To my parents.
The Study of β Disintegration in Nuclear Emulsions.

Thesis submitted by

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B.Sc.

for the Degree of
Doctor of Philosophy

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Chapter I.

Introduction.

Radium D is lead 210, with 82 protons and 128 neutrons. It is a member of the uranium series of radioactive substances \( A = 4n + 2 \), the lower members of which are

\[
\alpha \rightarrow \text{Ra} \rightarrow \text{Rn} \rightarrow \alpha \rightarrow \text{RaA} \rightarrow \alpha \rightarrow \text{RaB} \rightarrow \alpha \rightarrow \text{RaC} \rightarrow \alpha \rightarrow \text{RaD} 
\]

The modes of disintegration of the succeeding members of the chain down to stable lead 206 are shown in some detail in fig. 1. When the present work was undertaken, it was known that the main mode of disintegration of the RaD nucleus was by \( \gamma \)-decay to an excited state of RaE followed by a direct transition to the ground state. Yet, while the experimental evidence suggested that this mode of disintegration did not occur in all cases, no alternative mode had been detected in a wide variety of experiments using several methods of electron and \( \gamma \)-ray spectroscopy. The present work began as an open-minded attempt to see whether the nuclear emulsion method, with its own peculiar advantages and disadvantages, could throw new light on any of the features of the RaD disintegration, particularly on the possibility of the existence of a hitherto undetected mode of disintegration. Information was indeed obtained on the latter topic, and this provided a stimulus for preparing a confirmatory experiment using two proportional counters in anti-coincidence.
0+ [RaD (Pb\textsubscript{210})] 22 y.

\[ \beta^- \sim 15 \text{ keV}, \log f_t \sim 5.5 \]

0−

1− [RaE (Bi\textsubscript{214})] 5 d.

\[ \beta^- 1.17 \text{ MeV} \]

\[ \log f_t = 8 \]

0+ [RaF (Pb\textsubscript{210})] 138 d.

\[ \Delta 5.3 \text{ MeV} \]

\[ \text{Pb}_\text{82} \]

**FIGURE 1**
2.

The History of Experiments on RaD.

Soon after the separation of radium from pitchblende by Madame Curie in 1898, several radioactive substances were isolated from uranium-bearing minerals, among them an active form of lead called radon-lead. The active constituent of this was later named radium D.

In 1911, following the discovery by von Beyer, Hahn and Meitner (1) of the monoenergetic groups present in electron spectra, Danysz (2) performed the first electron-spectroscopy of RaD using the semi-circular focusing method which he had introduced. He found four discrete lines. When the $\gamma$-ray conversion mechanism was explained by Rutherford (3) in 1914 these lines could be interpreted as the conversion electron lines of monoenergetic $\gamma$-rays of about 47 keV converted in various levels of the electrons surrounding the emitting nucleus. From 1922 to 1926, the experiment of Danysz was repeated with greater accuracy by Meitner (4), Ellis (5), Black (6) and Curtiss (7). The work of Curtiss in 1926 was particularly accurate and is of some historical interest since it was one of the three experiments which finally settled the question of whether the $\gamma$-ray is emitted before or after disintegration. In disintegrations where a $\gamma$-ray of energy $h\nu$ is emitted, the conversion electrons have energies $h\nu - E_p$ where $E_p^Z$ is the ionisation energy of the P-shell (say) of an atom whose atomic number is Z. In 1925,
Meitner (8) had shown that in the case of $\alpha$-disintegrations (she used $\text{RaAc}$ and $\text{AcX}$) the appropriate $Z$ was that of the daughter and not that of the parent nucleus, and that, therefore, the $\gamma$-ray was emitted and converted after the nuclear transformation. To establish this conclusion for $\beta$-emitters by the same method was more difficult since in this case $Z$ changes by only one, and so the distinction between parent and daughter nucleus is more critical. Curtiss met this difficulty by choosing $\text{RaD}$, whose $\gamma$-ray energy of about 47 keV is of the same order as the ionisation potentials involved. He was able to show conclusively that the $\gamma$-ray was converted in the atomic shells of the daughter nucleus. Experiments performed independently by Ellis and Wooster (9) with $\text{Ra}(\text{B} + \text{C})$ gave the same result.

The net conclusion from these experiments in the electron spectroscopy of $\text{RaD}$ was that $\gamma$-rays of a single energy are emitted during the disintegration.

In 1927, Kikuchi (10) suspended a fibre impregnated with an equilibrium source of $\text{Ra}(\text{D} + \text{E} + \text{F})$ in a cloud chamber. Since he did not observe any cases in which a $\text{RaD}$ conversion electron track was accompanied by the track of a primary $\beta$-particle, he concluded that the energy of the $\beta$-particles could not exceed a few keV. Petrova (11) in 1929 did a similar experiment with low pressure filling, and he put the maximum energy at about 10 keV. An ingenious variant of this cloud
chamber technique was introduced in 1937 by Richardson and Leigh-Smith (12) who used gaseous lead (RaD) tetramethyl as their source, and so avoided the difficulties of absorption which are encountered when a solid source is mounted on a solid support. They found that the maximum energy of the $\gamma$-emission was about 20 keV.

At this time, then, the disintegration of RaD was thought to occur in a single mode: a very low energy $\gamma$ decay to a 47 keV excited state of RaE followed by a direct transition to the ground state.

However, in 1939, Amaldi and Rasetti (13), using a method of selective absorption, reported finding a $\gamma$-ray of 43 keV, and this was the first of a train of experiments which seemed to show that there were intermediate levels between the 47 keV and the ground states of RaE. Talen (14) confirmed the existence of the 43 keV $\gamma$-ray and in a later experiment (15), using selective absorption in foils of the rare earths, he reported additional $\gamma$-rays of 37 keV and 32 keV together with K X-rays of 77 keV and 87 keV. These were confirmed by Frilley (16) by means of crystalline diffraction $\gamma$-spectroscopy. In two further experiments, Talen and Marty (17) allowed a beam of $\gamma$-rays from RaD to ionise the filling of a cloud chamber and measured the quantum energies by observing the lengths of the tracks of the photoelectrons. In addition to the previously reported $\gamma$-rays, they found others at 7 keV, 23 keV and about
In the post-war years there has been a recession from this high-water mark of reported $\gamma$-rays. Salgueiro (18) in 1944, repeating Frilley's crystalline diffraction experiment observed the 32 keV $\gamma$-ray but no others (apart from the 47 keV line). In 1949 Curran et al. (19), performing $\gamma$-ray and X-ray spectroscopy by means of the proportional counter technique which he had largely developed, found the 23 keV line and the intense line at 7 keV previously reported by Tsien and Marty. In 1950, Cranberg (20), studying the electron spectrum of RaD, reported a conversion electron line at an energy appropriate to the conversion of a 32 keV $\gamma$-ray but he gave reasons for doubting its genuineness. Frilley, Gokhale and Valdéses (21) in 1950, using a curved mica crystal for the diffraction and focusing of $\gamma$-rays, saw the 32 keV line clearly; the 37 keV line was hardly visible and no other lines were seen. An experiment by Bannerman and Curran (22) in which $\gamma$-rays from RaD were counted by two scintillation counters in coincidence seemed to show that $\gamma$-rays of 16-30 keV were emitted (almost) simultaneously that is, that they were in cascade, and, in a later paper by Insch, Balfour and Curran (23) this cascade

This experiment was later repeated by Damon and Edwards (49) and no $\gamma$-$\gamma$ coincidences were found.
was assumed to explain the presence of a great number of small pulses in the $\gamma$-spectroscopy of RaD by proportional counter. The final (to date) appearance of the 7-37 keV $\gamma$-rays in the literature was in 1953 when Damon and Edwards (24) reported finding evidence for all the previously observed $\gamma$-rays and K X-rays in their proportional counter pulse spectrum.

However, by 1953, it had become virtually certain that all of the previously reported $\gamma$-ray transitions in the region of 7-37 keV do not in fact occur, or, if they do, they are of negligible intensity. The electron spectrum of RaD has been carefully examined by means of high resolution magnetic spectrometers, by Cranberg (20) in 1950, by Cork et al. (25) in 1951, by Bashilov et al. (26) in 1953, and by Wu et al. (27) in 1953. Apart from one unexplained line obtained by Cranberg from only one of several RaD sources, these workers are in agreement that there are no detectable electron conversion lines associated with $\gamma$-rays of $\lambda_2$, 37 or 31 keV. Work of lower resolving power by Ouang et al. (28), Braga (29), Butt and Brodie (30) and Pringle (31) is also in agreement. Further work by Wu et al. (26) using a proportional counter for $\gamma$-ray spectroscopy showed that the 7 and 23 keV $\gamma$-rays also did not occur, or were of undetectably small intensity. In 1952, the RaD $\gamma$-spectrum in the region of 10-60 keV was examined by G.T. Ewan and M.A.S. Ross.
by means of a curved crystal spectrometer, and none of the disputed \( \gamma \)-rays in the region were detected. It seems at present, therefore, that only the 47 keV excited level of RaE plays any part in the RaD \( \rightarrow \) RaE disintegration.

Is RaD \( \rightarrow \) RaE \( \rightarrow \) RaE the only mode of disintegration? If it were, one would expect each disintegration to be associated with either an emitted \( \gamma \)-ray or an emitted conversion electron. When the present work was begun, the most reliable estimated intensity of excitation of the 47 keV state (i.e. \( \gamma \)-ray intensity added to conversion electron intensity) was 77.5 \( \pm \) 5 per hundred disintegrations (20), leaving some 20\% of disintegrations to be accounted for. It therefore seemed worth while to study the decay by a technique which had not yet been applied to the problem - the nuclear emulsion technique - in order to see whether any contribution could be made. The most recent \( \beta \)-spectroscopic experiments by Wu et al. and by Bashilov et al. are widely discrepant in their conversion electron intensities, and do not remove the desirability of studying the decay by an independent technique.

**Summary of Experimental Information on RaD.**

RaD is probably one of the most studied of radioactive substances. Yet, although the decay scheme is, according to the present view, an extremely simple one,
there is a marked lack of agreement in the experimental estimations of its quantitative features. The disintegration energy, about 60-70 keV, is so low that it is difficult to make accurate measurements and to eliminate instrumental errors. Since in the present work, it will be necessary to make use of the quantitative observations of others, this section is devoted to a critical summary of the evidence on the various features of the decay.

General.

RaD, with 82 protons and 128 neutrons is an even-even nucleus and therefore, in its ground state, the total angular momentum \( I \) of its nucleus is 0 and its "parity" is even (fig. 1). The "spin" of the RaD nucleus in its ground state has been recently determined by Smith (33) to be 1. Its parity, from considerations of shell-model theory and of \( \beta \)-decay systematics, is necessarily odd. As will be seen later, the 47 keV de-excitation is a magnetic dipole transition, which implies that the parity of the excited state is also odd. The selection rules of the \( \gamma \)-transition, together with consideration of the ft.-value of the \( \beta \)-transition indicate that the spin of the excited state is 0.

Since, according to the shell-model theory, 82 and 126 are magic numbers, it should be noted that RaD has two neutrons and no protons outside closed shells and
RaB has one neutron and one proton.

The $\beta$-transition.

The $\beta$-transition is first forbidden spin-favoured ($0 \rightarrow 0$, yes), and it is virtually certain that, ignoring screening corrections, its spectrum has the "allowed" shape:

$$N(W)dw \propto W(W_0 - W)^2 F(z,W) dw.$$ 

This will be discussed more fully in a later section (page 84). The log ft. value is 5-6 which is in line with other ($\Delta I = 0$, yes) transitions in the doubly magic region (34). The reported half-lives are:

<table>
<thead>
<tr>
<th>Worker</th>
<th>Date</th>
<th>$T$ (years)</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Schweidler</td>
<td>1929</td>
<td>22.1 ± 0.5</td>
<td>Ionisation chamber over 15 y.</td>
</tr>
<tr>
<td>Wagner</td>
<td>1950</td>
<td>25</td>
<td>Ionisation chamber over 250 d.</td>
</tr>
<tr>
<td>Tobailem</td>
<td>1955</td>
<td>19.40 ± 0.35</td>
<td>Differential ionisation chambers</td>
</tr>
</tbody>
</table>

Maximum Energy. The "end-point" energy of the spectrum has been variously reported as follows:
<table>
<thead>
<tr>
<th>Worker</th>
<th>Date</th>
<th>End-point</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Petrova</td>
<td>1929</td>
<td>~10 keV</td>
<td>Cloud chamber, solid source.</td>
</tr>
<tr>
<td>Richardson</td>
<td>1937</td>
<td>~16 keV</td>
<td>Cloud chamber, gaseous source.</td>
</tr>
<tr>
<td>Lee</td>
<td>1939</td>
<td>25.5 ± 1.0 keV</td>
<td>Geiger counter, gauze wall.</td>
</tr>
<tr>
<td>Saha</td>
<td>1946</td>
<td>29.2 keV</td>
<td>Geiger counter, gauze wall.</td>
</tr>
<tr>
<td>Bannerman</td>
<td>1952</td>
<td>18 ± 2 keV</td>
<td>Scintillation counters.</td>
</tr>
<tr>
<td>Ingech</td>
<td>1952</td>
<td>18 ± 2.5 keV</td>
<td>Prop. counter, solid source, 2.</td>
</tr>
<tr>
<td>Jaffe</td>
<td>1953</td>
<td>15.2 ± 1.0 keV</td>
<td>Prop. counter, gaseous source.</td>
</tr>
<tr>
<td>Huster</td>
<td>1953</td>
<td>23.0 ± 2.5 keV</td>
<td>Prop. counter, gaseous source.</td>
</tr>
<tr>
<td>Kobayashi</td>
<td>1953</td>
<td>16-18 keV</td>
<td>Electrostatic -spectrometer.</td>
</tr>
<tr>
<td>Lewis</td>
<td>1955</td>
<td>17 ± 2 keV</td>
<td>Source in NaI crystal.</td>
</tr>
</tbody>
</table>

The work of Petrova is early and unreliable. The high values obtained by Lee and by Saha (who used similar methods) may be discarded since some systematic fault in their arrangement led to results which, taken as a whole, are not consistent with known facts (see For the sake of brevity, it will sometimes be convenient to refer only to the first-named author of each reference.
below, intensity of conversion electrons). The results of Richardson, Bannerman, Insch, Jaffe and Lewis are consistent with each other and indicate a maximum energy of about 16 keV. Kobayashi's observations also support this value although the nature of his spectrum did not lend itself to a reliable determination of the end-point. The high value observed by Huster is markedly outside the 15-18 keV range reported by the other six workers, although it may be noted that, taking the extremes of the quoted uncertainty limits, it is barely consistent with the result of Insch.

On the question of the intrinsic reliability of the methods used, whereas the results of Bannerman and of Insch depend essentially on the single observation of the point at which the ordinate of their spectra becomes zero, those of Jaffe and of Huster (and probably that of Lewis) are the result of extrapolating a linear Kurie plot, embodying many observed points, to the energy axis.

It is unfortunate, therefore, that the experiments of Jaffe and Huster, which should be the most accurate, have produced the two extreme values, values which are utterly discordant with each other. It would seem that a considerable systematic error has occurred in one or other of the experiments; Jaffe's value is preferable since (a) it is more in line with the values obtained by other workers, (b) his resolution is
markedly better than that of Huster; and (c) the work of Lewis contradicts the conclusions of Huster (and supports those of Jaffe) not only in the maximum energy of the $\beta$ spectrum but also in its shape at energies below 10 keV. The best conclusion from the available evidence would seem to be that the end-point is probably around 15-16 keV but may be as high as 20 keV.

**Shape of Spectrum.** According to Jaffe and Cohen (35), the shape follows the allowed shape down to about 3.5 keV. Below 3.5 keV there are probably fewer electrons than predicted by the theory. Lewis (44) reports that his work is consistent with the result of Jaffe and Cohen, but not consistent with that of Huster (36) whose observed spectrum begins to fall below the allowed shape at 10-12 keV.

**The 47 keV Excited State of RaE.**

**Energy.** The most accurate value of the energy is probably that found by G.T. Ewan and M.A.S. Ross by curved crystal diffraction of the $\gamma$-ray. Their value is

$$46.52 \pm 0.02 \text{ keV}.$$  

The value found by the indirect method of adding the measured energy of the $L_I$ conversion electrons (7) to the ionisation potential of the $L_I$ level (42) is 46.55 keV which is in agreement.
Fink P.R. 106, 266, 1957.

V ray 4.5 %
L-V ray 24 %
ratio 6:1
Intensity of the $\gamma$-ray. The experimental values for the number of $\gamma$-ray quanta emitted per 100 disintegrations are as follows:

<table>
<thead>
<tr>
<th>Worker</th>
<th>Date</th>
<th>No. of quanta per 100 disintegrations</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bramson</td>
<td>1930</td>
<td>3.1 ± 1.2</td>
<td>Ionisation chamber.</td>
</tr>
<tr>
<td>Stahel</td>
<td>1931</td>
<td>2.4 ± 0.7</td>
<td>Ionisation chamber.</td>
</tr>
<tr>
<td>Gray</td>
<td>1932</td>
<td>&lt; 4</td>
<td>Ionisation chamber.</td>
</tr>
<tr>
<td>von Droste</td>
<td>1933</td>
<td>3.6</td>
<td>Cloud chamber.</td>
</tr>
<tr>
<td>Wu</td>
<td>1953</td>
<td>7</td>
<td>Indirect derivation.</td>
</tr>
<tr>
<td>Damon</td>
<td>1954</td>
<td>3.8 ± 0.6</td>
<td>Scintillation counter.</td>
</tr>
</tbody>
</table>

The result of Damon and Edwards is regarded as the most reliable one. It was obtained by the direct counting of quanta by means of a NaI scintillation counter. The source strength was determined by counting $\beta$-particles from the polonium in the RaD-E-F source.

The meticulous work of Bramson is probably reliable within the stated error and is in agreement. Stahel and Simoo corrected for the effect of non-existent $\gamma$-rays from RaE and for the escape of secondary radiation from their CH$_3$I filling and their value is possibly too low.

The discordant value of Wu et al. is not a direct
determination; it is a combination of the values of
(a) the intensity of L-conversion electrons (b) the
ratio of the intensities of L X-rays to emitted γ-rays
and (c) the L-fluorescence yield as reported by Kinsey
(50). There is considerable uncertainty about all of
these quantities. The reported values for (a) are not
in agreement, as will be seen later. For (b) Wu et al
finds a value of 4.4 ± 0.7 whereas Hughes (51) and
Curran et al. (19), using the same method, obtain
6.74 ± 0.5 and 11 respectively. Kinsey's value for
(c), i.e. 4.7%, has been questioned by Damon and Edwards
(49) and by Ross et al. (51). Damon asserts that
Kinsey's results are uncertain within the range 30 -
50% and he himself puts the fluorescence yield at 30 ± 5%. Ross also concludes from an examination of all
the relevant data that 4.7% is too high and chooses a
value of 40 ± 2%. Considering these large uncertain-
ties, especially in the value of the fluorescence yield,
it can be concluded that no great weight should be
given to the high value of Wu et al.

Moreover, as Ross (51) has pointed out, the
theoretical value for the internal conversion coeffic-
ient of the γ-ray (58) leads to a value of 3.5 γ-ray
quanta per 100 disintegrations, which supports the ob-
servation of Damon and Edwards. The latter value
(3.8 ± 0.6%) will therefore be given most weight in
this thesis.
15.

The Conversion Electrons - Absolute Intensities. A summary of reported results is given in Table IV.

<table>
<thead>
<tr>
<th>Worker</th>
<th>Date</th>
<th>No. of conversion electrons per 100 disintegrations</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kikuchi (10)</td>
<td>1929</td>
<td>~90</td>
<td>Cloud chamber.</td>
</tr>
<tr>
<td>Stahel (53)</td>
<td>1931</td>
<td>83</td>
<td>Geiger counter.</td>
</tr>
<tr>
<td>Lee (37)</td>
<td>1939</td>
<td>5-10</td>
<td>Geiger counter, gauze wall.</td>
</tr>
<tr>
<td>Ouang (28)</td>
<td>1943</td>
<td>8  4  12</td>
<td></td>
</tr>
<tr>
<td>Cranberg (20)</td>
<td>1950</td>
<td>54 20 74 ± 5</td>
<td>Magnetic spectrometers.</td>
</tr>
<tr>
<td>Butt (30)</td>
<td>1951</td>
<td>&gt;47 ± 4 &gt;12</td>
<td></td>
</tr>
<tr>
<td>Bashilov (26)</td>
<td>1953</td>
<td>45 ± 2 13 ± 1 58 ± 3</td>
<td></td>
</tr>
<tr>
<td>Wu (27)</td>
<td>1953</td>
<td>64 21 85 ± 5</td>
<td></td>
</tr>
</tbody>
</table>

Of the latter four results, those of Butt and Brodie, and of Bashilov et al. are probably too low and certainly comparatively unreliable because of the uncertainty of correcting for the backscattering of electrons from the source backing. (They used respectively 0.22 mg./cm.² of gold and 1.35 mg./cm.² of aluminium as backing). Cranberg and Wu avoided this difficulty by using vacuum evaporated sources on thin films of 5 and 15 μg./cm.² respectively. Of these two, the result of Wu is probably the less liable to error. Cranberg used photographic detection of the
electrons and his result depended on calculation of the transmission of the spectrometer, and on subsidiary experiments to find the sensitivity of his photographic film at various energies, and the absolute strength of his sources. In the experiment of Wu et al., the electrons from the Ra(D + E) source were counted directly by means of an extremely thin-windowed (~ 0.001 cm. 2) Geiger counter, and the absolute intensity of the conversion electron lines was estimated by comparing their areas with that of the RaE β spectrum measured on the same run. While the results of Wu et al. seem therefore to be the best available, it should be noted that a conversion electron intensity of about 80% would be barely consistent with both of these two most reliable observations.

