results. A sequence of levels with characters 0+, 2+, 4+, and 6+ is to be expected from the calculations of Edmonds and Flowers[15] on the independent-particle model, assuming *jj* coupling, but the 2+, 4+, and 6+ levels should then form a fairly close triplet. The ratio of the observed energies of the 2+ and 4+ levels, 2.34, and the ratio between the observed transition probability from the 0.99-Mev level[9] and the value given by the single-particle model, agree closely with the values found for even-even nuclei having $36 \leq N \leq 88$, which Scharff-Goldhaber and Weneser[16] interpret in terms of a weak-coupling version of the Bohr-Mottelson model. The spin-6 state might then be interpreted as a member of a higher multiplet with an excitation in the zero coupling limit of $3\hbar w$ relative to the ground state. If the spin-4 state at 3.22 Mev is also regarded as a member of this multiplet, it should decay preferentially to the 2+ and 4+ members of the two-phonon triplet. The triple-coincidence data of Casson *et al.*[3] show that such transitions are $\lesssim 7$ times as strong as the competing 2.23-Mev transition.

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# Gamma Radiation in RaD and RaE Decay*

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By using chromatographically separated, carrier-free sources, the electromagnetic spectra of RaD and RaE have been investigated from 4 to 60 kev with an argon-methane gas proportional spectrometer. No gamma rays or *L* x-rays were detected in the decay of RaE. Aside from the principal 46.5-kev transition, no gamma rays (<0.2% of the 46.5-kev photons) were observed in the decay of freshly-purified RaD. However, when equilibrium mixtures of RaDEF, from spent radon seeds less than 2 years old, were examined through aluminum absorbers of thickness sufficient to eliminate pile-up peaks (20 to 26 kev) from the *L* x-rays (10 to 16 kev), a broad peak at about 31 kev appeared; it was established conclusively that in reality this is external beta-bremsstrahlung of RaE, whose low-energy region has been attenuated selectively so as to give the appearance of a gamma-ray peak.

A chromatographic column separation of millicurie quantities of carrier-free RaD and RaE from each other, as well as from RaF and macro amounts of gold and mercury impurities, is described. The time required for separation is less than 2 hours.

## I. INTRODUCTION

THE work published from 1926 to the present[1–21] on gamma radiations from the decay of RaD ($Pb^{210}$) reveals a persistent controversy concerning the existence of gamma rays of energy lower than the 46.5-kev transition which accounts at least for 87.5% of RaD

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TABLE I. Summary of gamma-ray studies in RaD and RaE decay.

| Gamma rays observed other than 46.5 kev (kev) | Source used | Method | Reference |
|---|---|---|---|
| 31, 44, 88 | 9.1 mC equilib. RaDEF | crit. abs. in Pr, Nd, Sm oxides in Xe filled ioniz. chamber | 1 |
| 32, 37 | equilib. RaDEF | diffraction from rock salt cryst. | 2 |
| 32 | equilib. RaDEF | photoelectrons in air cloud chamber | 3 |
| 26, No 31 | separated RaD and equilib. RaDEF | gas prop. counter | 4 |
| 35.4(?) | separated RaD from natural radiolead nitrate | weak conv. electrons in photographic mag. spect. | 5 |
| 32, 37 | equilib. RaDEF(?) | bent cryst. spect. with photoplates | 6 |
| 37 | separated RaD | scint. spect. | 7 |
| 31(?) | gaseous $Pb(CH_3)_4$ | gaseous source in prop. counter | 8 |
| 31 | (a) 3$\mu$C separated RaD from aged radon seeds (b) 100 $\mu$C equilib. RaDEF from natural radiolead nitrate | gas prop. counter using both gaseous $Pb(CH_3)_4$ internal source and external sources | 9 |
| 31(?) | equilib. RaDEF | weak, unassigned conv. electrons in mag. spect. | 10 |
| none | Electrodeposited RaD from radon seeds | photographic mag. spect. | 11 |
| none | separated RaD source | abs. in Al and Cu in ioniz. chamber | 12 |
| none | unspecified | abs. in Al, Sr, Pb | 13 |
| none | separated RaD | cloud chamber | 14 |
| none | $Pb(CH_3)_4$ | gaseous source in cloud chamber | 15 |
| none | RaD electroseparated from radon seeds | lens spect. with post-focusing electron accelerator | 16 |
| none (<1%) | strong (equilib.?) sources RaD | photographic mag. spect. | 17 |
| none | unspecified | cryst. spect. | 18 |
| none | unspecified | cryst. spect. | 19 |
| none | unspecified | conv. electrons in spherical condenser mag. spect. | 20 |
| none | separated RaD from natural radiolead nitrate | lens spect. and gas prop. counter with external source | 21 |
| none | carrier-free RaD separated from radon needles less than 2 years old | gas prop. counter with external source, Lucite abs. | present work |

