31] F. Boehm and C. S. Wu (to be published); A. Migdal, J. Phys. (U.S.S.R.) **4**, 449 (1941); E. L. Feinberg, J. Phys. (U.S.S.R.) **4**, 423 (1941); J. S. Levinger, Phys. Rev. **90**, 11 (1953); H. Primakoff and F. T. Porter, Phys. Rev. **89**, 930 (1953).

tively suggest the simple decay scheme of RaD shown in Fig. 12.

The maximum energy of the low energy group of RaD last reported by proportional counter method is ~18 kev.[25] The maximum energy of the high energy group would therefore be around 64 kev. If one assumes 90 percent for the low energy group and 10 percent for the high energy group, then the respective *ft* values are 3×10^5 sec and 1×10^8 sec. The β-spectrum of RaE has a definitely forbidden shape, and the transition must be first forbidden as predicted by the parity assignment of the nuclear shell model. That is to say, the parity of the ground state of RaE must be "$-$." To interpret the β-spectrum of RaE on the first forbidden assumption, Petschek and Marshak[10] proposed the ($0\rightarrow0$ "yes") transition and the (T, P) combination of interactions. If the spin and the parity of the ground state of RaE is $(0, -)$, then the excited state must be $(1, -)$ as derived from the magnetic dipole transition of the 46.5-kev gamma-ray. It is interesting to see that the *ft*-value of the high energy group, which is again a ($0-0$ "yes") transition under this interpretation, should give a *ft*-value of $\sim10^8$ sec comparable to that of RaE. The *ft*-value of the low energy group ($0-1$ "yes") is 10^5-10^6 sec, which is low for first forforbidden transitions, but is consistent with the three exceptional cases of Hg^{205}, Pb^{209}, Tl^{206} being close to the magic numbers. The other alternative is to assign the spin of the ground state of RaE as 1 (2 is excluded; it will give a unique α-type spectrum for RaE) then the excited state of RaE could be either 0 or 2. Although the *ft*-value of 10^5 sec seems too short for a ($0-2$ "yes") transition, there are not enough known *ft*-values of the ($0-0$ "yes") type to exclude such an assignment. It would be most desirable to have the spin of RaE determined directly.

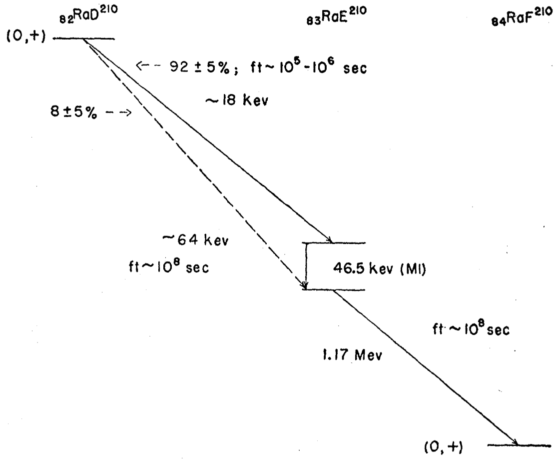

FIG. 12. Decay scheme of RaD and RaE.

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