The Conversion Electrons - Relative Intensities. The resolution of the conversion lines obtained by Wu, Boehm and Nagel (27) is the best so far reported; their values for the relative intensities are shown in Table V, together with those of several other workers, and compared with the values predicted by Gellman et al. (54) for γ-transitions of the indicated multipolarity. The comparison shows that the 47 keV transition is entirely or almost entirely "magnetic dipole" (W 1) (see page 8 for the implications of this).
<table>
<thead>
<tr>
<th></th>
<th>$L_I$</th>
<th>$L_{II}$</th>
<th>$L_{III}$</th>
<th>$M_{I-III}$</th>
<th>$M_{IV V}$</th>
<th>$N_{I-V}$</th>
<th>$N_{VI-VIII}$,0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wu (27)</td>
<td>100</td>
<td>7.5 ± 0.5</td>
<td>0.7 ± 0.3</td>
<td>25 ± 2</td>
<td>0.6 ± 0.2</td>
<td>7 ± 1</td>
<td>0.7 ± 0.3</td>
</tr>
<tr>
<td>Cranberg (20)</td>
<td>100</td>
<td>9 ± 1.5</td>
<td>1.9 ± 0.4</td>
<td>29 ± 2</td>
<td></td>
<td>20</td>
<td>1</td>
</tr>
<tr>
<td>Bashilov (26)</td>
<td>100</td>
<td>15 ± 3</td>
<td>0.9 ± 0.3</td>
<td>26 ± 3</td>
<td></td>
<td>8.0 ± 0.3</td>
<td></td>
</tr>
<tr>
<td>Curtiss (7)</td>
<td>100</td>
<td>6</td>
<td></td>
<td></td>
<td>50</td>
<td>20</td>
<td></td>
</tr>
<tr>
<td>Ellis (5)</td>
<td>100</td>
<td>4</td>
<td>1</td>
<td></td>
<td>40</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gellman (54), N 1</td>
<td>100</td>
<td>8.5</td>
<td>0.60</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Theory)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>E 1</td>
<td>85</td>
<td>112</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>E 2</td>
<td>3450</td>
<td>3450</td>
</tr>
</tbody>
</table>
Absolute Intensity of L X-rays. The absolute intensity of emission of L X-rays per 100 Rd disintegrations has been given as 30 by Gray (47), 25.1 by Stahel (55), 27 by Taeien (15) and 19 ± 3 by Damon and Edwards (49). Only the values of Damon and of Stahel merit consideration.

Stahel counted the number of L X-ray quanta indirectly, by observing the current produced in an air-filled ionisation chamber irradiated by the X-rays. The connecting relation is

\[ i = n_L \omega a \frac{E}{E_0} \]

where \( i \) is the current; \( n_L \) the number of quanta leaving the source; \( 4\pi \omega \) is the solid angle subtended by the source at the ionisation chamber window; \( a \) is the fraction of quanta absorbed in the air filling; \( E \) is the mean energy of the quanta; \( E_0 \) is the mean energy required to produce an ion-pair in the filling; \( e \) is the charge of the electron. It is clear, first of all, that although no error is indicated, it must be very much greater than that implied by recording the result to three significant figures. Bramson, who used a similar method (45) for the intensity of the 47 keV \( \gamma \)-ray, gave her result as (3.1 ± 1.2)%; this is a 39% error. Yet, whereas Bramson was observing monoenergetic \( \gamma \)-rays whose energy was quite accurately known from the work of others, Stahel was concerned
19.

with a discrete X-ray spectrum of many components ranging from 10.8 to 16.3 keV. He was obliged to use in his calculations a "mean wavelength" found by measuring the absorption in foils of copper, silver and aluminium, and then to use an air absorption coefficient corresponding to this wavelength. The "mean wavelength" thus appears twice in the formula, since it also governs the mean number of ion-pairs per quantum. From these considerations alone, it is to be expected that Stahel's result is subject to a percentage error of the same order as that of Bramson.

An examination of Stahel's data together with tables of experimental (59) and theoretical (60) absorption coefficients, appears to show that either (1) $E$ is correct but $a$ is too low (factor of about 16\%) or (2) $a$ is correct but $E$ is too low (factor of about 8\%). In either case, the reported intensity would be reduced by the indicated factor, and so the result of Stahel is probably too high. But however that may be, it is clear that the difficulties of the experiment were such that the reported intensity is subject to a large probable error. It is therefore not in disagreement with the recent value found by Damon and Edwards. This value, $19 \pm 3\%$, was found directly by counting quanta with a NaI scintillation counter, and is probably the best determination to date.

L Level Fluorescence Yield. It has already been
mentioned (page 14) that there is a huge uncertainty about the value of this quantity. Ross et al. (51) have examined the relevant data; the quoted values range from 21% to 56%.

By combining Damon's result for the intensity of $L$ X-rays with that of Wu for the frequency of occurrence of $L$-holes, one arrives at a value of $30 \pm 5\%$ and Damon and Edwards (49) have pointed out that this value brings Wu's estimation of the intensity of the emitted $\gamma$-ray into line with those of all other workers. Rose has remarked that the observations of Wu and of Hughes on the ratio of $L$ X-rays to emitted $\gamma$-rays lead to values of $25 \pm 4\%$ and $38 \pm 3\%$ respectively, when combined with the theoretical internal conversion coefficient (58); and that the latter, taken with Wu's determination of the frequency of occurrence of $L$-holes, leads to a value of $3.5\%$ for the $\gamma$-ray intensity. All of these facts are concordant with Damon's proposed value of the fluorescence yield, or, in other words, the observations of Damon, Wu, Rose (58), and Bramson are mutually consistent as the following résumé shows. (The $N$'s are intensities; the suffixes are self-explanatory.)

**Primary Data.**

(1) Wu et al.:

\[
\frac{N_{Le}}{N_{\gamma}} = 64 \pm 4% \\
\frac{N_{LX}}{N_{\gamma}} = 4.4 \pm 0.7\%
\]
21.

(2) Damon and Edwards: $N_{LX} = 19 \pm 3\%$

\[ N_\gamma = 3.8 \pm 0.6\% \]

(3) Branson: $N_\gamma = 3.1 \pm 1.2\%$

(4) Rose et al. (theoretical calculation of internal conversion coefficient):

\[ \frac{N_{Le}}{N_\gamma} = 17.77 \pm 0.35. \]

Derived Quantities.

(Compared with experimentally observed quantities.)

From (1) and (2)

L fluorescence yield $= \frac{N_{LX}}{N_{Le}} = \frac{19}{64} = 30 \pm 5\%.$

From (2)

\[ \frac{N_{LX}}{N_\gamma} = \frac{19}{3.8} = 5.0 \pm 1.2. \]

of. Wu 4.4 \pm 0.7.

From (1) and (2)

\[ N_\gamma = \frac{N_\gamma}{N_{LX}} \cdot N_{LX} = \frac{19}{4.4} = 4.3 \pm 1.0\%. \]

of. Damon and Edwards

\[ N_\gamma = 3.8 \pm 0.6\% \]

and Branson

\[ N_\gamma = 3.1 \pm 1.2\%. \]

From (1) and (4)

\[ N_\gamma = \frac{N_x}{N_{Le}} \cdot N_{Le} = \frac{64}{17.77} = 3.6 \pm 0.25\%. \]

of. Damon and Edwards

\[ N_\gamma = 3.8 \pm 0.6\% \]

and Branson $N_\gamma = 3.1 \pm 1.2\%.$
Yields of the L sub-shells. In the cases where the γ-ray is internally converted, the ionised daughter atom (Bi²¹⁰) de-excites by the transfer of the ionisation to successively higher levels. The energy released by each transfer, equal to the difference in the binding energies of the shells between which the transfer takes place, may be emitted as an X-ray quantum or may result in the ejection of an electron from shells for which ionisation is energetically possible. The latter possibility, which may with certain reservations (61), be regarded as the internal conversion of an X-ray, is called the Auger effect, after its discoverer. It is possible for Auger transitions to occur in cases where the corresponding X-ray transition is forbidden by the selection rules, and in cases where the corresponding X-ray transition is allowed but never observed. The RaD L\textsubscript{I}→L\textsubscript{II},\textsubscript{III} Coster-Kronig transitions (see below) are an instance of the latter.

As shown in Table V (page 17), the frequencies of ionisation of the L\textsubscript{I}, L\textsubscript{II} and L\textsubscript{III} shells during the disintegration of RaD are roughly in the ratio 100:8:1. The ratio of the intensities of the X-rays originating in these levels is, however, about 100:83:230, showing that an appreciable transfer of ionisation from the L\textsubscript{I} to the L\textsubscript{II},\textsubscript{III} sub-shells takes place. Such a radiationless transfer between the sub-shells of one shell is called a Coster-Kronig transition (62).
Ross, Cochran, Hughes and Feather have made a detailed examination of the X-ray, Auger-electron and Coster-Kronig yields of the $L_I$, $L_{II}$ and $L_{III}$ sub-shells. They summarise the results of Frilley et al. (63), Ivan (56) and Cochran (64) on the relative intensities of the lines of the L X-ray spectrum, and the results of Kobayashi (38) and Bashilov (26) on the intensities of the Auger electron spectrum. On the assumption that the total fluorescence yield of the L shell is $40 \pm 2\%$, they come to the following conclusions:

$$
\begin{align*}
\omega_1 &= 0.11 \pm 0.01 \\
a_1 &= 0.105 \pm 0.025 \\
f_{12} &= 0.19 \pm 0.05 \\
\omega_2 &= 0.32 \pm 0.04 \\
a_2 &= 0.64 \pm 0.14 \\
f_{13} &= 0.59 \pm 0.05 \\
\omega_3 &= 0.39 \pm 0.05 \\
a_3 &= 0.61 \pm 0.05 \\
f_{23} &= 0.04 \pm 0.14 
\end{align*}
$$

$\omega$, $a$ and $f$ are the fluorescence, Auger and Coster-Kronig yields respectively. The suffixes refer to the sub-shell(s) involved.

These results, and the results of the analysis by M.A.S. Ross of the Auger electron spectrum will be referred to in a later section.

**Yield of the M Shell.** Although the M shell undergoes primary ionisation in only about $20\%$ of disintegrations, the number of M-holes per disintegration is greatly increased by the Coster-Kronig, Auger and X-ray transitions of the L shell. The total is about 120 M-holes per 100 disintegrations (65). The fluorescence yield of the M-shell is $3.7 \pm 0.7\%$ according to A.A. Jaffe (65).
Chapter II.

Preparation of the Source.

The disintegration of RaD was studied by impregnating an Ilford G 5 nuclear emulsion with a solution of a salt of RaD, and then studying the tracks produced in the developed plate under a microscope.

Following the experience of previous workers who have used the impregnated emulsion method, the source was prepared in the form of lead (RaD) citrate. Yagoda (66) and Jarvie (67) found that the heavy elements, in general, tend to form chemical bonds with the outer layers of the emulsion. Zajac and Miller (68) pointed out that this difficulty is avoided by using the citrate.

The sources were prepared in the following way, in the first instances by Dr. N. Miller of this department, and later by the author, following Dr. Miller's instructions:

The primary source was lead (RaD) nitrate from which the daughter products RaE (Bi²¹⁰) and RaF (Po²¹⁰) had to be removed. Forty drops of the primary solution were put in a centrifuge tube, and a little bismuth nitrate and lead nitrate solutions added as carriers. Some concentrated sulphuric acid was added, so that after mixing well, the (lead + RaD) sulphate came out of solution. The mixture was centrifuged and the liquid poured off. A little sodium
carbonate solution was added to the precipitate to convert it to the carbonate, and the mixture centrifuged. The precipitate was washed with distilled water several times, and citric acid added to the final precipitate to give the citrate. Finally, using universal indicator, the solution was neutralized by the addition of caustic soda solution. The work of Picciotto (69) has shown that the use of neutral solutions is desirable.

**Efficiency of the Separation Process.**

After observations on the RaD tracks in the nuclear emulsion had been completed it became apparent that it would be useful (see page 93) to know with what efficiency the above chemical process removed RaE from the primary source.

The experimental determination of this efficiency will now be described so that the result may be introduced later at the appropriate place.

**Theory of the Experiment.**

The experiment consisted essentially of observing the growth of RaE (half-life about 5 days) in the prepared sources. This was done by counting (with a Geiger counter) the rate of emission of RaE β-particles (end-point energy 1.16 MeV) since this rate is proportional to the amount of RaE present in the source.

If a source containing \( N_0 \) atoms is pure RaD at a
26.

At time \( t = 0 \), the number of RaE atoms, \( N_T \), present at time \( t = T \) is

\[
N_T = \frac{N_0 \lambda_1}{\lambda_2 - \lambda_1} (e^{-\lambda_1 T} - e^{-\lambda_2 T})
\]

where \( \lambda_1, \lambda_2 \) are the disintegration constants of RaD and RaE respectively. \( \lambda_2 \) is so very large compared with \( \lambda_1 \) (factor of 22 years/5 days \( \approx 1,600 \)) that

\[
N_T = \frac{N_0 \lambda_1}{\lambda_2 - \lambda_1} (1 - e^{-\lambda_2 T})
\]

This curve is shown in fig. 2.

If at the time of separation, there is some RaE present, the source is, in effect, already aged by an amount, say, \( t = t_s \) (fig. 2). The age of the source at a later time can be determined by measuring the RaE activities, \( A \), at two different times, say \( t = T \), and \( t = T + k \). Then

\[
\frac{A_T}{A_{T+k}} = \frac{N_T}{N_{T+k}} = \frac{1 - e^{-\lambda_1 T}}{l - e^{-\lambda_2 (T+k)}}
\]

(the suffix 2 may now be dropped from \( \lambda_2 \))

\[
e = \frac{N_{T+k} - N_T}{N_{T+k} - N_T e^{-\lambda k}}
\]

and so \( T \), the age of the source at the time of the first observation, can be found. But since the lapse of time \( (T - t_s) \) between the time of separation and the time of the first observation is known, the "age" of the source at separation, \( t_s \), can be deduced. The
efficiency with which RaE has been removed from the equilibrium Ra(D + E) solution is then \[ \frac{N_\infty - N_{ts}}{N_\infty} \]
where \( N_\infty \) is the equilibrium concentration of RaE. I.e the efficiency of separation is \( \eta = e^{-\lambda t_s} \).

**Experimental Arrangement.**

The main radiations from a RaD source with its daughter products are the 47 keV RaD \( \gamma \)-ray, the RaD X-rays of about 12 keV, the RaD conversion electrons of about 30 and 40 keV, the RaE \( \beta \)-particle of maximum energy 1.16 MeV, and the RaF \( \alpha \)-particle of 5.3 MeV. It is required to count only the RaE \( \beta \)-particles.

The range of the RaD conversion electrons in air is less than 4 cm. and so they were shielded off by placing the counter window 7 cm. from the source. The same air path stops the Po\(^{210} \) \( \alpha \)-rays (range 3.9 cm.) but in any case these never attained appreciable intensity over the period of the experiments. The RaD X-rays and \( \gamma \)-rays could not be shielded off but their counting rate (which was constant for any one source over the period of the experiment) was assessed by placing the apparatus between the poles of an electromagnet and so preventing any electrons from reaching the counter window. It was found, by means of a fluxmeter, that with a coil current of 6 amperes, the electromagnet supplied a field of 4,300 gauss over the region of the air space between the source and the
counter window. Since the fastest RaE $\beta$-particles have an $H_p$ value of about 5,300 ($\rho = 1.1$ cm.), this field value was sufficient for the purpose.

The experimental arrangement is shown in fig. 3. The wooden holder for the source and counter was designed to ensure that the geometry remained constant for all sources and for all times of observation. Its dimensions, apart from the 7 cm. air space, were chosen so that it could fit in a reproducible way between the poles of the electromagnet, with the air space directly between the poles. The wooden spar AB was designed to hold a Co$^{60}$ source (used as a standard) at a convenient and reproducible distance from the counter. The Co$^{60}$ source was contained in a closed brass cylinder of about 4 mm. diameter which could be pushed into the close-fitting hole C. A brass locating collar, fixed permanently to the cylinder, ensured that the Co$^{60}$ source could always be replaced in an accurately reproducible position.

The sources were in the form of RaD sulphate prepared as described above, and evaporated in cylindrical brass cups. Some distrene cement, poured in a thin film over the sources and allowed to dry, ensured that the sources remained in a fixed position and presented no health hazard. The brass cups fitted fairly closely into a recess in the wooden holder.

The Geiger-Muller counter was type G.E.C. ERM2
FIGURE 3.

Geiger counter
air space.

magnet pole
magnet pole
metal plate
wooden support
for counter

wooden base
and source holder

recess for source

10 cm.
with a mica end-window of 1.9 mg./cm.$^2$. It was chosen from several since its plateau was satisfactory (0.025% per volt at 1,600 volts). The probe unit accepted the negative pulses from the counter and delivered amplified positive pulses to the scalar.

**Experiment.**

Dead-time. It was first necessary to find the "dead-time" of the apparatus. Each component, (that is, the counter, the probe unit and the scaler) has its own dead-time, the time the component takes to regain its original receptive condition after it has been triggered by a pulse (or by an incoming radiation, in the case of the counter). The effective dead-time of the system is that of the component which has the greatest dead-time. It is clear that the "loss" of counts through this paralysis of the counting system becomes more serious as the counting rate is raised.

The formula connecting the true number of counts $N_o$ with the observed number $N_c$ is

$$N_c = \frac{N_o}{1 + N_o \tau}$$

where $\tau$ is the dead or paralysis time. The dead-time was measured in three ways, using two sources of Co$^{60}$ (1.17, 1.33 MeV $\gamma$-rays). In each case, one source was placed in such a position as to give a convenient counting rate; the other was then brought up and placed so that, roughly, the counting rate doubled;
then the first source was removed and the counting rate produced by the second source alone was observed. Let these three counting rates be called \( n_1, n_2 \) and \( n_3 \). There are in general lost counts in all three rates. The three methods were:

(a) \( n_1, n_2 \) and \( n_3 \) were observed at very low counting rates and it was verified that there were no lost counts in \( n_2 \); i.e. \( n_1 + n_3 \) equalled \( n_2 \) within the statistical error. A second series was then observed \( n'_1, n'_2 \) and \( n'_3 \) so chosen that \( n'_1 \) and \( n'_3 \) were about equal to \( n_2 \). Then, since there was no loss in \( n'_1 \) and \( n'_3 \), the loss in \( n'_2 \), namely \( n'_1 + n'_3 - n'_2 \), could be found. Proceeding to high counting rates in this step-by-step fashion, it was possible to find the lost counts appropriate to some high counting rate and then to apply the formula

\[
\mathcal{C} = \frac{N_0 - N_0}{N_0 N_0}.
\]

It was found, however, that the statistical errors accumulated at each step in the process, so that the final result was subject to a large error: 

\[
\mathcal{C} = (2.4 \pm 0.5) \times 10^{-4} \text{ sec.}
\]

(b) \( n_1, n_2 \) and \( n_3 \) were observed for a high counting rate \( (n_3 \sim 16,000/\text{min.}) \). The dead-time was then found by a method of successive approximations. The first approximation is:

\[
\mathcal{C}_1 = \frac{(n_1 + n_3) - n_2}{(n_1 + n_3)n_2} = 1.16 \times 10^{-4} \text{ sec.}
\]
(This assumes tentatively that there are no lost counts at counting rates $n_1$ and $n_3$).

This first approximation to $\mathcal{C}$ was then used to find the first approximation to the true counting rates of $n_1$ and $n_3$:

$$n_1' = \frac{n_1}{1 - n_1 \mathcal{C}_1}, \quad n_2' = \frac{n_2}{1 - n_2 \mathcal{C}_1}.$$ 

Then the second approximation to $\mathcal{C}$ was obtained:

$$\mathcal{C}_2 = \frac{(n_1' + n_3') - n_2}{(n_1' + n_3') n_2}.$$ 

This cycle was repeated until the successive values for $\mathcal{C}$ became nearly equal. The final result was

$$\mathcal{C} = (2.34 \pm 0.20) \times 10^{-4} \text{ sec}.$$ 

(o) A formula due to Beerø (70) was finally used:

$$\Delta = \frac{\Delta n_2}{2n_1 n_3} + \frac{1}{8(n_2 + 2B)} \left( \frac{\Delta n_2}{n_1 n_3} \right)^2$$

where $\Delta = n_1 + n_3 - n_2$

and $B = \text{background counting rate}$.