disintegrations, according to a recent discussion by Ross, Cochran, Hughes, and Feather.[22] Gamma rays reported in the literature are listed in Table I together with the method of investigation.

Since a considerable number of radon needles less than 2 years old were on hand[23] from other work and because a new chromatographic chemical purification technique had been perfected for other studies in connection with RaDEF,[24] it was thought worthwhile to reinvestigate the low-energy gamma spectrum in carrier-free, freshly-separated RaD and RaE sources to search for the "31-kev" gamma radiation which is the subject of many reports (Table I).

## II. EXPERIMENTAL

By using a chromatographic column separation described in the appendix, carrier-free sources consisting of $6\times10^6$ dis/min RaE and about $4\times10^6$ dis/min RaD were isolated and mounted on flat glass planchets. Growth of RaE into the RaD source was followed, and indicated that the latter was quite free of RaE immediately after separation. The sources were examined immediately using a gas-filled proportional counter fitted with a 164 mg/cm$^2$ beryllium window.

The counter was filled with 2.05 atmos of a 10% methane–90% argon mixture, which was purified for 10 days by convection circulation over hot calcium turnings at about 300°C. The counter was then closed off; the pressure, resolution, and energy calibration have remained constant for a period of more than 5 months. The counter is of brass with an aluminum liner to eliminate fluorescent radiation from the brass. Within the sensitive volume, the photon path length is about 10 cm. A stabilized positive potential of 3540 volts was applied to the 4-mil diam stainless steel central wire, and the output, after amplification through a Higinbotham non-overloading amplifier, was fed through a single-channel pulse analyzer into a scaler. All points were taken manually.

The sources were counted through a Lucite absorber, and through increasing thicknesses of aluminum ab-

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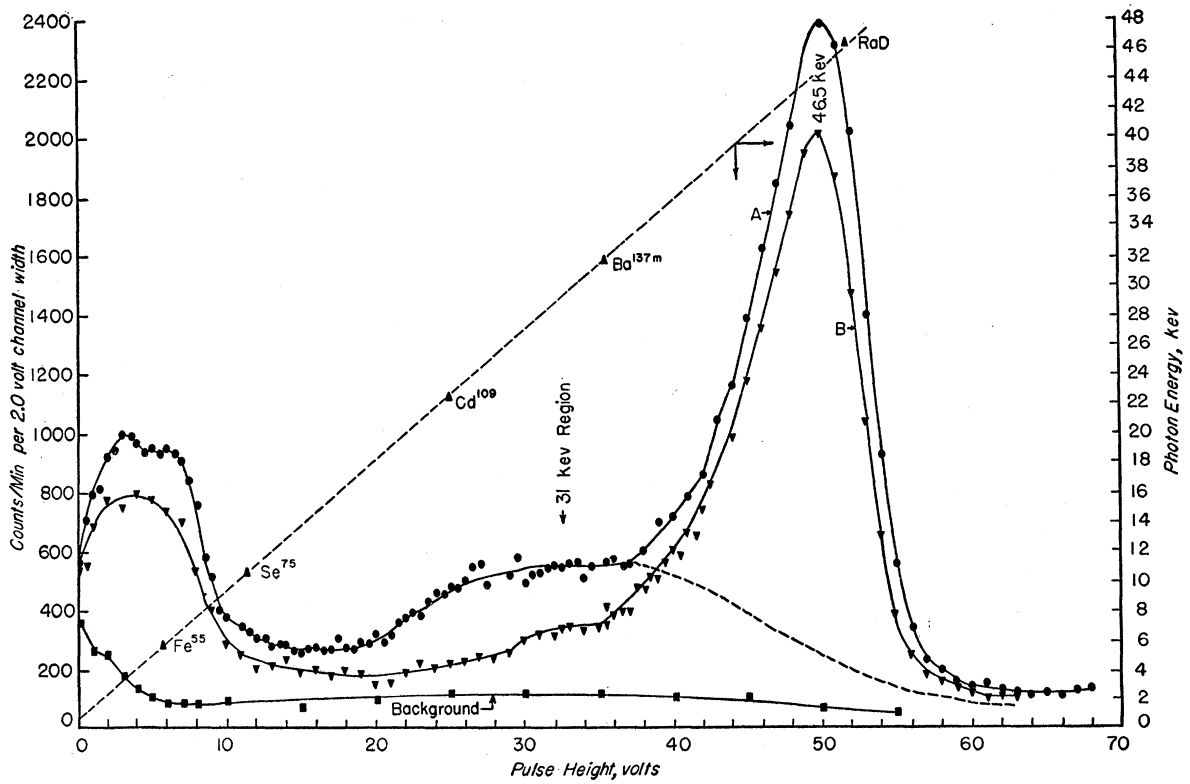


FIG. 1. Electromagnetic spectrum of equilibrium mixture of RaDEF. Curve $A$—through 861 mg/cm² aluminum absorber, representing about 10 half-thicknesses for 13-kev x-rays. Thus, Bi $L$ x-rays are highly attenuated. Note the broad hump in the region of "31 kev." Curve $B$—through 1498 mg/cm² aluminum absorber. The calibration of the instrument as well as the background is also shown, the former based on $Fe^{55}$, $Se^{75}$, $Cd^{109}$, $Ba^{137m}$, and RaD (ordinates shown to the right). A small shift in the position of the peak occurs at high counting rates due to the increased current drain in the instrument.

sorber. As is evident in Figs. 2 and 3, Lucite is far more transparent to low-energy radiation than is aluminum.

### III. RESULTS

From data taken using an equilibrium RaDEF source (Fig. 1), one might infer the presence of gamma rays from 25 to 32 kev, but examination of data taken using freshly-separated, carrier-free RaD (Fig. 2), reveals that there is no gamma radiation in this region ($<0.2\%$ of the area under the 46.5-kev peak), even when the transparency to electromagnetic radiation is increased considerably through the use of Lucite rather than aluminum absorber. Moreover, as RaE grows in, the hump reappears, the curves having the appearance of those in Fig. 1. It is evident that this effect is due to beta-bremsstrahlung from RaE whose low-energy region has been attenuated selectively by the aluminum absorber. An examination of data taken using the RaE source (Fig. 3), confirms this effect, showing how RaE beta bremsstrahlung might give the appearance of gamma radiation in the 31-kev region when equilibrium RaDE is studied in this manner. From Fig. 3, there is no evidence for gamma rays in RaE decay; furthermore, the absence of any appreciable intensity of $L$ x-rays, except those due to autoionization from beta decay which are of negligible intensity, confirms the absence of low-energy, highly-converted gamma rays.

### IV. DISCUSSION

While the data in Figs. 1–3 are conclusive in showing the absence of 31-kev gamma-rays in the decay of purified RaD, the possibility of a totally converted transition is of course not ruled out by this work. Moreover, there is a possibility that in aged, unpurified, equilibrium RaDEF sources there might exist an impurity, or hitherto undetected daughters from a rare mode of decay which would give rise to 31-kev gamma rays, but chemical purification shows that the latter cannot be assigned to the decay of either RaD or RaE. No 31-kev radiation was present in the spectra from an intact radon needle less than 2 years old. Spectra taken more recently with improved resolution (13%) exhibited a peak at 40.5 kev definitely resolved from the 46.5-kev line; however, the energy and intensity of this peak agree closely with the expected energy and intensity of the modified line due to Compton backscattering.