This gave an answer similar to that of method (b) -

$$\mathcal{C} = (2.30 \pm 0.20) \times 10^{-4} \text{ sec}.$$ 

During the course of observations, many opportunities arose of testing and confirming the reliability of this value. It corresponds roughly to the nominal paralysis time of the probe unit (200 $\mu$s) and so can be assumed to be independent of the counting rate (this is not in general true of the counter dead-time).

Observations on Six Sources. Six sources were prepared
by the process described above. Since, in the opinion of Dr. Miller, the amount of bismuth carrier was the only factor likely to affect the efficiency of separation, the amount added to sources 3 and 4 differed from the amount added to sources 5 and 6 by a factor of 10 (5 and 50 drops of bismuth nitrate solution respectively. This range certainly brackets the amount added in the preparation of the source which was used in the impregnation of the nuclear emulsion plate).

The observations made on each source were as follows. The time was noted at the time of separation that is, when the concentrated sulphuric acid was added to the \((RaD + RaE + Pb + Bi)\) nitrate solution. The counting rate of RaE \(\beta\)-particles from the mounted source was then observed (as soon as possible after separation) at a noted time. These observations were repeated from three to ten times, at intervals, over the ensuing five days.

In detail, the counting involved the following operations:

The scalar (1009A) was checked by means of the internal testing system. Internally generated pulses with a recurrence rate of about 3 per second, and the voltage peaks of the mains input (about 50 per second) could be counted. Small adjustments and replacements of components were made until the counting was satisfactory. The mains frequency test was also used to
find the accuracy with which a counting interval of (nominally) 1 minute could be reproduced using a stop watch. This accuracy depends on the constancy (not, of course, the absolute accuracy) of the watch and the constancy of the reaction times between the eye and the two hands. It was found, over ten observations, that the standard error was about 1/20 sec, in 1 nominal minute, that is, this error was involved in the starting and stopping of the watch and in the (ideally) simultaneous operation of the scalar switch. The background counting rate of the counter was found at frequent intervals and was usually 16-20 counts per minute. On starting each morning it was found that the background rate was higher than this, and that it could be brought down by counting a heavy flux of γ-rays for a few seconds.

The RaD γ-ray and X-ray counting rate for each source was found (with two or three repetitions at intervals) by putting the apparatus in the magnetic field. It ranged from 30-60 counts per minute and was constant in time for any one source. By using an external source of γ-rays it was verified that the magnetic field did not in itself affect the sensitivity and counting rate of the counter. Except when making these observations, the apparatus was removed from the electromagnet since it was not certain that when the activating current was switched off, the residual field
would always return to a constant strength.

The observation of the counting rate of RaE $\beta$-particles from each source involved three counts:

(a) the source was inserted, the E.H.T. ($\sim 1,600$ volts) was switched on and a count taken over 5 minutes; (b) leaving the source in, and the E.H.T. on, the Co$^{60}$ standard source was placed in position (as described before) and a count taken over 2 minutes; (c) the E.H.T. was switched off, the source was removed, the E.H.T. switched on, and a count taken of the Co$^{60}$ standard alone over 5 minutes. The purpose of count (c) was to monitor the efficiency of the counter; this guards against long-term instability of the E.H.T. supply and gradual deterioration of the counter. Ideally the monitoring should be done under the same conditions as the actual count, but the fact that, for safety, the E.H.T. had to be switched off to insert or remove a source and then switched on again, introduced a (barely) possible cause of variation in the value of the E.H.T. Count (b) was intended to provide a check against this, and it also, incidentally, furnished a repeated check on the reliability of the observed value of the paralysis time, since for true counting rates, $(a) + (c) = (b)$. The background counting rate was usually taken before and after each run of observations.

Each count was corrected for the loss of counts using the formula $N_0 = \frac{N_0}{1 - \frac{N_0}{C}}$ and the contribution
due to background and to X-and γ-rays was subtracted. The resulting "true" count of RaE β-particles was corrected according to the departure of its monitor count from a convenient "normal".

For any one source, any two of the true, normalised counting rates, \( N_T \) and \( N_T + k \), separated in time by an interval \( k \), could be inserted in formula (1) (page 26) and hence the efficiency of separation found.

In general, for each source, several such calculations were made yielding the results shown in Table VI. Not all the results in any one column are derived from completely independent data.

**Table VI.**

*Results of Experiment on Efficiency of Source Separation.*

<table>
<thead>
<tr>
<th>Source</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Efficiency of separation</td>
<td>93.6</td>
<td>99.2</td>
<td>97.7</td>
<td>98.35</td>
<td>95.1</td>
<td>99.2</td>
</tr>
<tr>
<td>%</td>
<td>93.5</td>
<td>99.2</td>
<td>97.8</td>
<td>98.35</td>
<td>95.4</td>
<td>99.2</td>
</tr>
<tr>
<td>Error in each</td>
<td>± 2.0</td>
<td>± 0.2</td>
<td>± 1.0</td>
<td>± 1.0</td>
<td>± 1.3</td>
<td>± 0.3</td>
</tr>
<tr>
<td>Average</td>
<td>94.1</td>
<td>99.2</td>
<td>97.8</td>
<td>98.3</td>
<td>95.5</td>
<td>99.2</td>
</tr>
</tbody>
</table>

The error for source 1 is large since, because of a
counter break-down, it was impossible to obtain a count soon after separation.

It is concluded that the efficiency with which RaE was removed from the primary equilibrium source was 97.5 ± 2.0%. Although the error, which is in fact the standard error of the six averages, does not embrace the low value for source 1, it is considered adequate in view of the large error in this lot value. Comparison of the results 3 and 4 with the results 5 and 6 shows that there is no apparent correlation (within the range of quantities used) between the amount of bismuth carrier and the efficiency of removal of RaE.
Chapter III.

The Nuclear Emulsion Technique.

The so-called "nuclear" emulsions are photographic emulsions which have been specially developed for the study of ionising particles - cosmic rays; the particles produced by nuclear fission, nuclear reactions, and nuclear disintegrations.

The action of single $\alpha$-particles on a photographic plate was first observed by Kinoshita at Manchester in 1910. In 1927, Blau and Wambacher, following a suggestion by Myssowsky and Tschishow prepared thick emulsions so that long tracks of fast protons could be observed. Interest in the technique was stimulated when Powell in 1939-40 showed by a series of experiments that the method was capable of giving accurate results for particle energies. After the war, efforts to produce emulsions of greater sensitivity (notably by Ilford in collaboration with Powell) culminated in the production in 1948 of the Kodak NT 4, and later, the Ilford G 5 emulsions. These emulsions combined a high silver halide content with small grain size and high sensitivity so that for the first time it became possible to follow tracks at minimum ionisation energies. Since for electrons this energy is only 1 MeV, these emulsions were the first to be fully electron-sensitive, and made possible the study of $\beta$-decay and $\gamma$-ray conversion with nuclear emulsions.
Zajac and Ross (71) in 1949 calibrated the FT 4 emulsion by measuring the ranges and the grain counts of tracks produced by electrons of known energy. The calibration covered the range 30 to 250 keV and the observed ranges agreed closely with the theoretical formula of Bethe. The grain counts and ranges were found to be given by expressions of the type

\[ g = a E^k \quad r = a' E^{k'} \]

where \( E \) is the electron energy and \( a, a', k, k' \) are constants. The standard deviation of the range and grain count distribution for a monoenergetic group of electrons was about 20% of the mean value throughout the energy interval 30-250 keV. Under the supervision of Dr. Ross, work by Zajac (72), Jarvis (73), Bisgard (74) and Bayman (75) on the conversion electrons of \( \alpha - \gamma \) transitions proved the validity and usefulness of the method. In particular the work of Bisgard had shown that grain-counting could give results for low energy electrons whose ranges were too short to be measured with accuracy.

Some time before the author began work on RaD, Spurgin, working with Dr. Ross, had shown that the nuclear emulsion method could be used for purely electronic events, that is, in cases where there was no easily seen \( \alpha \)-particle to identify the site of the disintegration. Working with the \( \beta, \gamma \)-emitter \( {\text{MeTh}}_2 \), he had observed events in which a \( \beta \)-particle track
started from the same point as a γ-ray conversion electron. This indicated that the problems of the RaD→E→F γ-decay chain might be approached in the same way with some prospect of success.

Impregnation and Development of Plates.

The technique of impregnation and processing of the plates was that used successfully by previous workers in the nuclear emulsion laboratory. For each impregnation a solution of RaD citrate was prepared (by Dr. N. Miller) as described in Chapter II. Since the absolute disintegration rate of the solution was only roughly known, and since in any case the most suitable density of events in the developed plates could only be found by trial and error, the impregnating solution was made up in a series of concentrations, each one two times more dilute than the preceding. The plates (both Kodak NT 4 and Ilford G 5, 200 μ thick, were used in different impregnations) were cut into 1 inch square sections, marked with identifying marks, and soaked for 30 minutes in the solutions. They were then dried, first with filter paper and then by lying on a hot plate for an hour or so. After lying for a time to ensure that they were dry, they were put in a light-tight box and stored in a lead "castle". Since the range of an electron in the emulsion, and the number of grains it produces in the developed plate, depend on
the moisture content of the plate, this was kept constant by including in the box some potassium carbonate. This kept the relative humidity of the air at 44%.

After periods ranging from 1 to 4 weeks, the plates were developed, following essentially the method of Dilworth, Occhialini and Payne. The developer used was the following.

**Solution A.** (prepared immediately before use).
- Crystalline sodium sulphite 25 gm.
- "Elon" or "Metol" 3 gm.
- Distilled water to 1,000 cc.

**Solution B.** (prepared once a fortnight).
- Anhydrous sodium carbonate 50 gm.
- Anhydrous sodium bicarbonate 50 gm.
- Distilled water to 1,000 cc.

Solutions A and B mixed in equal volumes.

The developer was put in large test-tubes, and cooled in ice for about 30 minutes. The sections of plate were put in, one in each test-tube, and soaked for 30 minutes. The test-tubes were then transferred to a "warm" bath (22°C) and left there for 20-30 minutes, so that development took place. The developed plates were put in a "stop-bath" (5 gm. sodium bisulphite in 100 cc. of water) for quarter of an hour. Usually, the plates at this stage had an opaque layer of silver on their surfaces, and this was rubbed off with the finger before the plates were put into the
41.

fixer (300 gm. of hypo in a litre of water). After about 24 hours the fixing was complete, and the fixing bath was diluted, very gently at first, with a stream of tap water. The washing of the plates was judged to be sufficient when no residual hypo could be detected in a sample of the wash-water by the potassium permanganate test (a drop of dilute KMnO₄ solution disappears when dropped into water containing a trace of hypo).

The plates were taken out, left to dry and then mounted with Canada balsam on 3" x 1" glass slides for examination.

It was found, in agreement with the experience of previous workers, that the behaviour of Kodak NT 4 emulsions under the above treatment was not satisfactory. The emulsions tended to peel off either during impregnation or during development or after drying. Of the 65 plates, most had an unexplained dense layer of fog grains somewhat above the glass-emulsion interface. One batch, however, was completely satisfactory, clear and well-developed. This batch had been left (inadvertently) for about 3 hours on the hot plate after impregnation and perhaps this played some part in producing the satisfactory result. The work to be described was done almost entirely on one plate from this batch, chosen for its convenient density of events.
Description of the Events.

The disintegration of RaD (half-value period about 22 years) is followed in many cases by the disintegration of RaE (half-value period about 5.0 days). Fig. 4 shows the growth of RaE (curve A) and RaF (curve B) in a pure source of RaD. It will be seen that, ignoring the small RaE contamination in the impregnating solution, curve A may be interpreted as the density (in the developed plate) of events associated with RaD disintegration, plotted as a function of the time between impregnation and development (exposure time). Similarly, curve B gives the density of events for which the RaD disintegration is followed by the disintegration of RaE, leaving RaF. Only the latter events were studied, since the long RaE $\beta$-particle tracks made it possible to locate and identify with certainty the short RaD tracks with which they were associated.

The figure shows that for the exposure time of the observed plate, 19 days, the ratio of RaD,E to RaD disintegration was 2.2. To make evident the nature of the RaD tracks, it is convenient to classify the disintegrations as in Table VII, using the results discussed in Chapter I.
FIGURE 4

No. of atoms (units $N_0 \times 10^{-4}$)

Exposure time (days)

Curve B (RaF)

Curve A (RaE)
Table VII.

<table>
<thead>
<tr>
<th>Particles</th>
<th>Frequency of occurrence</th>
<th>Energies keV</th>
<th>Expected total grain-count (after Zajac and Ross)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{\beta} \beta_{47} + \gamma$</td>
<td>3.8%</td>
<td>15, 47</td>
<td>0 - 4</td>
</tr>
<tr>
<td>$^{\beta} \beta_{47} + e_L + X_L$</td>
<td>19%</td>
<td>15, 30, 12</td>
<td>10 - 14</td>
</tr>
<tr>
<td>$^{\beta} \beta_{47} + e_L + A_L$</td>
<td>45%</td>
<td>15, 30, 8</td>
<td>12 - 17</td>
</tr>
<tr>
<td>$^{\beta} \beta_{47} + e_{MNO...}$</td>
<td>21%</td>
<td>15, 43</td>
<td>16 - 19</td>
</tr>
<tr>
<td>others?</td>
<td>11%</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^{\beta} \beta_{47}$ denotes $\beta$-particles of the $\beta$ transition to the 47 keV excited state of RaE. $\gamma$-rays, X-rays, conversion electrons and Auger electrons are denoted by $\gamma$, $X$, $e$ and $A$ respectively.

Taking into account the spread of grain-counts about the expected mean value, it is seen that most of the RaE events have 8-22 grains; the expected track ranges are 5-12 microns. Such tracks have a high grain density and are highly scattered so that some RaE events looked like small, knobby, opaque, round masses of silver. Some tracks, on the other hand, showed a definite path with increasing grain density along the direction of travel. But the majority were clusters of grains more or less elongated along the supposed trajectory.

The RaE $\beta$-particle tracks started near the RaE cluster, comparatively straight and lightly grained.
(the average grain-spacing near the origin of the RaE tracks observed was about 2μ). Many of these left the emulsion at the surface or the glass-emulsion interface, but some could be followed 100-600μ to their ends, the grain density and amount of scattering increasing along the trajectory.

Photographs of some events are shown in fig. 5. Fig. 6 shows drawings of twelve typical events copied from notebooks.

**Background Tracks.**

Besides the events of interest, there was a considerable number of cosmic-ray background tracks in the plate. (Fog grains occurred too but in negligible numbers.) These consisted of (1) the straight tracks of fast electrons, protons, mesons etc. which could not be confused with RaE β tracks, and (2) curved tracks produced by electron showers. Comparison of the tracks in the observed plate with those in a "control plate" (whose history was identical with the observed plate except that it was not impregnated) showed that the long tracks in the observed plate were 45% RaE tracks, 22% curved cosmic-ray tracks of the same appearance as RaE β tracks but not starting in the emulsion, and 33% straight cosmic-ray tracks going directly from surface to glass.

It will be of interest, later, to know with what
FIGURE 5

12 grains

3 grains

12 grains

8 grains
FIGURE 6(a)
probability a curved cosmic ray track might be mistaken for a RaE $\beta$ track starting in the emulsion. In a survey of 200 such tracks in the control plate, 3 tracks were found which could conceivably have been judged to start in the emulsion. Since these tracks, if they had occurred in the plate under observation, would not in fact have been recorded as RaE $\beta$ tracks (they looked like what in fact they must have been—tracks which had got broken up and lost in passing through an insensitive or distorted part of the emulsion) it is certainly safe to say that no more than 3 in 200 curved cosmic ray tracks would be mistaken for RaE $\beta$ tracks starting in the emulsion. This result will be used later. (Page 94)

The Measurement of RaD Tracks.

Grain Counting.

The group of grains attributable to RaD electrons usually formed a single opaque cluster although sometimes there were two or, rarely, three separate clusters. Only very rarely was it possible to see the greater part of the boundary of a single grain. The grain counting had therefore to be done by observing the irregularities along the boundary of the cluster (often the cluster was elongated, one or two grains wide), and, in the case of thicker clusters, by fitting imaginary standard grains inside the pattern of the
boundary grains. The conceptual image of the standard grain was continually being checked against the single grains seen along the RaE $\beta$ tracks, and against the single grains which could often be seen jutting out of the RaD cluster. It will be seen later that after some experience of grain counting the process led to remarkably reproducible grain-count histograms, and that, over a region of the spectrum about which a great deal is known, the grain-count histogram tallied very well with the expected shape.

Range Measurement.

It is possible to measure the three-dimensional ranges of tracks by means of a calibrated eyepiece graticule (for projections of ranges on the horizontal plane) and by a calibrated fine focusing adjustment. In the opinion of the author, however, it is of little value to attempt to measure ranges of electron tracks which do not have spaced grains along an appreciable portion of their length. With fairly long tracks, the trajectory is well defined and the total range can be found by summing the lengths of fairly straight segments of track. Since each segment has a perfectly visible grain at each end the fine focusing can be done with some accuracy. With short tracks attributable to electrons of 0-40 keV, on the other hand, the track is usually 2 or 3, and is frequently 4 or 5 grains
thick. It can only be said that the true trajectory lies somewhere in the region occupied by the cluster, that it will in general be highly scattered, and that an unknown number of grains are actually 1 or 2 grain branch tracks which are expected to occur in detectable numbers at these low energies. Thus the apparent range, even if it could be measured accurately, would always be less than the length of true trajectory. But it is difficult to measure even the apparent range except for tracks which are inclined at small angles to the horizontal. For the others, the terminal grain at the bottom end of the track is always obscured, if not by the actual superposition of higher grains, then by their out-of-focus diffraction patterns (which are much larger than the grains themselves) so that it is impossible to focus accurately on the end grain.

Of 40 RaD clusters observed, the majority had apparent ranges between 2 and 6 $\mu$. The effective upper limit was about 9 $\mu$ with one long track at 13 $\mu$. These values are very much lower than those predicted by the Zajac-Rose calibration which gives $7 \pm 1.5 \mu$ for the range of the main group of L conversion electrons and $13 \pm 3.5 \mu$ for the MN conversion group. That this discrepancy is explicable as the discrepancy between the true trajectory length as predicted by the calibration and the apparent range as measured in the emulsion is shown by the following argument.
(a) For 30 keV electrons in NT 4 emulsion, Zajac found that the mean range was 7.0 μ and the mean grain-count 11.0.

(b) In Ilford G 5 emulsions the mean diameter of developed grain is 0.6-0.7 μ.

(c) As will be shown later, the grain counts of the author for electrons of a given energy in G 5 emulsion was much the same as those of Zajac in NT 4 emulsion.

It follows from these facts that if a 30 keV track in G 5 emulsion, with 11 grains, had a measurable range of 7.0 μ, the grains would have to be arranged end-to-end in "single file" along the trajectory. In fact, such tracks are virtually never observed so that a 7 μ, 11 grain track in G 5 emulsion would be a freak. The evidence shows that the trajectory of a 30 keV electron is highly tortuous and that the apparent "range" of the cluster is a factor of 1.5-3 smaller than the true trajectory length.

In view of the considerations set out in this section, the observations on RaD events were confined solely to grain-counting.

The Structure of the RaD Cluster.

Combining the range-energy and the grain-energy relations of Zajac and Ross, it is found that the grain density appropriate to an electron travelling with energy E is given by
\[ \frac{dg}{dr} = 4.62 \times 10^{-0.4}. \]

The grain-spacing \( \frac{dr}{dg} \) for various energies is then

<table>
<thead>
<tr>
<th>Energy (keV):-</th>
<th>5</th>
<th>10</th>
<th>20</th>
<th>30</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \frac{dr}{dg} (\mu) ):-</td>
<td>0.41</td>
<td>0.54</td>
<td>0.72</td>
<td>0.85</td>
</tr>
</tbody>
</table>

Considering that the mean diameter of a developed grain is \( \bar{d} = 0.6\mu \), and that the average path length through a grain is \( \frac{2\pi}{3} \), it is apparent that right down to 5 keV (about 2 grains), the grains of a cluster do not form a more or less solid mass but remain, in general, separate. They appear to form an opaque mass in the microscope field partly because spaces between the upper grains are obscured by the presence of grains underneath, and partly perhaps because the diffraction image of such a close packed three-dimensional cluster of grains fails to reproduce the pattern of individual grains. This looseness of the grain cluster must explain why grain-counting can be done at low energies without great loss of resolving power. For, if the grains were touching each other, a number of grains would be totally obscured, the number varying widely according to the orientation of the track, so this is true only if the trajectory of a track may pass anywhere through a developed grain. This in turn depends on the precise mechanism by which an activated grain (diameter \( \sim 0.2\mu \)) becomes a developed grain.
that the "line width" for monoenergetic electrons due to the random "straggle" of grains brought up would be increased by this cause.