As pointed out by Ross, Cochran, Hughes, and

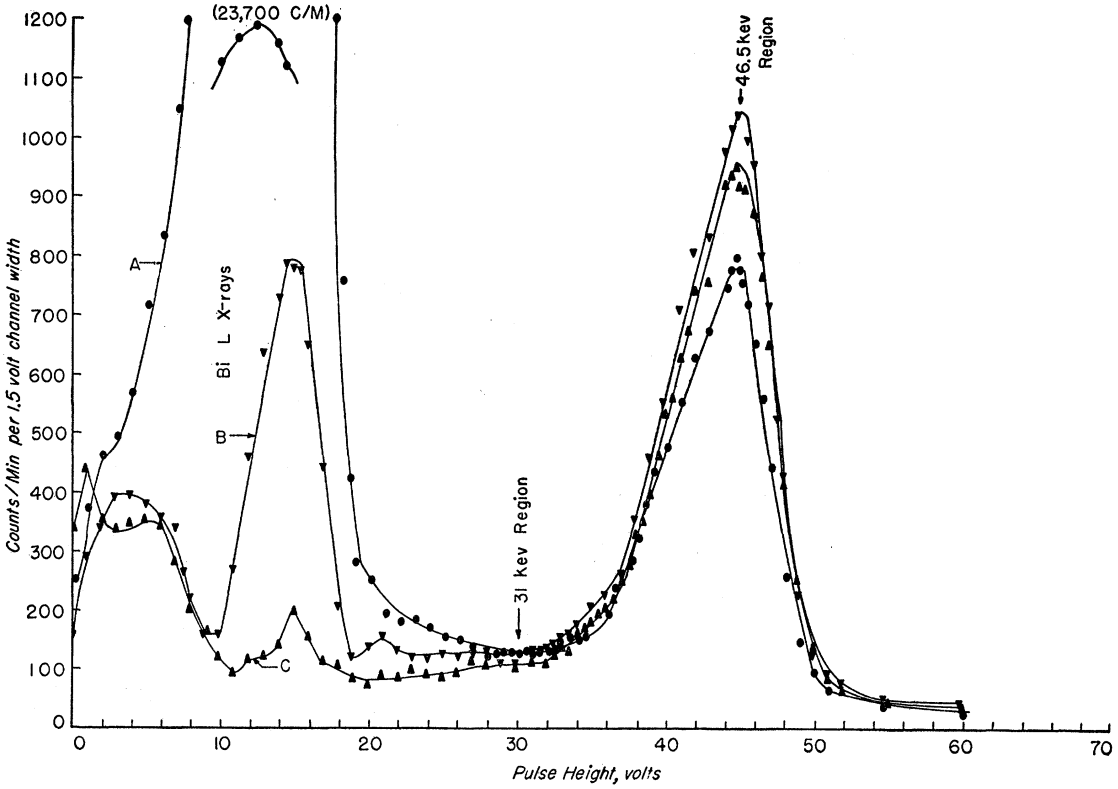


FIG. 2. Purified carrier-free RaD about 6 hr after separation. Source consisted of about $4\times10^6$ dis/min on glass planchet. Curve *A*—through 1071 mg/cm² Lucite. The Bi *L* x-ray peaks (10 to 13 kev) rise to 23 700 counts/min per 1.5-volt window (window width not calibrated). Note absence of 31-kev gamma rays. Curve *B*—through 444 mg/cm² aluminum. Bi *L* x-rays are highly attenuated. Note absence of 31-kev gamma ray. The resolution of the 46.5-kev gamma ray is about 19%, full width at half-height, because of the contribution of the 40.5-kev modified Compton backscattered line. (See discussion in text.) Later spectra taken under improved resolution (13%) clearly resolved the Compton back-scattered line. Curve *C*—through 702 mg/cm² aluminum. No 31-kev gamma ray is evident. Absence of beta bremsstrahlung from RaE shows clean chemical separation.

Feather,[22] in order to advance a convincing argument that all RaD decays proceed through the 46.5-kev transition, an accurate measurement is needed of the total number of *L* x-ray quanta emitted per disintegration and of the mean *L*-fluorescence yield of bismuth. Measurements of the former are being undertaken.

## V. ACKNOWLEDGMENTS

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## APPENDIX. PREPARATION OF CARRIER-FREE RaD AND RaE SOURCES

Recently, anion exchange resins have been applied successfully to the carrier-free separation of RaDEF,[25–27] but the only paper chromatographic separations reported[28–30] were of a type requiring as long as two days for development. Warren and Fink[31] have published a carrier-free separation using ascending paper chromatography which requires only a few minutes. Because the spent radon seeds available for this work were gold needles containing appreciable mercury from the pumps used in transferring radon, it was necessary to modify the technique so as to remove macro quantities of gold and mercury and at the same time separate RaD and RaE carrier-free.