The number of "branch" grains is given by the formula

\[ B = \frac{N}{E} \left( \frac{1}{W} - \frac{2}{E} \right) \text{ branches/cm.} \]

where \( N \) is the number of electrons per cc. and \( W \), the threshold energy for producing a grain, is taken to be about 2.5 keV. The result is quite small, 0.2 to 0.1 branch per \( \mu \) over the 1-30 keV range.

The implication is that the true trajectory passes through nearly every developed grain, which further implies that the path is very highly tortuous. This is confirmed by a calculation using the Rutherford scattering formula (76). Making the rough assumption that the effective nuclear charge is the mean nuclear charge of the constituents of the emulsion (\( \sim 15 \)), it is found that for a 25 keV electron there is a 60% chance of being singly scattered through an angle greater than 30° in a path length of 1 micron.
Chapter IV.

When observations were started on the events in the nuclear emulsion plate, there was no clear idea of what line the investigation might take. It was known that knowledge of the RaD disintegration was incomplete, and that "electron-star" events in the emulsion could be found and measured - it remained to be seen what kind of information the method could be made to yield. It was always kept in mind, however, that the outstanding question was whether or not the transition was the only mode of decay. It was realised that, in the absence of any cascade de-excitation of the 47 keV excited state, the only possible alternative was a transition to the ground state of RaE or to very low-lying excited level (or levels) of RaE.

Most of the microscope work described in the thesis was done with a 1/12" objective, numerical aperture (with oil-immersion) 1.28. The eyepiece had a magnifying power of 10.

First Group of Observations: 91 Events.

Following the method of previous workers the events were found by scanning the plate systematically, searching each field of view (about 120$\mu$ diameter) several times from top to bottom of the emulsion. The site or origin of each event was found by producing the RaE $\beta$-particle track to intersect the RaD cluster.
Very often the cluster had a structure suggestive of multiple electron emission; for instance it might have a gap, or a narrow waist, or a sharp bend, or a sharply projecting spur at or near the supposed site of the event. Or, in the case of an elongated cluster, the origin might seem to be in the middle rather than at one or other of the ends. Examples of such tracks are shown in fig. 7. On the tentative assumption that the apparent component tracks emanating from the origin were indeed produced by separate electron emissions (e.g. L conversion electron + Auger electron + primary β-particle), an estimation was made of the grain-count of each component of the structure.

Of 91 events observed, 33% seemed to have a single component, 60% two components and 7% three components. The following brief examination shows that the results do not fit the known facts about the disintegration.

(a) The large number of "double" events could only be accounted for by taking the most extreme of the results discussed in Chapter I. If one assumes that the L fluorescence yield is 30% (page 20) and that 19 L X-ray quanta are emitted per 100 disintegrations (page 19) the expected number of "double" events would be 44% (L conversion electron + Auger electron). The few events in which the β_{4.7} track would be seen (e_L + X_L + β_{4.7}) could not raise this number to 60%.

(b) The number of grains in the component tracks of
FIGURE 7
"double" events did not correspond to the expected pattern. An L conversion electron track is expected to have $10 \pm 3$ grains, an L Auger electron track 2, 3 or 4 grains. Even if no assumption is made about the grain-count energy relation, it is still clear that the grain counts of the two groups should be mutually exclusive. In Fig. 8, the histogram of grain counts of the longer components of "double" events is compared with the histogram for shorter components. The histograms overlap considerably; nor does either, in shape or in mean grain count, resemble the distributions expected for the groups they have been tentatively assumed to represent.

(c) Fig. 9 is the histogram of grain counts of all the individual components observed. It fails to show groups corresponding to $A_L (2 - 4$ grains), $e_L (10 \pm 3$ grains) and $e_{MN} (16 \pm 4$ grains).

It was apparent that the appearance of a Rad cluster as consisting of 2 or more components was largely governed by chance and bore little or no relation to the detailed mode of disintegration.

An attempt was made to re-classify the observed events according to a more stringent criterion of multiplicity - events were considered to be "double" only if there was a definite gap or narrow waist between the two components at the origin. This produced a result which seemed to tally fairly well with the
possible facts. The number of "double" events became 27% of all, in agreement with a fluorescence yield of 47% (see page 14) and an X-ray intensity of 25% (Stahel, page 18), results which were at that time thought to be the most reliable. The longer components of the "double" events, moreover, corresponded roughly, in grain count, with L conversion electrons (fig. 10).

The apparent success of this criterion for "double" events (the author now believes that the appearance of even these "double" events was largely due to chance - see page 63) led to the method adopted for the next group of observations.

It should be noted that of the 91 events, there were 6 whose total grain count was 6 grains or less (fig. 10). This will be recalled later.

Second Group of Observations: 139 Events.

The method of observation used for this group was intended as a preliminary attempt to reveal the presence or absence of a ground state to ground state transition. (This will be referred to as the $\beta_o$ transition, the suffix indicating the energy of the final state with respect to the ground state.) The plate was scanned systematically as before, but observations were confined to only two kinds of event. (1) Events which could with certainty be deemed to have a single
Figure 8

Figure 9

Figure 10
component. The origin clusters of such events were markedly elongated and the origin was unambiguously at one or other end of the cluster; there were no gaps or waists or sharp spurs near the origin. (2) Events which could with certainty be deemed to have two components; in the origin clusters of these events, a gap or narrow waist coincided with the origin. It was assumed, tentatively but with reasonable justification, that group (1) would be comprised of events $\beta_{47} + e_L + \chi_L$, $\beta_{47} + e_{\text{MNO}}$, and $\beta_0$, if any, and that group (2) would be comprised of events $\beta_{47} + e_L + \alpha_L$. It was assumed that the majority of $\beta_{47}$ particles would be of too low energy to form distinguishable component tracks. It will be seen later that about 90% of them have only 1 or 2 grains. It was proposed to derive from the longer components of the group (2) "double" events the grain count distribution of L conversion electrons. By subtracting this distribution from the grain count distribution of group (1) "single" events (which also contains L conversion electrons) it was hoped to "lay bare" a distribution of grain counts in the 8-16 grain region which would be accounted for only by the presence of a $\beta_0$ continuous spectrum. This method could be described and justified in greater detail, but its importance, considered in retrospect, does not warrant this.

The grain count distribution of observed "single"
events (group (1)) is shown in fig. 11 together with the corresponding "smoothed" curve. The latter was obtained by means of Spencer's 15-term smoothing formula (78). The total number of events represented is 122.

Only 17 certain "double" events (group (2)) were found. No further comment will be made on this small sample except to note that the infrequency of occurrence of these events was one of the reasons for not pursuing this method or any variant of it.

The histogram of "single" events (fig. 11), when smoothed, showed 4 main groups: a, b, c and d. Roughly, they were distributed as follows:

<table>
<thead>
<tr>
<th>Group</th>
<th>Grain count limits</th>
<th>Number in group</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>2 - 7</td>
<td>30</td>
</tr>
<tr>
<td>b</td>
<td>8 - 15</td>
<td>70</td>
</tr>
<tr>
<td>c</td>
<td>14 - 20</td>
<td></td>
</tr>
<tr>
<td>d</td>
<td>20 - 28</td>
<td>18</td>
</tr>
<tr>
<td>others</td>
<td>&gt; 28</td>
<td>4</td>
</tr>
</tbody>
</table>

Although at this stage it was impossible to say how much, if at all, the author's grain-counting deviated from that of Zajac, it seemed evident, both from the grain counts and from the relative intensities of the groups, that groups b and c consisted of L and MNO conversion electrons respectively. The Ross and Zajac calibration predicts a mean grain count of 10 and 16
FIGURE II
respectively, and the areas are expected to be about
equal (intensity of $\beta_{47} + \varepsilon_L + \chi_L$ is 19-25 per 100
disintegrations, see page 18, and that of $\beta_{47} + \varepsilon_{MNO}$
is about 20%, see page 15).

The presence of groups a and d in fairly high
intensities was at first inexplicable, and it was the
stimulus of considering these groups which led even-
tually to the method which was used for the observa-
tions on the final group of 801 events.

The Searching Procedure.

The occurrence of the apparently separate group of
high grain count events confirmed what had been sus-
pected from a consideration of the subjective process
by which events were found, namely that the method of
systematic searching of the plate introduced distortion
in the true grain count spectrum.

Previous workers who had used the field-by-field
method of searching were concerned with events in which
there was at least one $\alpha$-particle track. Since it is
virtually impossible to overlook the presence of such
a track in the searched field of view, these workers
could be certain of observing all the events in a given
area of plate and so were sure of obtaining a true
random sample of the events being studied. Events
which involve only electrons, however, are by no means
certain of being found in a systematic search. Each
event in a searched area of plate has a probability of being found which depends on (a) the size and orientation of the RaD cluster, (b) the distinctness of the RaE β-particle track, (c) the alertness of the observer, (d) the conceptual image of an event which happens to be uppermost in the observer's mind at the time of the search, and (e) the presence and orientation of background electron tracks near the event.

(a) obviously favours the observation of long tracks with high grain counts, lying in the horizontal plane. (b) should produce no distortion. The effect of (c) and (e) is probably that both tiredness and the presence of confusing background tracks accentuate the effect of (a) and (d). This latter distorting effect arises from the fact that the finding of an event (whether α tracks are present or not) is largely a matter of "recognition" - the eye registers or does not register a given pattern according to whether it does or does not approximate to pattern retained in the mind. (For example, it is sometimes possible, by visualising the appearance of a certain number, to pick it out from a list of numbers without actually reading all the numbers). The author has reason to believe that in the tedium of searching (usually only 5 or 6 events were found in a day) a run of events of a certain kind was sometimes allowed to fix the pattern by which succeeding events were picked out.
To the distortion produced by the method of finding events, there was probably added in the present instance, a distortion produced by the process of accepting or rejecting events. The histogram of fig. 11 is intended to represent these events in which there is no L-Auger electron, but in fact it is (on the most favourable interpretation) the histogram of events which were "single" and also looked "single". It is probable that the proportion of true "single" events which also look "single" is a function of grain count. The tracks of highest grain count have the highest probability of leaving the origin with low grain density, the least probability of having misleading spurs (due to scattering) at the origin. At the other end of the scale, events of 2 to 5 grains are unlikely to appear "double" since clusters of this size are usually compact, with no apparent bends or gaps. It appears likely then that it was in the middle region of 10-20 grains that the highest rate of rejection of the "single" events occurred.

These possible causes of distortion explained the occurrence of the high grain count group in fig. 11 and indicated that reliable results would only be obtained if the method of observation was such that (a) all events found were used for observation and (b) the probability of finding an event did not depend on the size, orientation or appearance of the origin.
cluster of the event.

To realise condition (b) the following method of finding events was used in the observations to be described in Chapter V. In any field of view, an electron track was picked out which looked as if it might be a RaE $\beta$-particle track, that is, a track having the grain density and degree of scattering appropriate to an electron of 200-1,000 keV. The track was followed, if possible, to its "ends". Often the track was a cosmic ray track which entered the emulsion at one point and left at another, or which entered and ended within the emulsion. Sometimes it was "lost" through becoming entangled with other tracks. But in some cases the electron track was found to originate in the emulsion. (An "end" of a track could be unambiguously distinguished from an "origin" by its showing a great increase in grain density towards the end, accompanied by an increasing tortuousness of path.) Clearly, the factors which governed the choosing of the track to be followed, and the possibility of following it to its origin had no dependence on the nature of the event to be found at the origin, so that the events so found were a true random sample of all events. In order to ensure that no event found in this way was observed twice at different times, the notebook in which the events was recorded had a page reserved for each square millimetre of the area to be searched, and each event was drawn on
the page appropriate to the square in which it was found.

**The Ground State to Ground State Transition.**

The question remained: what is the explanation of the group at 1 to 7 grains? Although, as has been mentioned, these events, once found, had a low probability of rejection, it appeared that this would be outweighed or at least counterbalanced by the comparative difficulty of finding them in the first place. It seemed, therefore, that 1-7 grain events could not be grossly over-represented in the histogram — certainly not to such an extent that the natural "low grain count tail" of the $E_L$ peak could be inflated to become comparable with the peak itself. (On the basis of work by Zajac and Bisgard (74) the intensity of the $E_L$ group should be completely negligible below 6 grains.) The presence of this group then was either indicative of a mode other than the $\beta_{47,e}$ mode or was due to some as yet unexplained manifestation of this mode. (At this stage there was plenty of room for conjecture: was it possible for instance that 9 grain tracks, say, could appear to have 2-5 grains if they went straight down?) The low grain count events could be explained as internal conversion electrons of an intermediate $\gamma$-ray of the 47 keV excited state, on the assumption that the accompanying $\gamma$-ray escaped. But this would involve
such a large total intensity of soft conversion electrons that they could not have failed to be detected by $\beta$-spectroscopy.

At about this time, the author was considering a crystalline diffraction method (after the method of von Friesen (77)) for the $\beta$-spectroscopy of RaD, and this involved consideration of the shape of the $\beta_0$ spectrum. Roughly it is
\[
N(w)dw \propto (w - W_0)^2 dw
\]
(fig. 12, see page 83) where $W_0$ is the maximum energy $\approx 15 + 47 = 62$ keV. Later, on looking again at the histogram of fig. 11, it became apparent that this spectrum was so much concentrated at the low energy end that, in conditions of low statistical reliability, it might well appear as a low grain count group. Thus the occurrence of a large number of low grain count events was both an intimation that the $\beta_0$ transition probably takes place, and an indication of the only method which could conclusively demonstrate its existence - an examination of the frequency of occurrence of low grain count events.

The fact that several low grain count events were also found in the first group of observations (fig. 10 page 54) tended to substantiate the conclusion that there was a significant number of 1-7 grain events. The conditions of observation of the first group were such that low grain count events were almost certainly under-represented.
\[ N(W) dw \sim (W_0 - W)^2 dw. \]
Examination of the Ends of Tracks.

Before beginning observations of the last group of 801 events it was decided, with the intention of finding out whether any information could be derived from the appearance of each RaE cluster, to examine the frequency with which true "single" events could appear to be "double" events. This was done by studying the ends of RaE $\beta$-particle tracks or background tracks. For example, the last 13 grains, say, of a track ending in the emulsion simulates a true "single" event of 13 grains. Any appearance of multiplicity at the assumed origin is necessarily spurious. 54 ends of tracks were studied, each being drawn as shown in the two examples in fig. 13. For each end, the assumed origin was imagined to be at successive positions along the track and notes were made on the probable interpretation of the simulated event for various orientations of an imagined RaE $\beta$-particle track leaving the assumed origin. It is difficult to give quantitative expression to results of such a survey but the conclusion was nevertheless quite definite that no information whatever could be derived from appearance. For instance: the most frequent cause of interpreting an event as "double" was the occurrence of a clear break in the cluster near the origin (the latter defined by the direction of the RaE $\beta$ track). The following table gives the probability (deduced from
these observations) that a true "single" track of given grain count has at least one break in it.

<table>
<thead>
<tr>
<th>g-count</th>
<th>4</th>
<th>6</th>
<th>8</th>
<th>10</th>
<th>12</th>
<th>14</th>
<th>16</th>
<th>18</th>
<th>20</th>
</tr>
</thead>
<tbody>
<tr>
<td>probabiiity %</td>
<td>6</td>
<td>12</td>
<td>27</td>
<td>50</td>
<td>54</td>
<td>73</td>
<td>77</td>
<td>80</td>
<td>90</td>
</tr>
</tbody>
</table>

A 21 grain track has a 60% probability of having 3 or more breaks in it. No further comment is required.

Conclusions.

This preliminary study revealed the scope and the limitation of the method. There was no possibility of adding to information on the following disputed topics:

- The shape of the $\beta_{47}$ spectrum below 3 keV.
- The maximum energy of the $\beta_{47}$ transition.
- The intensities and relative intensities of conversion electron lines, the Auger yield of the L-shell etc.

On the other hand, it seemed likely that the peculiar advantages of the method - the absence of the extraneous scattering of electrons (by source backing, walls etc.) which afflicts other methods - could lead to a conclusion on the topic which is at present of greatest interest, the occurrence of the $\beta_0$ mode.

The method of making the final group of observations also became clear.

(1) The events were to be found in the random
way already described.

(2) All events found were to be accepted for observation; only the total grain count of each cluster is of any significance.

(3) The presence of the $\beta_0$ transition would be revealed, if at all, only by an examination of the low grain count end of the grain count distribution.
Chapter V.

The Final Group of Observations:

801 Events.

The events were found as described in Chapter IV. The observations made on each origin cluster of grains were as follows:

(1) The total number of grains in the origin cluster of grains was counted.

(2) It was realised that since it can only be said that the RaE $\beta$-particle track starts somewhere in the origin cluster, there is a possibility that one or more of the grains within the cluster belongs to the RaE $\beta$ track. To furnish data for estimating the effect of this, two further observations were made:

(a) In events 29 - 801, the first 20-25 blobs of the RaE $\beta$ track were examined and the number of 1 grain blobs, 2 grain blobs etc. found. The length of track occupied by the first 20-25 blobs was measured. (The number of blobs examined was largely governed by the convenience in measuring the length.)

(b) In events 200 - 801, a length $d$ was measured - the length of the intercept on the line of the RaE $\beta$ track cut off by the area of the origin cluster. The true site of each event lay somewhere on this intercept (which in cases where the origin cluster
was highly curved might, rarely, consist of two separate parts). These lengths were measured in three dimensions. The horizontal projections of measured segments of track were estimated by means of an eyepiece graticule whose divisions were calibrated against a stage micrometer (1 division was 1.37 \( \mu \)). The vertical projections of the segments were measured by focusing on the top and bottom ends of each segment and reading the divided rim of the fine-focusing knob. For very accurate work it is essential to find the thickness of unprocessed emulsion which corresponds to the thickness of the processed emulsion; this can be done by a study of the ranges of a monoenergetic group of \( \alpha \)-particle tracks distributed isotropically in the emulsion (see, for instance, B.F. Bayman, Thesis 1954, p. 31). For the present work, however, great accuracy was not required in range measurement, and, following the experience of Bayman and other workers, the thickness of unprocessed emulsion was taken to be 190 \( \mu \). This implies that after development about 10 \( \mu \) of emulsion is removed in rubbing off the layer of silver (page 40). The fine-focusing adjustment could thus be calibrated (in microns of unprocessed emulsion) by focusing on the top and the bottom of the emulsion and equating the difference in readings to 190 \( \mu \). This was done once a day, since the thickness of the processed emulsion
varies according to atmospheric humidity.

Fig. 14 shows the grain count distribution of the origin clusters of grains for the 801 events. Origin clusters of 1-5 grains looked just like, and possibly were, the first blobs of the RaE β tracks. The smooth curve was calculated for the main peak, using a 7-term smoothing formula (79):

\[ u'_0 = \frac{1}{21} \left\{ 7u_0 + 6(u_1 + u_{-1}) + 3(u_2 + u_{-2}) - 2(u_3 + u_{-3}) \right\}^2 \]

Since it would seem at first sight that the grain counting of compact clusters would necessarily be highly subjective and that the subjective standards set up might vary over long periods of observation, it was of interest to compare the grain count histogram of the first 400 events with that of the second 400 (fig. 15). The smoothed curves are also compared. Although a period of 3 months elapsed between ending the first series of observations and beginning the second, there is remarkable agreement between the two sets.

Fig. 16 compares histograms of the events taken by hundreds. They show little detailed resemblance to each other and demonstrate that a peak must be 40-80 events high before the group profile approaches reliability.

It is not sufficient to demonstrate only the reproducibility of the distribution of fig. 14, since factors which introduce distortions may well be
FIGURE 14

No. of events

Total no. of grains
FIGURE 15
FIGURE 16
reproducible. The question arises: does the histogram correspond with what might be expected on the basis of the information available? Since the intensity of the X-ray is about 3.6% (page 13) this is also the fraction of events in which the $\beta_{47}$ particle is the only electron emission. These events must contribute to the 1-5 grains group. The main group from 5 to 36 grains must be composed largely of those events (about 85%, page 15) in which the $\beta_{47}$ particle is accompanied by a conversion electron and possibly Auger electrons. The expected grain count distributions for the origin clusters of events of these kinds will now be calculated. Since the expected grain count spectrum for the hypothetical $\beta_0$ transition will be required later, and since its calculation follows the same procedure, it will also be derived here. A preliminary discussion is given of the effect of counting grains from the RaE $\beta$ tracks as part of the RaD cluster. The results will be required in the calculations.

Allowance for grains of RaE $\beta$ track.

The observations 2(a) (page 66), taken over 773 events, established the blob constitution of the average observed RaE $\beta$ track. The relative frequency of occurrence of 1 grain blobs, 2 grain blobs, etc. was found to be
It is required to find the probability that $n$ blobs of the RaE $\beta$ track are counted as being part of the origin cluster. Let $d$ be the length of the intercept measured in observation 2(b) and $b$ be the number of blobs per micron on the RaE $\beta$ track. Let $x$ be measured along the intercept as shown in the diagram.

What is the probability $p(x)dx$ that the $\beta$ track starts in the interval $dx$? It is impossible to formulate any general answer to this question. For an electron of low energy forming a tight cluster tending to the spherical, the most probable position of the origin would be at the centre of the intersect $d$. For straighter, less highly scattered tracks, the most probable position would be at one or other of the
ends, depending on the appearance of the track. It will be mathematically convenient to assume a rectangular probability distribution, \( p(x)dx = \frac{dx}{d} \). This will serve to calculate the magnitude of the effect, which will later be seen to be small. Assuming that blobs occur at random with an average frequency of \( b \) per micron, the probability of \( n \) blobs (of a track starting in \( dx \)) occurring within the cluster, i.e. in the interval \( (d - x) \) is given by the Poisson probability distribution

\[
\mathbb{P}(n) = \left( \frac{b(d - x)}{n!} \right)^n e^{-b(d - x)}.
\]

The probability of \( n \) blobs occurring in the cluster is therefore

\[
P(n) = \int_0^d p(x) \, dx \cdot \mathbb{P}(n)
\]

\[
= \int_0^d \frac{1}{d} \left( \frac{b(d - x)}{n!} \right)^n e^{-b(d - x)} \, dx.
\]

Evaluating the integral, it is found that

\[
P(0) = \frac{1}{bd} \left( 1 - e^{-bd} \right)
\]

\[
P(1) = \frac{1}{bd} \left\{ 1 - e^{-bd}(1 + bd) \right\}
\]

\[
P(n) = \left\{ \frac{1}{bd} \left[ 1 - e^{-bd}(1 + bd + \frac{1}{2}bd^2 + \ldots + \frac{1}{n!}bd^n) \right] \right\} (l)
\]
Using these results it was possible to estimate the correction to be applied to the main peak of the histogram. Several methods were considered, all open to objections of one sort or another, and some more laborious than others. Eventually it was decided to apply the correction to each of a sample of observed events and to take \( b \) as being simply the total number of grains per micron of the RaE track associated with each event. The implication here is that the only mechanism for the formation of blobs of more than one grain is the random occurrence of two or more grains in such a small interval of track that they appear as a continuous group. If this were true, all blobs would be a "single file" of grains along the trajectory. In fact, a study of 168 events showed that a third of all 2 grain blobs and about a half of all 3, 4 and 5 grain blobs (in all, about 10% of all blobs) did not lie along the track. Nevertheless, this method was the best of the ones available.

The correction was applied as follows: graphs of \( P(0), P(1), P(2), P(3) \) were drawn as functions of the product \( abd \). The value of \( bd \) was found for each event from observations 2(a, b), and the probabilities \( P_m(n) \) found from the graph. \( P_m(n) \) symbolises the probability that the \( m \)th \( N \)-grain event has \( n \) RaE track grains counted in its origin cluster, i.e. that this \( N \)-grain event is really an \((N - n)\) grain RaD event.
The height of the \(N\)-grain column of the corrected histogram is then

\[
\sum_{m} \frac{N_{m} P(0)}{m} + \sum_{f} \frac{N_{f} P(1)}{f} + \sum_{s} \frac{N_{s} P(2)}{s} + \ldots
\]

where \(\sum_{f}\), for example, indicates summation of \(P(1)\) over all the \((N + 1)\) grain observed events.

Fig. 17 shows the effect of the correction applied to a sample of 166 events. It is concluded that the effect is to raise the ordinates of the low grain count edge by a factor of 14% and to depress the trailing edge ordinates by a similar factor. When this correction is applied to the smoothed curve of fig. 14, the net result is that the main peak is moved bodily through 0.5 grain in the negative direction.

It can therefore be said that in calculating the expected grain count distribution for events in which the \(\beta_{47}\) particle is accompanied by a conversion electron, the effect of RaE \(\beta\)-particle grains is allowed for by adding 0.5 grain.

For events of lower grain count (0 - 6 grains) the origin clusters have a less variable appearance, and it is possible (and desirable) to calculate the effect in a more straightforward fashion.

In this case it proved unnecessary to make a simplifying assumption about the mechanism of formation.
of blobs along the RaE β track. Let $t_{s}P(n)$ be the probability that $n$ $t$-grain blobs are counted along with an $s$-grain RaD cluster, so that the observed origin cluster has $(s + nt)$ grains. Then

$$t_{s}P(n) = \text{function}(d_{s}, t)$$

where the function is defined in formula (1) page 71. $d_{t}$ is the average number of $t$-grain blobs per micron of RaE β track. A little thought shows that $d_{s}, t$ must be taken to be $(d_{s} + \frac{1}{2}d_{t})$ where $d_{s}$ and $d_{t}$, found by observation, are the average diameters of $s$- and $t$-grain clusters (or blobs) respectively.

$t_{s}P(n)$ was calculated for $s = 1$ to 6 grains, $t = 1$ to 4 grains and $n = 0, 1, 2$. The probability of $g$ grains from a RaE β track being associated with an $s$-grain RaD cluster could then be found. For example, the probability of 3 RaE β grains being counted with a 4 grain RaD cluster is

$$\begin{align*}
\frac{1}{4}P(3) & \frac{2}{4}P(0) \frac{3}{4}P(0) \ldots \ldots \quad \text{(chance of 3 1-grain blobs)} \\
+ \frac{1}{4}P(2) & \frac{2}{4}P(1) \frac{3}{4}P(0) \ldots \ldots \text{(chance of 2 1-grain and 1 2-grain blobs)} \\
+ \frac{3}{4}P(1) & \frac{1}{4}P(0) \frac{2}{4}P(0) \frac{4}{4}P(0) \ldots \ldots \quad \text{(chance of 1 3-grain blob)}.
\end{align*}$$

The results of a series of such calculations (which are less difficult than they look) are given in the following table which tabulates the probability (%) of an $s$-grain RaD cluster being associated with $g$ grains.
from the RaE β track.

<table>
<thead>
<tr>
<th>RaE grains (g)</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>RaD grains (s)</td>
<td>1</td>
<td>83</td>
<td>11</td>
<td>4.5</td>
<td>0.5</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>78</td>
<td>15</td>
<td>6</td>
<td>0.8</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>70</td>
<td>18.5</td>
<td>8.5</td>
<td>2.5</td>
<td>0.2</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>66</td>
<td>20</td>
<td>10</td>
<td>3.0</td>
<td>0.3</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>62</td>
<td>22</td>
<td>12.5</td>
<td>3.3</td>
<td>0.3</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>60</td>
<td>23</td>
<td>13.0</td>
<td>3.6</td>
<td>0.6</td>
</tr>
</tbody>
</table>

Table X.

The β\(_{47}\) Spectrum.

As shown in Chapter I, pages 8, 9 (see also fig.1) the β\(_{47}\) transition is first forbidden with zero spin change. The β-spectrum is therefore expected to have the "allowed" shape and this is confirmed by the work of Jaffe and Cohen (35). The energy spectrum is then

\[ N(W) \, dw \propto P(Z,W) \, pW(W_0 - W)^2 \, dw \]

where \( p \) is the momentum of the β-particle and \( W \), the energy, includes the rest mass energy \( m_0 c^2 \). \( W_0 \) is the maximum energy.

\( P(Z,W) \), the Fermi coulomb field factor, is, for very low values of \( W_0 \), approximately equal to \( w/p \) (80). Since also, for low values of \( W_0 \), \( W \) is nearly constant,
76.

the spectrum becomes

\[ N(E) \, dE \propto (E_0 - E)^2 \, dE \]

where \( E \) is the kinetic energy.

In order to calculate the corresponding grain count spectrum the extrapolated Zajac-Ross calibration was used. It will be seen later that the author's grain counting corresponded well with that of Zajac, but, in any case, the final results will not depend strongly on the complete precision of the calibration used.

It was assumed that energies corresponding to half-integral grain counts were "watershed" values; i.e. that electrons of all energies between two "watershed" values produce the median integral number of grains. The table illustrates this, and gives the probabilities of a \( \beta \) particle producing an \( n \)-grain cluster. These probabilities are given by

\[
\int_{E_1}^{E_2} (E_0 - E')^2 \, dE' \div \int_{0}^{E_0} (E_0 - E)^2 \, dE
\]

where \( E_1 \) and \( E_2 \) are adjacent watershed values. In accordance with the conclusions reached on page 12, the calculations are done for maximum energies of 15 keV and 20 keV.
Table XI.

<table>
<thead>
<tr>
<th>grain limits</th>
<th>0.5</th>
<th>1.5</th>
<th>2.5</th>
<th>3.5</th>
<th>4.5</th>
<th>5.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>'watershed' energies (keV)</td>
<td>2.4</td>
<td>6.0</td>
<td>9.2</td>
<td>12.3</td>
<td>15.0</td>
<td>18.0</td>
</tr>
<tr>
<td>RaD grains (s)</td>
<td>0</td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>4</td>
<td>5</td>
</tr>
<tr>
<td>47 grain spectrum (%)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>15 keV</td>
<td>42</td>
<td>38</td>
<td>15</td>
<td>4</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>20 keV</td>
<td>33</td>
<td>34</td>
<td>18</td>
<td>9</td>
<td>4</td>
<td>1</td>
</tr>
</tbody>
</table>

These results are corrected for the presence of grains from RaE \( \beta \) tracks by using Tables IX (page 70) and X (page 75). For example, from Table X, it is seen that there is a 6% probability of 2 grain RaD cluster appearing as a 4 grain origin cluster. For zero-grain RaD events, the "origin cluster" is simply the first blob of the RaE \( \beta \) track. The expected grain count spectrum of this first blob is given in Table IX.

In principle, there should be an additional correction for the finite energy resolving power of the grain counting process. But, at low grain counts the resolution (in absolute terms) is so sharp that the correction is negligible. The final grain count spectrum of origin clusters of events in which the \( \beta_{47} \) particle is the only emitted electron is then
Table XII.

<table>
<thead>
<tr>
<th>No. of grains</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Corrected $\beta_{47}$ spectrum (%)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>15 keV</td>
<td>63</td>
<td>24</td>
<td>8</td>
<td>3.6</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>20 keV</td>
<td>54</td>
<td>25</td>
<td>13</td>
<td>6</td>
<td>1.6</td>
<td>0.3</td>
</tr>
</tbody>
</table>

Calculated Spectrum for Events with Conversion Electron.

This calculation was based on the following data.

(1) The Zajac-Ross calibration gives the mean grain counts for the conversion electron groups. The work of Wu et al. (page 17) gives the relative intensities of the groups.

Table XIII.

<table>
<thead>
<tr>
<th>Conversion electron</th>
<th>$L_I$</th>
<th>$L_{II}$</th>
<th>$L_{III}$</th>
<th>$M_{I-III}$</th>
<th>$M_{IV-V}$</th>
<th>$N_{I-V}$</th>
<th>$N_{VI-VIII-O}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy (keV)</td>
<td>30</td>
<td>31</td>
<td>33</td>
<td>43</td>
<td>46</td>
<td>46</td>
<td></td>
</tr>
<tr>
<td>Mean grain count</td>
<td>10.0</td>
<td>10.5</td>
<td></td>
<td>16.0</td>
<td>17.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Relative intensity</td>
<td>100</td>
<td>7.5</td>
<td>0.7</td>
<td>25</td>
<td>0.6</td>
<td>7.0</td>
<td>0.7</td>
</tr>
</tbody>
</table>

(2) The only information on line-width and line shape at low grain counts is that due to Bisgard (74). His line-shape for a grain count group centred at about
6 grains (produced by nearly monoenergetic conversion electrons) was

<table>
<thead>
<tr>
<th>No. of grains</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
<th>9</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. of events</td>
<td>16</td>
<td>55</td>
<td>83</td>
<td>129</td>
<td>75</td>
<td>36</td>
<td>14</td>
</tr>
</tbody>
</table>

The "line-width" (standard deviation) is about 32% of the mean. This is larger than that found by Zajac (about 20%) and perhaps indicates the effects of not seeing some grains in tightly bunched clusters. The work of Zajac shows that the line-width is proportional to the mean grain count.

(3) The spectra of L Auger electrons were calculated from data given by Ross et al. (51).

Calculation.

Curves were drawn representing the L_I, L_II, M and N conversion groups, each having the shape of the Biogard distribution but broadened in proportion to energy. They were centred at the appropriate grain count and their relative areas adjusted to the values given in Table XIII.

Each electron in these groups is accompanied by a $\beta_{47}$ particle whose grain count spectrum is given in Table XI (a maximum energy of 15 keV was chosen, page 12) and the groups were corrected to take this into account.
For example, the contribution of the N events (say) in the 10 grain column, to the 10, 11, 12, 13 and 14 grain columns of the corrected curve is 0.42N, 0.38N, 0.15N, 0.04N and 0.01N, since 0.42 ... 0.01 are the probabilities of 0, 1, 2, 3 and 4 βU grains being added to the 10 grain conversion electron track.

A certain fraction $x_1$ of $L_I$ conversion electrons and $x_2$ of $L_{II}$ conversion electrons are also accompanied by Auger electrons. These fractions are

$$
x_1 = a_1 + a_2 f_{12} + a_3 f_{13} + a_3 f_{23} f_{12}
$$
$$
x_2 = a_2 + a_3 f_{23}
$$

where $a$ and $f$ are the Auger and Coster-Kronig yields of the L sub-shells; the suffices denote the level(s) concerned.

Assuming a total L fluorescence yield of 40%, Ross et al. (51) arrived at the values for $f$ and $a$ tabulated below. Values calculated by the author to see the effect of assuming a 30% fluorescence yield (see page 20), are also given, together with the derived values of $x_1$ and $x_2$.

<table>
<thead>
<tr>
<th>Fluor.</th>
<th>$a_1$</th>
<th>$a_2$</th>
<th>$a_3$</th>
<th>$f_{12}$</th>
<th>$f_{13}$</th>
<th>$f_{23}$</th>
<th>$x_1$</th>
<th>$x_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>40%</td>
<td>0.105</td>
<td>0.67</td>
<td>0.61</td>
<td>0.18</td>
<td>0.605</td>
<td>0.60</td>
<td>0.60</td>
<td>0.67</td>
</tr>
<tr>
<td>30%</td>
<td>0.123</td>
<td>0.76</td>
<td>0.70</td>
<td>0.17</td>
<td>0.58</td>
<td>0</td>
<td>0.71</td>
<td>0.76</td>
</tr>
</tbody>
</table>
Tables 5 and 7 in the paper of Ross et al. supply the information needed for the breakdown of the Auger electrons into their energy groups. For example, of the L\textsubscript{I} conversion electrons, a fraction $a_{21}^{12}$ are accompanied by L\textsubscript{II} Auger electrons, and these in turn are divided in the ratio 12:4 between the groups $L_{II} \rightarrow M(M(7.5 - 10.4 \text{ keV})$ and $L_{II} \rightarrow M(N)(10.7 - 13 \text{ keV})$.

A series of calculations of this kind produced the following grain count distributions (%) for Auger electrons accompanying L\textsubscript{I} and L\textsubscript{II} conversion electrons.

<table>
<thead>
<tr>
<th>No. of grains:</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distribution (%)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Auger electrons with $e_{L_{I}}$</td>
<td>55</td>
<td>40</td>
<td>5</td>
</tr>
<tr>
<td>Auger electrons with $e_{L_{II}}$</td>
<td>37</td>
<td>50</td>
<td>13</td>
</tr>
</tbody>
</table>

Fractions $x_1$ and $x_2$ of the L\textsubscript{I} and L\textsubscript{II} conversion electron groups respectively were corrected for the presence of these grain count spectra, in a way similar to that described for the $F_{47}$ correction.

All conversion electrons are accompanied by 1, 2 or 3 M Auger electrons but almost none of these are above the 2.4 keV threshold for the activation of one grain, and so they can be ignored.

The curve resulting from the above calculations was finally corrected for the presence of Ra\textsubscript{E} $\beta$ track
grains by moving it through 0.5 grain towards higher grain counts (see page 73). The final curve will be referred to as spectrum A.

Fig. 18 shows spectrum A (full line) compared with the histogram of fig. 14 (page 69). It is also shown (dotted line) moved to the left through 1 grain. This is done to show the extent of the discrepancy and to show how well the shape of the experimental histogram agrees with the shape expected on the basis of the available data. The version of spectrum A shown is that calculated for an L fluorescence yield of 40%. The other is so nearly identical that to draw it too would confuse the figure. Its peak is 0.2 grain to the right of the peak shown, and most of it is 0.2-0.3 grain to the right. The high grain count tails coincide.

At first sight it would seem that the author's grain counting is about 1 grain less than that of Zajac at about 11 grains. However, for the following reasons, it is concluded that this is not so: the 8-15 grain region of the calculated curve is largely made up of $({}^{3}P_{46} + e_L + A_L)$ events. When one low and two very low energy electrons are emitted from the same point in the emulsion, it seems reasonable that the observed grain count should be somewhat lower than the sum of the three expected grain counts, either because
the probability of a grain being hidden is increased in such conditions, or because there is a large chance that two separate electrons pass through and activate the same grain. This suggestion is supported by the fact that the experimental and calculated spectra agree very well above 18 grains - the region where MN conversion electrons, unaccompanied by Auger electrons, supply the main contribution.

The conclusion from figs. 15 and 18 can thus be stated. The grain counting process of the author was steady and reliable and did not depart significantly from that of Zajac. The method of finding events was truly random and did not introduce systematic distortion in the 6-36 grain region.

The results of this chapter will wholly depend on the demonstration that there is an excess of events in the 1-6 grain range. Any imaginable cause of distortion in this region would, if anything, cause a deficiency of events. This will be discussed later.

The $\beta_0$ Spectrum.

Discussion of the Energy Spectrum.

The $\beta_0$ transition, if it takes place, is between the RaD ground state (spin 0, even parity) and the RaE ground state (spin 1, odd parity). It is therefore a first forbidden transition, with the spin change equal to the degree of forbiddenness, and it would in general
be expected to have the "allowed" shape. However, an unques­tioning assumption that this is so might in this particular case lead to error. Of the many first forbidden \((\Delta I = 0,1)\) \(\beta\)-transitions whose spectra have been adequately measured, only that of RaE departs appreciably from the allowed shape. The RaD transi­tion is similar to that of RaE. The RaD \(\rightarrow\) RaE \(\rightarrow\) RaF sequence is \((0, +) \rightarrow (1, -) \rightarrow (0, +)\); the particles outside closed shells are \((2\text{ neutrons}) \rightarrow (1\text{ neutron,}
\) \(1\text{ proton}) \rightarrow (2\text{ protons})\) so that there is a striking symmetry between the two transitions. It is therefore necessary to consider the factors which affect the shape of the RaE \(\beta\)-spectrum, and see if they are likely to affect the \(\beta_0\)-spectrum.

The energy spectrum is

\[
N(w) \, dw \propto S(z, w) \cdot F(z, w) \cdot p(w - w_0)^2 \, dw.
\]

The Shape Correction Factor \(S\). In general, the shape factor for first forbidden, \(I = 1\), transitions is (81)

\[
S \propto \left\{ (1 - \Lambda) C_T + \frac{1}{\varepsilon} C_S \right\} \frac{\alpha z}{2R} + S'(C_T, C_S, \varepsilon, \omega)
\]

where \(C_T\) and \(C_S\) are respectively the coefficients in the linear combination of the tensor \((T)\) and scalar \((S)\) interactions (only these are now believed to play a part in the interaction); \(\Lambda\) and \(\varepsilon\) are ratios of the nuclear matrix elements:
\[ \Lambda = \frac{\int \beta \chi}{\int \beta \sigma^a \Lambda R} \frac{2}{2} \rho \left( i \int \beta \sigma \frac{R}{\beta R} \right) \]

\( \mathcal{L}_{Z/R} \) is the coulomb potential energy of an electron at the nuclear radius \( R \).

Since \( \mathcal{L}_{Z/R} \) is always much greater than the maximum kinetic energy \( (W_0 - 1) \) of the \( \beta \)-particle, the energy dependent term \( S' \) is in general negligible. \( S \) is then a constant which affects the \( ft \)-value, but not the shape. The shape of the RaE spectrum, however, departs radically from the allowed shape and it is assumed that this is caused by the near cancellation of the large energy-independent term; that is,

\[ C_T (1 - \Lambda) \approx \gamma \approx 0. \]

Is there any reason to believe that such a cancellation occurs in the case of the RaD \( \rightarrow \) RaE transition? This question is of more than incidental significance and it will be discussed fully in the final chapter. A long digression will be avoided here by merely quoting the results of this discussion.