The method[32] involves the use of a column 17 mm in diameter and about 80 cm long. Whatman No. 1 ashless cellulose powder is slurried with the eluant to be used, in this case *n*-butanol saturated with 3*N* HCl, and poured into the column; additional eluant is

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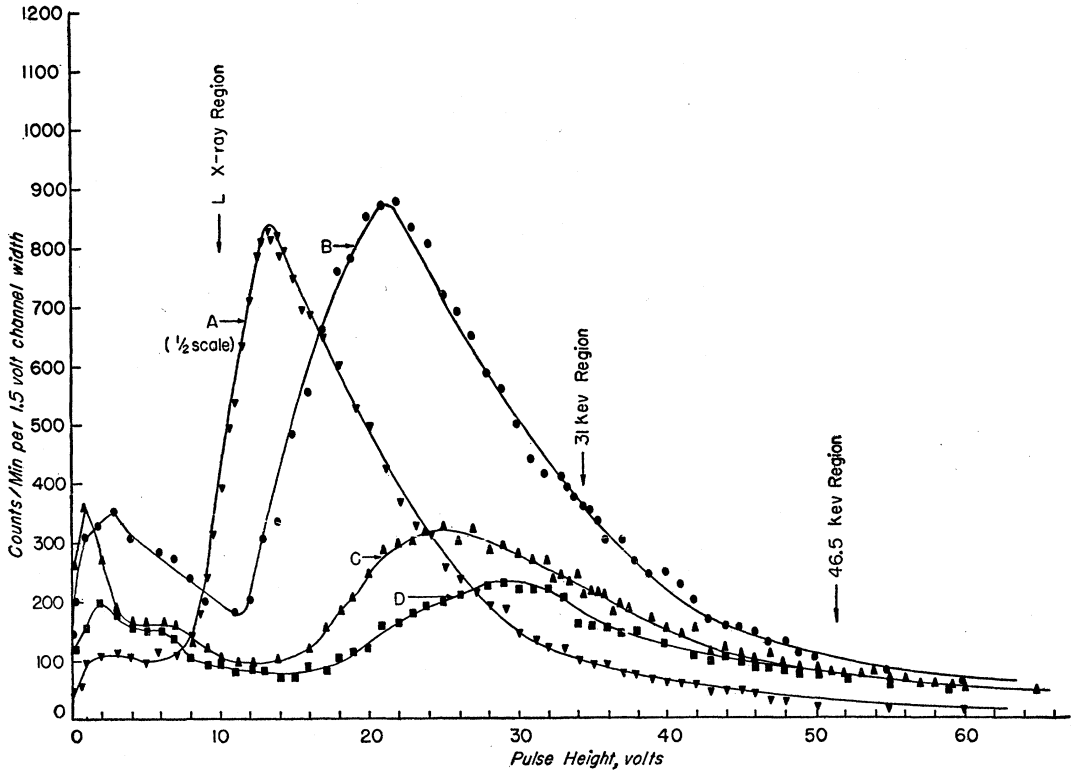


FIG. 3. Purified carrier-free RaE. Source consisted of about $6\times10^6$ dis/min on flat glass planchet. Curve *A*—through 1071 mg/cm² Lucite, but plotted ½-scale. Curve *B*—through 444 mg/cm² aluminum absorber. Curve *C*—through 702 mg/cm² aluminum absorber. Curve *D*—through 1009 mg/cm² aluminum absorber. The counter window thickness for all curves, Figs. 1–3, was 164 mg/cm² beryllium. Note absence of 46.5-kev gamma ray of RaD, showing completely clean chemical separation. Note absence of *L* x-rays, showing no converted gamma rays in RaE decay. (*L* x-rays arising from autoionization are of negligible intensity.) No gamma ray is visible in the region of 31 kev. Note the shift in the beta-bremsstrahlung peak due to selective absorption of the lower-energy region as absorber thickness is increased.

allowed to drop through the column overnight to pack down the powder to a depth of about 40 cm. The RaDEF solution containing Au and Hg is made 3*N* in HCl and mixed with the eluant and some cellulose powder, and the slurry is added to the top of the column. The system is then eluted with *n*-butanal saturated with 3*N* HCl and the fractions collected. Gold, mercury, and polonium come off first and quantitatively. About one to two column volumes later, carrier-free RaE, free of RaD (Fig. 3), comes off, and finally, one or two column volumes later, carrier-free RaD, free of RaE, is eluted. The fractions then may be evaporated to convenient volumes from which samples may be mounted. The entire elution procedure requires about 90 minutes.

It is important to note that if the macro quantities of gold and mercury are absent, our faster method[31] should be employed.