The evidence is not sufficient to define the shape factor. It can only indicate that it is likely that some significant degree of cancellation takes place, and predict what the shape factor will be in the extreme case of complete cancellation.

The shape factor is given conveniently by the
formula of Plassmen and Langer (83)

\[ s = x^2 - \frac{2}{3}(1 - \frac{f}{2})q\pi - 4x(1 + \frac{f}{2}) \frac{p^2 + \frac{Q^2Z^2}{3}}{(Q + 1)(2Q + 1)} + \frac{q^2}{6}(1 + 2\frac{f}{2})^2 + \frac{4}{3}q(1 - \frac{f}{2})^2 \frac{p^2 + \frac{Q^2Z^2}{3}}{(Q + 1)(2Q + 1)} + 4(1 + \frac{f}{2})^2 \frac{p^2 + \frac{Q^2Z^2}{3}}{(Q + 1)^2(2Q + 1)^2} + \frac{1}{1 + q}L_1 \]

where \( x = \frac{Q^2}{2N}(\Lambda - 2 \frac{1 + f}{q}) \)

\[ f = \frac{1}{C} \frac{C_S}{C_T} = \frac{C_S}{C_T} \int \beta \gamma \]

\[ \lambda = \frac{1}{137} \]

\[ q = (1 - \frac{Q^2Z^2}{3})^\frac{1}{2} \]

\( L_1 \) is a tabulated function (84)

\( p \) and \( q \) are the momenta of the \( \beta \)-particle and neutrino respectively.

The greatest degree of energy dependence is produced when the energy independent term is zero, i.e. when \( x = 0 \). It will be shown in Chapter VII that the best guess at the corresponding value of \( f \) is \( f = 0.70 \). The dominant term is then the penultimate one. It turns out that this "extreme" shape factor \( S_{\text{EXT}} \) is closely linear.

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>0</th>
<th>20</th>
<th>40</th>
<th>60</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shape factor ( S_{\text{EXT}} )</td>
<td>1.00</td>
<td>1.20</td>
<td>1.40</td>
<td>1.60</td>
</tr>
</tbody>
</table>
In what follows, calculations will be done using the two extreme cases - complete energy independence ("allowed" shape $S = 1$), and "extreme" energy dependence ($S = S_{\text{EXT}}$).

The Coulomb Factor $F$. The function $G = \frac{pF}{\sqrt{W}}$ is tabulated by Rose (85). The values of $pWF = GW^2$ are given below. They are linear against $W$.

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>0</th>
<th>20</th>
<th>40</th>
<th>60</th>
</tr>
</thead>
<tbody>
<tr>
<td>$pWF$</td>
<td>1.000</td>
<td>1.061</td>
<td>1.123</td>
<td>1.184</td>
</tr>
</tbody>
</table>

The Effect of Screening. The theoretical "allowed" $\beta$ spectrum shape is calculated for a bare nucleus. The effect of the atomic electrons is to modify the Fermi coulomb field factor in such a way as to reduce the transition probability, particularly for low energy $\beta$-particles. Fig. 19 shows the percentage deduction to be made from the allowed shape according to (a) the value of an approximate formula tabulated by the U.S. National Bureau of Standards (56) and (b) the work of Rietz (57) who calculated the Fermi function numerically for the atom as a whole, assuming that the electrons have the Thomas-Fermi distribution. Rietz's curve shows that the approximate formula cannot be relied on, but unfortunately it does not go below 25 keV. The effect is likely to be significant only at energies below a few keV. It will be ignored in the subsequent
FIGURE 19

% deduction

Rietz \( Z = 84 \)

N.B.S. \( Z = 80 \)

Energy of \( \beta \)-particle

0 100 200 300 keV
The \( \beta_0 \) Grain-Count Spectrum.

The \( \beta_0 \) energy spectra are calculated from the formula

\[
N(W) \, dw \propto (W - w_0)^2 (cW^2) \, dw
\]

taking the maximum energy \((w_0 - 1)\) to be 62 keV and 67 keV and putting \( S = 1 \) and \( S = S_{\text{EXT}} \) in both cases. The spectrum shapes are drawn on graph paper to supply the interpolated ordinate values needed for the calculation below.

To obtain the grain count spectrum, the energy "watershed" values are found as before by putting \( g = 0.5, 1.5, 2.5 \ldots 28.5 \) in the Zajac-Ross calibration formula

\[
E(g) = 4.13g^{0.84}.
\]

The relative intensity of, for example, the 9-grain column of the grain-count spectrum, is then given by the area of energy spectrum enclosed between \( E(8.5) \) and \( E(9.5) \). The four grain-count spectra are shown in Table XV (page 92) (the intensities are in arbitrary units).

**Effect of Finite Resolving Power.** In deriving these grain count spectra there is an implicit assumption that, apart from the effect produced by the integral
nature of grain counts, the "line-width" of a mono-
energetic group is zero. The method of correction for
the actual line width is as follows.

To ease the labour of calculation, it is assumed
that the line-shape is given by the normal error curve

\[ n(g) = \frac{1}{\sigma_0 \sqrt{2\pi}} e^{-\frac{1}{2} \left(\frac{g - g_0}{\sigma_0}\right)^2} \]

where \( g_0 \) is the mean grain count of the group and \( \sigma_0 \),
the standard deviation, is taken to be 0.32 \( g_0 \) in
accordance with the line shape of Bisgard. \( N(g_0) \)
is the total intensity of the group centred at \( g_0 \) and \( n(g) \)
is the height of the ordinate at \( g \). The ordinate of
the corrected curve at \( g = g_1 \) is then the sum of the
contributions made at \( g_1 \) by all the groups of the
uncorrected spectrum. Formally, it is given by

\[ \sum_{g_n = 0}^{g_{\text{max}}} \int_{g_n - \frac{1}{2}}^{g_n + \frac{1}{2}} \frac{N(g_n)}{\sigma_n \sqrt{2\pi}} e^{-\frac{1}{2} \left(\frac{g - g_n}{\sigma_n}\right)^2} \, dg. \]

The calculation will be made clear by reference to
fig. 20. Only two columns of the uncorrected (62 keV,
allowed shape) spectrum are shown, at 4 grains and 7
grains, of intensities 415 and 270 respectively. It
is required to find the contributions made by these
two columns to the corrected intensity at 4 grains.
The contribution by the uncorrected 4 grain column is
**FIGURE 20**

[Graph showing a distribution with labeled bars and values.]
the area of its line shape enclosed between \( g = 3.5 \) and \( g = 4.5 \). These limits correspond to deviations of \( \frac{1}{4} \) grain (or 0.39 \( \sigma_g \)) from the median. The area can be conveniently found in tables of the normal curve of error \((86), (277)^{-\frac{1}{2}} e^{-\frac{1}{2}t^2}\), by putting \( t = 0.39 \). The contribution is found to be

\[
2 \times 0.1517 \times 415 = 125.9.
\]

The contribution of the uncorrected 7 grain column is obtained by noting that the limits of deviation from the median are 2\( \frac{1}{2} \) grains (1.12 \( \sigma_g \)) and 3\( \frac{1}{2} \) grains (1.56 \( \sigma_g \)). The error table then shows that the contribution is

\[
(0.4406 - 0.3686)270 = 19.44.
\]

The total corrected intensity at 4 grains is obtained by summing all such contributions from all the columns of the uncorrected spectrum.

The potential usefulness of completely corrected grain count spectra did not justify the labour of obtaining them. It will be seen later that it is important to know the precise distribution of the calculated spectra in the 1-5 grain region. Therefore the effect of non-zero line-width was calculated only for 1-5 grain columns of the four \( \beta_0 \) spectra. The results are shown in Table XIV page 92.
RaE $\beta$-track grains. In cases where the $\beta_0$ particle is expected to produce no grains, the "origin cluster of grains" is simply the first blob of the RaE $\beta$-track, and so, for these events, the grain count distribution is given by Table IX (page 70). For other cases, the number in the $m$th row and $n$th column of Table X (page 75) gives the percentage probability that an $m$-grain RaD event appears to have an $(m+n)$ grain origin cluster. The correction for these effects has been carried out for the 0-5 grain region of the spectra. The results are shown in Table XIV (page 92).

**Examination of the Histogram.**

It has been pointed out in Chapter IV that an examination of the low grain count region of the spectrum would be crucial for the detection of the $\beta_0$ spectrum. The calculated spectra derived above make it possible to subtract the known contributions to this 1-5 grain region. They are as follows.

1. In the $3.8 \pm 0.6\%$ of disintegrations in which the $\gamma$-ray is emitted and undetected, only the $\beta_{47}$ particle produces grains. Therefore, of the 801 events observed, 30 $\pm$ 7.3 events are distributed in the $\beta_{47}$ grain count spectrum (Table XII, page 78). The error reflects not only the uncertainty in the experimental observation of Damon and Edwards, but
Table XIV.

Calculated $\beta_0$ grain count spectra - 62 keV.

<table>
<thead>
<tr>
<th>Grains</th>
<th>S = 1</th>
<th></th>
<th></th>
<th>S = EXT</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>X</td>
<td>Y</td>
<td>Z</td>
<td></td>
<td>X</td>
<td>Y</td>
</tr>
<tr>
<td>0</td>
<td>512</td>
<td>517</td>
<td>576</td>
<td>1</td>
<td>735</td>
<td>752</td>
</tr>
<tr>
<td>1</td>
<td>735</td>
<td>815 1091</td>
<td>629</td>
<td>2</td>
<td>579</td>
<td>612</td>
</tr>
<tr>
<td>2</td>
<td>477</td>
<td>447 462</td>
<td>399</td>
<td>3</td>
<td>415</td>
<td>404</td>
</tr>
<tr>
<td>3</td>
<td>352</td>
<td>341</td>
<td>339</td>
<td>4</td>
<td>310</td>
<td>367</td>
</tr>
<tr>
<td>4</td>
<td>238</td>
<td>295</td>
<td></td>
<td>5</td>
<td>177</td>
<td>226</td>
</tr>
<tr>
<td>5</td>
<td>128</td>
<td>170</td>
<td></td>
<td>6</td>
<td>91</td>
<td>125</td>
</tr>
<tr>
<td>7</td>
<td>35</td>
<td>52</td>
<td></td>
<td>8</td>
<td>6.5</td>
<td>15</td>
</tr>
<tr>
<td>10</td>
<td>0</td>
<td>0</td>
<td></td>
<td>12</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Total</td>
<td>5024</td>
<td>2920 5821</td>
<td>3123</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

X - uncorrected
Y - X corrected for non-zero line width
Z - Y corrected for addition of RaE track grain.
Table XIV.

Calculated $\beta_0$ grain count spectra - 67 keV.

<table>
<thead>
<tr>
<th>Grains</th>
<th>$S = 1$</th>
<th></th>
<th></th>
<th>$S = S_{EXT}$</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$X$</td>
<td>$Y$</td>
<td>$Z$</td>
<td>$X$</td>
<td>$Y$</td>
<td>$Z$</td>
</tr>
<tr>
<td>0</td>
<td>602</td>
<td>669</td>
<td></td>
<td>608</td>
<td>678</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>865</td>
<td>961</td>
<td>1286</td>
<td>890</td>
<td>1000</td>
<td>1325</td>
</tr>
<tr>
<td>2</td>
<td>686</td>
<td>668</td>
<td>751</td>
<td>730</td>
<td>723</td>
<td>800</td>
</tr>
<tr>
<td>3</td>
<td>565</td>
<td>537</td>
<td>552</td>
<td>626</td>
<td>599</td>
<td>606</td>
</tr>
<tr>
<td>4</td>
<td>502</td>
<td>494</td>
<td>483</td>
<td>565</td>
<td>560</td>
<td>545</td>
</tr>
<tr>
<td>5</td>
<td>435</td>
<td>424</td>
<td>419</td>
<td>500</td>
<td>495</td>
<td>483</td>
</tr>
<tr>
<td>6</td>
<td>385</td>
<td></td>
<td></td>
<td>455</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>305</td>
<td></td>
<td></td>
<td>375</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>236</td>
<td></td>
<td></td>
<td>300</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>177</td>
<td></td>
<td></td>
<td>236</td>
<td></td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>132</td>
<td></td>
<td></td>
<td>182</td>
<td></td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>61</td>
<td></td>
<td></td>
<td>90</td>
<td></td>
<td></td>
</tr>
<tr>
<td>22</td>
<td>21</td>
<td></td>
<td></td>
<td>32</td>
<td></td>
<td></td>
</tr>
<tr>
<td>26</td>
<td>2</td>
<td></td>
<td></td>
<td>4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>6364</td>
<td>3491</td>
<td>7434</td>
<td>3759</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* $X$ - uncorrected
  
  $Y$ - $X$ corrected for non-zero line width
  
  $Z$ - $Y$ corrected for addition of RaE track grain.
also the standard deviation, \(\sqrt{30}\), to be expected from statistical fluctuations. The spectrum is shown in Table XV, page 96, together with the other spectra listed below.

II(a). It was concluded in Chapter II that the process by which the RaD impregnating solution was prepared was 97.5 \(\pm\) 2.0\% efficient in removing RaE from the Ra(D + E + F) source. That is, at the time of separation, there was 2.5 \(\pm\) 2.0\% of the equilibrium concentration of RaE, present in the solution. Since two hours elapsed between the preparation of the source and the final drying of the plates after impregnation, it can be said that the concentration of RaE at the time when tracks began to be formed in the emulsion was 3.7 \(\pm\) 2.0\% of the equilibrium concentration. This concentration is \(0.037 \frac{\lambda_D}{\lambda_E}\) or \(2.3 \times 10^{-5}\) per RaD atom. After 19 days exposure a fraction \((1 - e^{-\lambda_E t})\) or 93\% of these RaE atoms had disintegrated giving RaE \(\beta\)-tracks. During this time the number of RaD \(\rightarrow\) RaE \(\rightarrow\) RaF disintegrations was

\[
1 - e^{-\lambda_D t} - \frac{\lambda_D}{\lambda_E} (1 - e^{-\lambda_E t})
\]

or \(105 \times 10^{-5}\) per RaD atom.

The fraction of all observable RaE tracks which were produced by RaE \(\rightarrow\) RaF disintegrations was therefore
The total number of such events occurring in the histogram is then $16 \pm 9.6$. The grain count distribution of their "origin clusters" is the grain count distribution of the first blob of the average RaE track (Table IX, page 76).

II(b). It has been mentioned in Chapter III, page 45, that about 1/3% of all cosmic ray background tracks seem to originate in the emulsion and are similar in appearance to typical RaE tracks. This figure will be used here, although, as has been said, it is certainly an overestimate. The figures given in Chapter III show that for every 45 RaE tracks observed, there are 22 curved cosmic ray tracks of which 0.82 track seem to originate in the emulsion. That is, in 801 observations on events associated with RaE-type tracks, at most $14 \pm 3.7$ involve background tracks. As in II(a), the grain count distribution of their "origin clusters" is given by Table IX. So altogether, $30 \pm 10.3$ events have this distribution, as shown in Table XV.

III. It is seen from spectrum A, fig. 18, page 82, that the expected contribution from the conversion electron peak in the 1-5 grain region is about 3 events, 2 at 5 grains, and 1 at 4 grains.

The total number of events in the 1-5 grain region
is therefore expected to be $63 \pm 12.6$. In fact, the histogram of fig. 14 shows that there are $108 \pm 10.4$ events in this region, so that there is an excess of $45 \pm 14.4$ events. (The error here is not the square root of the sum of the component squared errors, since the quantities represented by 60 and 108 are not independent - if the first quantity is larger than the medial value, the second would tend to be larger too. The error given is therefore $12.6 + \sqrt{48}$.) The probability that the expected group of $63 \pm 12.6$ events should by a chance fluctuation give rise to 108 observations is about $2 \times 10^{-4}$. Therefore, on the face of it, the occurrence of this excess is in itself a demonstration that there is some mode other than the (RaD $\rightarrow$ RaE$^{47} \rightarrow$ RaE) mode. (The analysis will be taken forward without critical digressions; a critical discussion follows.) However the grain count distribution of the excess is still more convincing.

Table XV shows the distributions discussed above, the observed distribution, and the distribution of the excess. In the calculation of the errors it was noted that each tabulated entry for the calculated spectra is subject to two errors, the constant error proper to the spectrum as a whole, and the statistical error proper to the entry itself. For example, the entry 18.9 for the 15 keV $\beta_{47}$ spectrum is subject to the fractional error $\frac{18.9 \times 0.6}{3.8}$ stemming from the
<table>
<thead>
<tr>
<th>Entrain</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>Totals</th>
</tr>
</thead>
<tbody>
<tr>
<td>I (β{\text{s}}) (15 keV)</td>
<td>18.9</td>
<td>7.2</td>
<td>2.4</td>
<td>1.1</td>
<td>0.3</td>
<td>0</td>
<td>30 ± 7.3</td>
</tr>
<tr>
<td></td>
<td>16.2</td>
<td>7.5</td>
<td>3.9</td>
<td>1.8</td>
<td>0.5</td>
<td>0.1</td>
<td>30 ± 7.3</td>
</tr>
<tr>
<td>II (RaE, background)</td>
<td>21.9</td>
<td>5.3</td>
<td>1.5</td>
<td>0.6</td>
<td>0.3</td>
<td>0.1</td>
<td>30 ± 10.3</td>
</tr>
<tr>
<td>III (conversion peak)</td>
<td></td>
<td></td>
<td>1.0</td>
<td>2.0</td>
<td></td>
<td></td>
<td>3</td>
</tr>
<tr>
<td>Total (15 keV)</td>
<td>40.8</td>
<td>12.5</td>
<td>3.9</td>
<td>2.7</td>
<td>2.6</td>
<td></td>
<td>63 ± 12.6</td>
</tr>
<tr>
<td>(20 keV)</td>
<td>38.1</td>
<td>12.8</td>
<td>5.4</td>
<td>3.4</td>
<td>2.8</td>
<td></td>
<td>63 ± 12.6</td>
</tr>
<tr>
<td>Errors</td>
<td>± 9.2</td>
<td>± 4.0</td>
<td>± 2.1</td>
<td>± 1.7</td>
<td>± 1.6</td>
<td></td>
<td>( \sqrt{\sum \sigma^2} = 12.6 )</td>
</tr>
<tr>
<td>Observed Distn.</td>
<td>42</td>
<td>23</td>
<td>19</td>
<td>13</td>
<td>11</td>
<td></td>
<td>108</td>
</tr>
<tr>
<td>Excess (15 keV)</td>
<td>1.2</td>
<td>10.5</td>
<td>15.1</td>
<td>10.3</td>
<td>8.4</td>
<td></td>
<td>45 ± 14.5</td>
</tr>
<tr>
<td>(20 keV)</td>
<td>3.9</td>
<td>10.2</td>
<td>13.6</td>
<td>9.6</td>
<td>8.2</td>
<td></td>
<td>45 ± 14.5</td>
</tr>
<tr>
<td>Errors</td>
<td>± 9.4</td>
<td>± 5.2</td>
<td>± 4.4</td>
<td>± 3.6</td>
<td>± 3.3</td>
<td></td>
<td>( \sqrt{\sum \sigma^2} = 14.5 )</td>
</tr>
</tbody>
</table>
uncertainty in Damon and Edwards' observation of the total intensity, and the statistical error $\sqrt{18.9}$. Since the errors under the headings "15 keV" and "20 keV" differ little from each other, a mean value is quoted for both.

Table XV.
(see opposite)

The point of the comment made above is now clear. The chance, for example, of the expected group at 3 grains (3.9 ± 2.1) giving rise by chance to 19 observed 3-grain events is utterly remote (deviation = 7\sigma).

Several tentative deductions can now be made as follows.

The excess events are to be ascribed to the $\beta_0$ spectrum. This spectrum is expected to increase in intensity with decreasing grain count whereas the observed excess falls off at 1, 2 grains. To avoid basing conclusions on a region which is possibly subject to observational error, therefore, the 3, 4, 5 grain region is used as a base for the estimation of the $\beta_0$ intensity. Table XV shows that the excess in this region is 33.8 ± 6.5 and 31.4 ± 6.5 events in the two cases (15 keV and 20 keV). Using these figures to "normalise" the spectra of Table XIV it appears that the expected $\beta_0$ spectrum (expressed in events) is
Table XVI.

<table>
<thead>
<tr>
<th>Grains</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\beta_0 \ { S = 1 }$</td>
<td>31</td>
<td>18</td>
<td>13</td>
<td>11</td>
<td>9</td>
</tr>
<tr>
<td>62 keV $\ { S = S_{\text{EXT}} }$</td>
<td>28</td>
<td>17</td>
<td>13</td>
<td>11</td>
<td>10</td>
</tr>
<tr>
<td>$\beta_0 \ { S = 1 }$</td>
<td>28</td>
<td>16</td>
<td>12</td>
<td>10</td>
<td>9</td>
</tr>
<tr>
<td>67 keV $\ { S = S_{\text{EXT}} }$</td>
<td>25</td>
<td>15</td>
<td>12</td>
<td>10</td>
<td>9</td>
</tr>
</tbody>
</table>

The observed excess is deficient by about 29 27 24 22 events (keeping the order used above) in the 1 grain column and by about 7 6 6 5 in the 2 grain column.

There are three possible explanations of this.

1. The $\beta_0$ spectrum departs radically from the expected shape at energies below 6 keV. In this case, since there are $30 \pm 10.3$ non-RaD events in the histogram, the true population of RaD events, is $771 \pm 29$.

2. The $\beta_0$ spectrum does not depart radically from the expected shape but
   (a) the allowance for background events was too great and the source separation was in fact more than averagely efficient, or
   (b) 1 grain events are seriously under-represented in the histogram.
In this case, the true population of RaD events is about \(771 +\) deficiency:

\[
807 \quad 804 \quad 801 \quad 798.
\]

These figures show that but for the allowance made for spurious events, the histogram would be consistent with a \(\beta_0\) spectrum of the allowed shape.

From Table XIV, page 92, the fractions of the \(\beta_0\) spectrum in the 3, 4, 5 grain and 6 - 30 grain regions are seen to be

\[
23.9\% \quad 23.0\% \quad 22.9\% \quad 22.0\%
\]

and

\[
41.9\% \quad 46.2\% \quad 45.1\% \quad 49.4\%
\]

respectively. Using the observed excess in the 3, 4, 5 grain region as base, it is found that the number of \(\beta_0\) events \(>5\) g is

\[
59 \quad 68 \quad 62 \quad 70.
\]

Since there are altogether 693 observed events \(>5\) g, it follows that

\[
634 \quad 625 \quad 631 \quad 623
\]

events are due to disintegrations in which the 47 keV \(\gamma\)-ray is converted.

From these figures the following results are derived. If explanation 1 is correct, the intensity of the \(\beta_0\) spectrum and the intensity of \(\gamma\)-ray conversion are respectively

\[
13.5\% \quad 14.7\% \quad 13.9\% \quad 14.9\%
\]

and
If explanation 2 is correct, these intensities are

\[ 17.5\% \quad 18.3\% \quad 17.1\% \quad 17.9\% \]

and

\[ 79\% \quad 78\% \quad 79\% \quad 78\% . \]

Taking into account both the inherent error in each value and the variance between different values the provisional conclusion is that the intensity of the $\beta_0$ spectrum is $16.2 \pm 4.7$ per 100 disintegrations and the intensity of $\gamma$-ray conversion is $80 \pm 5$ per 100 disintegrations.

**Critical Discussion and Conclusions.**

1. Is it possible that there is some factor other than the $\beta_0$ mode which might give rise to low grain-count events? A cascade de-excitation of the 47 keV state would give low grain-count events if one $\gamma$-ray was converted and the other(s) emitted. But $\sim 6\%$ of singly emitted conversion electrons would imply a much larger total percentage of soft conversion electrons which could not fail to have been detected by $\beta$-spectroscopy. Another possibility is the emission of soft electrons during the RaE $\rightarrow$ RaF disintegration. An examination of the low energy end of the RaE spectrum by Wu et al. (27) showed that there were no detectable discrete electron lines there. There are also no RaE $\gamma$-rays so that the RaE $\rightarrow$ RaF transition
is wholly ground-state to ground-state. However, the theory of Migdal (87), which has been experimentally confirmed for several $\beta$-decay nuclei ($\beta^-, \beta^+$ and K-capture), predicts that the disturbance caused by the charge reorganisation of the atom, results in the ejection of an orbital electron in $\frac{2}{3}$ 2$^{-2}$ cases per disintegration. This is $\sim 2 \times 10^{-4}$ for RaE, a result which has been confirmed by Novey (88, see also 89), and is entirely negligible in the present work.

The charge acceleration during the disintegration of RaE also produces bremsstrahlung (1 photon per disintegration). The photon spectrum has been studied by Novey (90) and by Bolgiano et al. (91). Now, the region of the spectrum of interest here is confined to 2.5 - 8 keV, since below 2.5 keV the photon has insufficient energy to bring up a grain (or at most would bring up only one) while above 8 keV, the photon would almost certainly travel far enough from the event before absorption, for the resultant grain(s) to appear unassociated. (For 8 keV, there is a 50% chance of the photon travelling more than 32 $\mu$ before absorption).

From the spectrum given by Novey (90), the author has estimated that the photon intensity in this energy interval is about $1.8 \times 10^{-3}$ per disintegration. Since the effect (the addition of 1 grain) is only of significance in the 1-5 grain region, the number of events affected is only $(60 \pm 12.6)1.8 \times 10^{-3}$ or about
0.1 event. This is negligible.

2. Is it possible that the "excess" of 1-5 grain events belongs to the low-grain-count tail of the conversion electron peak? Firstly, even if there were no information as to the precise shape of a mono-energetic "line" at low energies, there is no imaginable reason why the tail should not fall smoothly to zero. If the excess belonged to the peak it would constitute a subsidiary peak at 3 grains on its low grain count side. Even if, in the absence of more definite information, the smoothed profile of the main peak were to be extrapolated smoothly towards zero (see fig. 14), no more than half the observed excess would be accounted for. Therefore the minimal conclusion of this work is that the $\beta_0$ transition does occur. However, it seems certain from the observations by Bisgard on a peak centred at 6 grains (see page 79) that even in that case, the tail does not extend in appreciable intensity down to 1 grain. Work by Peat (unpublished) on the line shape of a group centred at 36 grains implies that in the present case the profile would approach zero at 7 grains. It is therefore likely that the main peak makes little contribution to the observed excess.

3. What is the significance of the discrepancy
between the observed excess and the predicted $\beta_0$ intensity at 1.2 grains? Is it possible that the $\beta_0$ spectrum falls radically below the predicted shape in the region (below 6 - 10 keV)? The experimental evidence on spectrum shapes at these energies is extremely sparse, and, except in cases where a gaseous source was used, unreliable. Inseh and Curran (92) using a proportional counter found that there was a gross deficiency below 1 keV in the spectrum of tritium. Waltner and Rogers (93) studied the RaE spectrum at low energies by introducing gaseous RaE (CH$_3$)$_3$ into a cloud chamber, and measuring track ranges. They found that the energy spectrum approaches zero intensity at 10 - 15 keV, and quote work by Madsen (94) in support. Only 131 tracks were measured. Wu et al. (27) investigated the same region by magnetic spectroscopy (thin source mount, Geiger counter detection); their spectrum cuts the $H_0$ axis at $\sim 250$ gauss-cm. ($\sim 6$ keV). They offer no comment on this result. On the other hand, Pniewsky (95) using a nuclear emulsion plate as detector in a $\beta$-spectrometer, found that the RaE energy spectrum cuts the intensity axis at zero energy. Jaffe and Cohen (35), in their measurement of the RaD $\beta_{47}$ spectrum shape do not find the sharp cut-off observed for RaE by Waltner and Rogers, but their results indicate a deficiency of as much as 41% in the intensity below 3 keV. A
tentative conclusion from these results would be that there is an extra-nuclear correction (screening, page 37) which (necessarily) affects the RaD and RaE spectra equally, and also a specifically nuclear effect which affects the RaE spectrum more than the RaD $\beta_{47}$ spectrum. If the latter effect does exist, it would possibly affect the RaD $\beta_0$ spectrum too.

Even if we suppose that only Jaffe and Cohen's maximum deficiency of 41$\%$ (due presumably to screening) affects the $\beta_{47}$ and $\beta_0$ spectra, this would be sufficient to reconcile the predicted and observed results within the limits of error. For, referring to Table XV, page 96, the 1 grain column of the $\beta_{47}$ spectrum would be reduced by $\sim 41\%$, making about 7 events available for the "excess". The expected intensity of the $\beta_0$ spectrum at 1 grain (Table XVI, page 97) would also be reduced, so that a predicted intensity of $\sim 15$ would compare with an observed excess of $8 \pm 9$.

It is also quite possible that too much was subtracted for the contribution of background track "events".

It is not believed that 1 grain events are seriously under-represented in the histogram, relative to the 2, 3, 4, 5 grain events. It is true that, since the author was aware that the final result would turn on the presence or absence of low-grain count
events, he was at particular pains to make sure that such events were genuine, i.e., to make sure that the RaE β track did not in fact continue past the supposed origin cluster after a break. This scrutiny, and the rejection of doubtful cases may conceivably have led to an under-representation of the 1-5 grain events as a group, but not to distortion within the group.

It is likely therefore that there is a deficiency in the 1 grain column of the true β₀ grain-count spectrum. Since this conclusion represents a bias in favour of the first set of values of the β₀ intensity given on pages 74, 75, it is finally concluded that the intensity of the RaD→RaE ground-state-to-ground-state transition is $15 \pm 5$ per 100 disintegrations and that the frequency of emission of conversion electrons of the 47 keV γ-ray is $80 \pm 5$ per 100 disintegrations. The error quoted in the former result is considered adequate to cover uncertainties as to the precise grain-count vs. energy calibration, as well as the uncertainties discussed above.

The latter result compares with the value $85 \pm 5$ found by Wu et al. (27) and the value $74 \pm 5$ found by Cranberg (20) (see page 75). It is definitely inconsistent with the results of other workers, e.g. Bashilov et al. (58 ± 3), and Butt and Brodie (59 ± 5).

The calculated log $f_0 t$ value for the β₀ transition is $7.84 \pm 0.20$. The error embraces the
uncertainty in the maximum energy as well as the uncertainty in the intensity.
Chapter VI.

The positive result of the nuclear emulsion experiment stimulated an examination of possible experimental methods of confirming the existence of the $\beta_0$ transition. The emulsion technique can be seen to suffer from two main disadvantages in the study of $\text{RaD}$. (a) The energy resolution is poor compared with that of other methods of electron spectroscopy. The analysis of the grain-count histogram (Fig. 14 page 68) was made difficult by the fact that, for example, the low grain-count tail of a group of 30 keV electrons is appreciable down to 1 grain (corresponding to 13 keV). (b) It is difficult, in a reasonable time, to amass sufficient observations to give statistical reliability for all parts of the grain count range. For instance, 400 observations of $N$-grain events are required to establish 5% accuracy for the $N$-grain column in the histogram. This means that estimated intensities are subject to a large error, and that it would be almost impossible to examine spectrum shapes. Statistical reliability is hardly a problem in other methods of spectroscopy. Why, then, have other experimental methods failed to detect the $\beta_0$ transition? And is it possible to devise a method which would overcome the reasons for failure?
Magnetic Spectroscopy.

The magnetic spectroscopy of low energy electrons involves, in general, problems of source mounting and electron detection. These may be largely overcome by having a thin uniform source on a backing of a few \( \mu g/cm \) of organic material, and by using a thin windowed Geiger counter as detector. Even if the general problems of low energy spectroscopy could be satisfactorily solved, however, it would be difficult to detect the \( \beta_0 \) spectrum. Fig. 21, which is the electron spectrum obtained by Bashilov (26) illustrates the difficulties. The \( \beta_0 \) spectrum is expected (in momentum dispersion) to have a maximum about mid-way between \( H \rho = 0 \) and the end-point, \( H \rho \sim 870 \), and its intensity (~15 per 100 disintegrations) is somewhat less than that of the M-conversion peak. It is seen that it is "buried" beneath the L and M conversion peaks and the L-Auger group (~45 per 100 disintegrations). In the 400-600 gauss-cm. region, there are L,M conversion electrons "degraded" by back scattering and self-absorption in the source, as well as background electrons scattered from the walls, slit-edges etc. of the apparatus. At the low energy end, there is the M-Auger group (~120%) and the \( \beta_{47} \) spectrum (100%). These are under-represented in Bashilov's spectrum because of detection difficulties.
FIGURE 21
It can be concluded that straightforward magnetic $\beta$-spectroscopy, in spite of the extremely high resolving power attainable, is unlikely to reveal the $\beta_0$ spectrum or even to demonstrate that it exists.

Proportional Counter Spectroscopy.

The only reliable way of studying the RaD disintegration by direct proportional counter spectroscopy is that used by Jaffe and Cohen (35) and by Huster (36). Detailed comment here will be confined to the experiment of Jaffe and Cohen.

They introduced the source into the counter in the form of gaseous lead (RaD) tetramethyl. There was thus none of the problems associated with solid sources. The pulse spectrum from the counter was equivalent to the distribution of the number of ion-pairs formed in the filling gas per disintegration. (It will be noticed here, and in what follows, that the ion-pair distribution is closely analogous to the distribution of grains per disintegration shown in fig. 14.) The dimensions of the proportional counter were such that there was an appreciable chance ($\sim 20\%$) of a conversion electron (a) hitting the wall before expending all of its energy or (b) travelling in the end-regions of reduced multiplication. When corrections (necessarily rather rule-of-thumb) were made for these effects, the amended pulse spectrum was as shown
in fig. 22. Peak A represents those events in which an L-X ray quantum escaped from the counter. Apart from this peak, which was not resolved in the grain-counting method, the pulse spectrum is similar to the histogram of fig. 14 and, since it does not approach zero intensity in the 10-15 volt region, it is susceptible to the same kind of analysis. (Jaffe and Cohen were mainly interested in deriving the shape and end-point of the $\beta_{47}$ spectrum from the shape of the main peak, and did not pursue an analysis of the low energy region). Taking the pulse spectrum at its face value, the author concluded that it was consistent with a $\beta_0$ intensity of 12.5% and a $\gamma$-ray ($\beta_{47} + \gamma$) intensity of 6.5%. The pulse spectrum calculated on this basis is shown as a dotted line. In a letter to the author, A.A. Jaffe gave his opinion that the uncertainties of the wall and end corrections prevented any definite conclusion about the $\beta_0$ spectrum but he did "think it unlikely for the true spectrum to be zero between 18 to 25 keV".

It would be of the greatest interest to have Jaffe and Cohen's experiment repeated under conditions which would eliminate or greatly reduce the uncertainties. The requirements are:

1. a long counter, with field-adjusting tubes at each end, to eliminate end-effect.

2. high pressure xenon or argon filling to
reduce electron ranges.

(3) a solenoid coil around the whole length of the tube. The magnetic field would "curl up" the electron tracks and cut down the wall-effect.

Since it appeared that there were difficulties in the way of buying or preparing active lead tetramethyl (even the inactive form is extremely poisonous), attention was turned to the consideration of other methods.

Liquid Phosphors:

A solution of p-terphenyl in xylene (～2 gm./litre) acts as a phosphor - that is, part of the energy lost by a charged particle in passing through it is transferred to molecules which de-excite by emitting quanta in the visible or near-visible region of the spectrum. The quantity of light emitted, is proportional to the total energy lost in the phosphor. It was proposed to dissolve a salt of RaD in a xylene-terphenyl solution and measure the quantity of light produced per disintegration by means of a photo-multiplier. The photo-multiplier pulse spectrum would again have been analogous to the grain count spectrum and the "wall" effect could have been made negligible.

Xylene is not the best of solvents but after a long series of trials with ThB (Pb^{212}) it was found that the myristate, Pb \( \cdot (C_{14}H_{27}O_2)_2 \), was soluble.
III.

However, a further study of the rather sparse data on the energy resolving power of the liquid phosphor led to the conclusion that it would be inadequate for the purpose. This was later confirmed by the experiment of Lewis (44) who added a trace of RaD to molten sodium iodide and used the resulting crystal in the manner described above. NaI is the most efficient of the phosphors — its light output per keV is about four times that of the xylene-terphenyl phosphor and its resolution, which depends on the statistical straggle of the numbers of quanta produced, is correspondingly enhanced. Yet, Lewis could draw no definite conclusions as to the occurrence of the \( \beta_0 \) transition. (Not that this was his intention — he was mainly interested in the lifetime of the 47 keV excited state.)

**Anti-coincidence Methods.**

Consideration was then given to evolving an arrangement which would discriminate in some way between \( \beta_{47} - \gamma, e_{LMN} \) disintegrations and \( \beta_0 \) disintegrations. The methods described above have depended on energy discrimination — disintegrations with a true total electronic energy emission of 20 — 30 keV could only be \( \beta_0 \) disintegrations. There is another factor which lends itself to discrimination. The \( \beta_0 \) transition involves (disregarding the
neutrino) a single particle emission whereas the 47 mode has at least 2 particles ($\beta^{47}$, $\gamma$) and may have as many as 6 ($\beta^{47}$, $e_L$, $A_L$, 3 $A_M$ + others < 2 keV).

If the radiations from RaD are allowed to fall on two detectors, A and B, and A counts only when there is no excitation of B, it is clear that A will discriminate in favour of $\beta_0$ particles, for if any other sort of particle passes through A, there is a chance that one or more of its companion particles will trigger B and so paralyse A. The greater the angle of acceptance of B, the greater will be the degree of discrimination.

We will consider the case where the total solid angle about the source is covered by either A or B. Let $4\pi \omega$ be the angle covered by A and $4\pi (1 - \omega)$ that covered by B. The RaD disintegrations are classified as follows. The intensities are given roughly.

<table>
<thead>
<tr>
<th>Case</th>
<th>Reaction</th>
<th>Intensity</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>$\beta^{47} + \gamma$</td>
<td>3%</td>
</tr>
<tr>
<td>(b)</td>
<td>$\beta^{47} + e_L + X_L + A_{MNO}$</td>
<td>19%</td>
</tr>
<tr>
<td>(c)</td>
<td>$\beta^{47} + e_L + X_L + X_{MNO}$</td>
<td>19%</td>
</tr>
<tr>
<td>(d)</td>
<td>$\beta^{47} + e_L + A_L + 2A_{MNO}$</td>
<td>43%</td>
</tr>
<tr>
<td>(e)</td>
<td>$\beta^{47} + e_L + A_L + 3A_{MNO}$</td>
<td>15%</td>
</tr>
<tr>
<td>(f)</td>
<td>$\beta^{47} + e_{MNO} + A_{MNO}$</td>
<td>20%</td>
</tr>
<tr>
<td>(g)</td>
<td>$\beta_0$</td>
<td>15%</td>
</tr>
</tbody>
</table>
Let the intensity of each group be denoted by, for example, $I(f)$, and let the efficiency of detector $B$ for the detection of each kind of particle be denoted by, for example, $E_B(\beta_{47})$.

We wish to find how successful the arrangement is in eliminating all but $\beta_0$ counts from detector $A$, which is in anti-coincidence with $B$. The rate of $\beta_0$ counting is proportional to $I(g)\omega$. Since, in considering any particular arrangement, it is important to know how the degree of discrimination depends on $\omega$, the non-$\beta_0$ anticoincident pulses at $A$ will be called 1st order, 2nd order, 3rd order, according as their intensities are proportional to $\omega$, $\omega^2$, $\omega^3$.

The calculation will be illustrated by considering only group $(f)$ say. For all arrangements $E_B(e_{LMNO})=1$.

The intensity of 1st order non-$\beta_0$ pulses from group $(f)$ is the multiple of the independent probabilities that the disintegration is of type $(f)$, that $e_{LMNO}$ goes into angle $\theta$, that $\beta_{47}$ and $A_{LMNO}$ go into angle $(1-\omega)$ and that $\beta_{47}$ and $A_{LMNO}$ are both undetected by detector $B$. That is, the intensity is proportional to

$$I(f)\omega (1-\omega)^2 \left\{1 - E_B(\beta_{47})\right\} \left\{1 - E_B(A_{LMNO})\right\}.$$ 

It will appear later that $\omega$ must be small, so that $1 - \omega \approx 1$. Because $E_B(e_{LMNO})=1$, it is impossible for $\beta_{47}$ or $A_{LMNO}$ to give a 1st order pulse at $A$. 
The intensity of 2nd order non-$\beta_0$ pulses from group $(f)$ involves the probability that $e_{\text{MNO}}$ and another particle go to detector $A$. It is thus proportional to

$$I(f) \omega^2(1 - \omega) \left\{ 1 - E_B(\beta_{47}) \right\}$$

$$+ I(f) \omega^2(1 - \omega) \left\{ 1 - E_B(A_{\text{MNO}}) \right\}.$$

The 3rd order intensity is proportional to $I(f)\omega^3$. It is to avoid high-order non-$\beta_0$ pulses that $\omega$ must be small.

By adding up contributions made by all the groups, the total intensity and spectrum of the non-$\beta_0$ pulses can be found. The merits of any proposed experimental arrangement can then be assessed when plausible values for the detection efficiencies are assumed.

Consideration of Experimental Arrangements.

In this section, a few experimental arrangements will be examined, involving different combinations of the various detectors.

Magnetic Spectrometer - Scintillation Counter.

In a spectrometer of suitable construction, it might be possible to replace the source mounting by a plastic phosphor. The phosphor would have a hole drilled in it, say 2 cm. deep and 1.5 cm. in diameter, and a $\text{Ra}(D + E)$ source would be evaporated from a
neutral solution on to the bottom surface of the hole. The phosphor would then act as detector B with an angle of acceptance $4\pi(1 - \omega) = 4\pi \times 0.035$. A photomultiplier, preferably in direct contact with the phosphor, would be in anti-coincidence with a thin-windowed Geiger counter acting as receiver for the spectrometer. Calculation then shows that the non-$\beta_0$ part of the received spectrum would be only 6% in intensity (of $\beta_0 \sim 15$%), that it would consist mainly of conversion electrons, which can be resolved sharply, and that only 0.15% would be L-Auger electrons. (The assumptions used here will be explained in the next section.) There seems little doubt that the $\beta_0$ spectrum could be detected under these conditions. An allowance, if necessary, for the presence of scattered or "degraded" conversion electrons could be made by comparing the anti-coincidence spectrum with the spectrum obtained when the photomultiplier was switched off. To minimise the effect of the magnetic field on the gain of the photomultiplier, the spectrometer would preferably be of the constant-field type (or of the electrostatic type used by Kobayashi).

**Two Scintillation Counters.**

A possible arrangement is that shown schematically in fig. 23. Detector B is of sodium iodide, anthracene or plastic. Detector A is of anthracene; since this
is less likely to reflect electrons than NaI, and has better resolution than plastic. Both phosphors are mounted in a vacuum-tight envelope; the reflector cuts down the probability of light from one phosphor entering the photo-multiplier of the other. The efficiency of detector B in detecting a particle of given energy is given by (96)

\[ E = 1 - e^{-N \alpha_p} \]

where \( N \) is the average number of photons produced by the particle, \( \alpha \) the fraction of these reaching the light-sensitive cathode of the photo-multiplier and \( \alpha_p \) the number of photo-electrons produced per photon \( (\sim 0.09) \). If the circuit associated with B is biased to cut out most of the thermal noise from the photo-multiplier (this is desirable but not absolutely necessary - a full discussion here is not warranted) the formula becomes

\[ E = (1 - e^{-N \alpha_p}) e^{-N \alpha_p}. \]

Assuming a hole depth of 2 cm. and a diameter of 1.5 cm. (for NaI, only this ratio is necessary down to any convenient size; for plastic, the detection of L-X rays requires these absolute dimensions at least) the detection efficiencies of detector B are calculated to be
### Calculations of the type described on page 113

Then show that, for NaI the intensity of non-$\beta_0$ pulses at detector A would be $2.7\%$ (noise rejected) or $1.0\%$ (noise accepted) and that all would of a size corresponding to $\geq 40$ keV. For plastic, the intensity would be $\sim 6.0\%$, all pulses corresponding to $\geq 30$ keV. (Cf. $\beta_0$ intensity $\sim 15\%$, mostly below 25 keV).

If no precautions are taken, there is a chance that a true anti-coincident pulse at A is annulled through a photon produced in A travelling back along the electron path and striking the photo-cathode of B.

The probability of this happening is

$$\frac{N}{4\pi} \cdot \frac{1}{\mu^2} \cdot \frac{\pi r^2}{D^2} \cdot p,$$

where $N$ is the number of photons produced in detector A; $\mu$ is the refractive index of phosphor A; $r$ and $D$ are as in fig. 23; $p$ has been defined above. For a 2 mm. hole in NaI, this probability is 2.5% and is negligible. With a hole of more convenient size, it
is intolerably large. The effect can only be overcome
by covering phosphor A with an opaque film of collodion-
aluminium of 25-50 \( \mu g/cm.2 \) thickness.

If such a film had to be used it would prevent
electrons of energy \( \sim 5 - 7 \) keV reaching A and would
make less certain the analysis of the pulse spectrum.
Since, moreover, there are uncertainties about the
source-absorption of K-Auger electrons, and about the
extent to which NaI reflects electrons (the available
information on this is discordant), it was thought
desirable to pursue the enquiry into alternative
arrangements further to see if one could be found for
which the prospects of success were more certain.

Proportional Counters.

It was realised that it would be most desirable to
have a proportional counter as detector A, since its
resolving power is very much better than that of the
best scintillator. At about 47 keV, the resolution of
a proportional counter (half-width of "line" at half-
height) is \( \sim 1.7 \) keV, compared with \( \sim 5.8 \) keV for
sodium iodide.

In order to consider the design of arrangements
involving proportional counters, it is necessary to
know the ranges of electrons in various gases. The
experimental data available for electrons of 0-50 keV
are very inadequate and unreliable. The theoretical
formulae depend on assuming a value for the "mean excitation potential", I, a quantity which is little understood (97). Tsien (98) measured the ranges of photo-electrons of 2-50 keV in an air-filled cloud chamber. His values agree with those predicted by the Bethe non-relativistic formula

\[ \frac{dE}{dx} = \frac{4\pi N a^2 N_z}{m v^2} \log_e \frac{0.58 mv^2}{I} \]

On the assumption that the formula holds for argon and helium, and that \( I \approx 11.5 z \), the ranges shown in Table XVII have been calculated for normal pressure and temperature.

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>2</th>
<th>6</th>
<th>10</th>
<th>11</th>
<th>26</th>
<th>34</th>
<th>42</th>
<th>60</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ranges (cm.)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Argon</td>
<td>0.02</td>
<td>0.09</td>
<td>0.22</td>
<td>0.58</td>
<td>1.10</td>
<td>1.74</td>
<td>2.52</td>
<td>5.1</td>
</tr>
<tr>
<td>Helium</td>
<td>0.07</td>
<td>0.47</td>
<td>1.17</td>
<td>3.4</td>
<td>6.6</td>
<td>10.7</td>
<td>15.5</td>
<td>31.5</td>
</tr>
</tbody>
</table>

The ranges for argon are in agreement with the four known experimental results (99). Those calculated for helium are roughly in agreement with the two known experimental results for hydrogen (99), which is to be expected from the theory. The only known
observations on ranges in helium do not go below 80 keV (100).

The tabulated ranges are smaller, by a factor of 2 or 3, than those predicted by the relativistic formula tabulated by Aron, Hoffman and Williams (97). This formula, however, behaves oddly at low energies (for $Z = 8$, $dE/dx$ has a maximum at $\sim 5$ keV, of experiment, $\sim 0.1$ keV) and disagrees with the observations of Tsien.

Several of the arrangements which have been considered will now be described.

(1) Fig. 24 (a). The principle is the same as in the 2-phosphor arrangement already described. Detector A is a long proportional counter; detector B is a Geiger counter; the gas filling is common to both. The source is mounted on a thin film supported on the end of a wire "cage" whose dimensions define the value of the solid angle $4\pi \omega$. As before, counter A is in anti-coincidence with counter B. The arrangement works as follows. If a $\beta_0$ particle is emitted into the proportional counter, the ionisation it produces inside the cage does not trigger the Geiger counter, because the cage is at uniform potential. The $\beta_0$ particle is therefore recorded by A. But if, for example, an L Auger electron is emitted into the proportional counter, there is a chance that one or more of its companion particles will either move to the
FIGURE 24
left, or travel rightwards through the film and through
the wall of the wire cage. In either case, the Geiger
counter is triggered and the $A_L$ pulse is not recorded.

For a cage 2 cm. deep and 0.5 cm. in diameter, it
has been estimated that the efficiency of detection of
the various RaD particles by the Geiger counter is

<table>
<thead>
<tr>
<th>Particle</th>
<th>$\beta_{47}$</th>
<th>$e_{LMN}$</th>
<th>$X_L$</th>
<th>$A_L$</th>
<th>$X_M$</th>
<th>$A_M$</th>
<th>$\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Efficiency %</td>
<td>45</td>
<td>98</td>
<td>0</td>
<td>80</td>
<td>0</td>
<td>45</td>
<td>0</td>
</tr>
</tbody>
</table>

This assumes that $\beta_{47}$ and $A_M$ are detected only if
they move leftwards.

Calculation then shows that the intensity of
non-$\beta_0$ pulses would be $\sim 10.5\%$ (of $\beta_0 \sim 15\%$) made
up of $4.5\% e_L$ pulses, $2.0\% e_{MNO}$ pulses, $0.5\%$ double
pulses $\geq 30$ keV, and $3.5\%$ due to the $\beta_{47}$ spectrum.

Detailed consideration of the arrangement shows
that there are many difficulties.

(a) It is essential to use the proportional
counter end-on (see (b) below). To avoid distorting
the pulse spectrum through the formation of ions in the
ill-defined end-region of variable multiplication, it
would be necessary to have field-adjusting tubes (101).
These would define a sensitive region of constant
multiplication.

(b) Since ions formed in the space between the
source and the end of the field tubes (about 3 cm. say)
would not contribute to the pulse produced by A, it would be necessary to have track lengths large compared with 3 cm. in order to make this loss negligible. Table XVII shows that hydrogen at a pressure of 1/10th of an atmosphere would give tracks of about 12 cm. for 10 keV electrons, and it shows further, that these electrons would lose only 1.4 keV in the first 3 cm. The absolute energy loss is less at higher energies—e.g. 0.5 keV at 35 keV.

To accommodate the electrons of highest energy, the counter would have to be 300 - 350 cm. long. This length is somewhat unusual but raises no great problems apart from the fact that for a given current-pulse, the voltage-pulse is inversely proportional to the capacity of the counter and its lead to the first grid of the amplifier. On the other hand, the working voltage would be conveniently small (order of 1,000 volts). The degree of scattering of electrons by the hydrogen filling is expected to be very small.

(c) Since the high voltage is common to both counters, the Geiger counter would probably not be at its optimum voltage. This could be overcome by having an additional voltage supplied to the Geiger counter wire.

(d) Electrons leaving the cage at an angle to the axis would, if undeflected, hit the wall of the proportional counter. This could be prevented by
having a solenoid along the length of the counter. The trajectories of the fastest electrons (\( \sim 62 \text{ keV} \)), emitted at the largest possible angle to the axis, would be constrained to a spiral of 2 cm. diameter by an axial field of 200 gauss. In order to ensure that not even highly scattered electrons would reach the wall, the counter diameter would need to be about 10 cm.

(e) The presence of the magnetic field would affect (but not grossly) the ideal working of the cage (for \( A_L, H \phi \sim 300 \text{ gauss-cm.} \)). This effect could be cut down by making the Geiger cathode of iron or steel.

(f) In the arrangements previously discussed, the presence of RaE in the source was of no importance. In this case, however, the fast, lightly ionising RaE \( \beta \)-particles emitted into the proportional counter would pass through, and produce small pulses corresponding to several keV. It would therefore be essential to do the experiment soon after the preparation of the pure RaE source, and to do monitor experiments with pure RaE. The source would be replaced by removing the Geiger counter case.

(g) Such a large counter would have a large number of background pulses due to cosmic rays. Most of them, however, would be very small (corresponding to \( \sim 1 \text{ keV} \)).

(h) The geometry of the Geiger counter is unconventional and it would require an unconventional
anode, in the form of a spiral around the cage. Such a counter has been used by C.A. Barnes (102). A good plateau would not be necessary.

(2) Fig. 24(b). This arrangement is two proportional counters in one, the two anode wires being separated by a glass bead. It is similar to one described by Insch and Curran (103). The source (on a film mounted on a wire frame) is supported on a metal "probe" held at the potential appropriate to its position in the electric field (104). A solenoid would again be necessary, but the required magnetic field would be very much greater (\( \sim 1,000 \) gauss) than in arrangement (1), since electrons travelling at all angles to the axis are accepted. In this case, as in the next, \( \psi = 0.5 \). The disadvantages of this method are the distortion of the field around the bead and the practical difficulty of finding and maintaining the voltage at which the "probe" must be held.

(3) Fig. 24(c). Detectors A and B are two proportional counters side by side, with the source supported between them. The gas filling is common to both. The acceptance angle of detector A is 2 \( \psi = 0.5 \).

Since finally, it was decided to perform an experiment using this arrangement, it will be considered in some detail, and its comparative merits will be discussed later.
Suppose the source is deposited in a thin uniform layer on an aluminium-collodion film of thickness \( \sim 10 \mu \text{g/cm}^2 \), and that it is on the side facing detector A. Then, using the data on the transmission of films given by Lane and Zaffarano (104), it is estimated that \( E_B (A_L) = 75\% \) and \( E_B (e_{LMNO}) = 97\% \). Roughly, it can be seen that the main function of the arrangement is to remove Auger electron pulses from the pulse spectrum of A. For if an Auger electron from the source enters A, its associated conversion electron must (neglecting small corrections) either enter A or enter B. In the former case the resultant pulse corresponds at least to the energy of \( (A_L + e_{LMNO}) \); in the latter case, there is no pulse. Therefore there is a region, corresponding to 15-25 keV, where the pulse spectrum should be susceptible to analysis to demonstrate the existence of the \( \beta_0 \) spectrum.

More detailed consideration shows that apart from the \( \beta_47 \) spectrum, which appears in full intensity, nearly all of the non-\( \beta_0 \) pulses form a group (intensity 64\%) fairly evenly distributed between 30 keV and 49 keV. The intensity of L-Auger electron pulses is 0.79\% (of \( \beta_0 \sim 15\% \)). Putting \( E_B (A_L) = 0 \) does not make much difference; it increases the intensity of pulses in the region 30 - 37 keV, from 27\% to 31\%.

In order to keep the electrons from hitting the wall of counter A, the apparatus must be put between
126.

the poles of an electromagnet.

(4) This method uses the anti-coincidence technique merely to eliminate the wall effect in an experiment of the type performed by Jaffe and Cohen. It also eliminates all cosmic ray background and so would be useful in all cases where large counters are necessary.

In fig. 25, A is an outer brass cylinder, and B represents 2 perspex formers held rigidly apart by brass rods and bolts C. A central anode wire is supported by the field- and guard-tube assembly D. The thick wires E (\( \sim 1 \text{ mm. diameter} \)), strung between the formers, form an open cylinder, and around the central wire. A still larger cylinder is formed by a thin wire (\( \sim 0.1 \text{ mm. diameter} \)) threaded back and forth between the teeth F. The cylinder A is sealed at each end by bolted end-plates and gaskets.

Cylinders A and B are at high voltage and act as cathodes. The central wire acts as anode to the central proportional counter, and the fine wires F as anode to the outer counter which merely acts as a detector. That the effect of cylinder B approximates to the effect of a continuous cylinder is confirmed by the experience of workers in the Glasgow University Physics Department. The working of the outer detector is not in doubt (e.g. see ref. 102). The anode leads are brought out through insulators (and a guard-tube
in the case of the central wire) in the end-plates.

The working of the arrangement, when RaD tetraethyl is introduced, is apparent. When the inner counter is in anti-coincidence with the outer detector, only particles whose trajectories lie entirely within the inner counter produce acceptable pulses. Thus pulses due to cosmic rays, and to electrons which would normally be absorbed in the "walls", are eliminated. Pulses from electrons which pass into the insensitive region at the ends could be made a negligible fraction of all by using a long counter. Some electrons, of course, would still hit the wires. But, under the conditions of Jaffe and Cohen's experiment, the fraction of pulses distorted would be reduced from 20% to 0.8%. The spectrum of distorted pulses produced by electrons which pass through the "wall" could be found by putting the inner counter in coincidence with the outer detector.

It should be noted that this rejection of distorted pulses does not affect all kinds of disintegrations equally. Events with MND conversion electrons would be rejected more frequently than those with L conversion electrons, since the probability of an electron passing through the "wall" depends on its track length. However, this would not greatly affect the experiment. The point is that potentially large pulses would be prevented from being thrown down by the
wall effect into that small-pulse region which is crucial for the detection of the $\beta_0$ spectrum. In fact, the action of this arrangement is to apply in a reliable experimental way exactly the same sort of correction that Jaffe and Cohen applied in an unreliable rule-of-thumb way.

This method, although attractive, was not considered further, because, as has been mentioned, it would have been difficult to obtain or prepare the RaD tetramethyl.

**Choice of Method.**

There are two levels of ambition in the investigation of the $\beta_0$ spectrum. The lower is merely to confirm the results of the nuclear emulsion experiment, i.e., to show conclusively that the $\beta_0$ transition does occur, and to estimate its intensity. The higher is to make, in addition, reliable observations on the shape of the spectrum. As will be seen in Chapter VII, this would be a valuable contribution to the experimental data required for the elucidation of $\beta$-decay theory.

Of the methods considered above, only the scintillation-counter-magnetic-spectrometer combination (page 114) gives promise of being able to measure the shape with sufficient accuracy. Perhaps a $\beta$-spectrometer would have to be specially designed for
such an experiment.

The possibility of using the Geiger counter—proportional counter arrangement (page 120) for shape measurement is more uncertain. The non-$\beta_0$ pulses would be less well resolved. The correction, especially at low energies, for the loss of ions in the insensitive region would be reliable only if $dE/dx$ were known accurately over the range $E = 0 - 65$ keV. The slight distortion, again at low energies, introduced by the action of the magnetic field on the effective geometry of the cage, while tolerable in estimating the total intensity of the $\beta_0$ spectrum, would be prejudicial to the accuracy required for shape measurement.

The choice of method therefore became a matter of considering the effectiveness of methods (1) and (3) (pages 120, 124) in merely detecting the $\beta_0$ transition and measuring its intensity. Nor is great accuracy in the latter measurement particularly desirable from the theoretical point of view, since the $ft$-value is insensitive to variations of the order of 20% in the intensity.

Method (3) was therefore chosen, since it is less complicated, requires counters of reasonable size, and seems capable of giving the required confirmation of the nuclear emulsion results. The experiment is effectively comparable, in this regard, with that of Jaffe and Cohen, with the crucial difference that there
Preparation of the Experiment.

Apparatus corresponding to arrangement (3) has been built and is at the stage where trials with RaD sources could be carried out. This has not yet been done.

The preparation of the experiment will be briefly described.

Design of the Apparatus.

Gas filling. It is desired to cut down scattering as much as possible so that almost no electrons are turned back to hit the source support or the wall. Since the degree of scattering is directly dependent on Z, it follows that the lightest gases, hydrogen or helium, are preferable. It is, of course, necessary to add a few per cent of methane or some other organic gas, to stabilise the multiplication.

Dimensions. The expected ranges of 42 keV and 60 keV electrons in hydrogen or helium at 760 mm. pressure are about 16 cm. and 32 cm. respectively. (Ranges in hydrogen are actually somewhat shorter than in helium due to the fact that its "mean excitation potential" I is smaller by a factor of $\sqrt{2}$ while NZ is the same for both.) A pressure of 2.0 - 2.5 atmospheres is therefore sufficient when the diameter of proportional
counter A is about 15 cm. This value for the diameter represents a rule-of-thumb compromise.

Smaller diameter \(\rightarrow\) higher pressure \(\rightarrow\) higher working voltage and heavier construction. Larger diameter \(\rightarrow\) larger cosmic ray background rate. The cathode used was a brass tube, of 14.5 cm. diameter, which happened to be available.

Since the dimensions of counter B, which has only to act as a detector, are largely arbitrary, a diameter of 7.5 cm. was chosen to permit the whole apparatus to fit between the poles of the available electro-magnet. For a given voltage, the multiplication in the smaller counter is larger than in the bigger, but this merely makes it more efficient as a detector. There is no need for it to work in the truly proportional region.

The length of the apparatus, 35 cm., was chosen as the smallest length likely to give uniform multiplication over the central region (\(\sim 4-5\) cm.) to which electrons are confined by the magnetic field.

The anode wires are of tungsten, 0.004 in. (\(\sim 0.01\) cm.) in diameter.

**Construction.** The design is shown in figs. 26 and 27. Fig. 28 is a photograph of the apparatus.

The cathodes, cut from brass cylinders, are soldered together, and soldered into grooves in the rectangular brass end plates. The source holder
FIGURE 27
(fig. 26(a)) is a grooved brass frame which slides along the "rails" formed by the junctions of the two cathodes. The frame is the same length as the counters and is soldered to a cover plate which can be screwed down, on an O-ring against the end-plate. The source support is a rectangle of 0.3 mm. thick aluminium held on to the frame by strips of spring brass. The sputtered formvar source-backing is stretched over a circular hole in the aluminium plate.

Each anode assembly consists of an earthed brass "guard-tube" and three ebonite insulators. The guard-tube ensures that in the event of momentary breakdown of the outer insulator, the current pulse flows to earth, and not to the wire. The guard-tube is waxed into the outer insulator. The other two joints are effectively sealed by O-rings. The small ebonite plug has a fine axial hole which centres the wire. The wire is threaded through a hole in the third insulator (the hole is waxed over) and anchored to a small brass bolt screwed into the insulator.

In order to give access to the inside for cleaning, there is a large brass plug, 12 cm. in diameter, coaxial with the large counter, screwed into one of the end plates.

At first the apparatus was soft-soldered, but after much trouble with leaks in the solder, it was eventually hard-soldered. The inner surfaces were
cleaned well, first with "Silvo" and then with acetone. Cotton hairs, etc., were blown out with a blast of compressed air.

Two ½ in. holes in the larger cathode, intended for the admission of X-rays for calibration, were covered with an aluminium "window" 0.01 cm. thick, in the following way. The brass was heated with a blow-lamp, some yellow wax put on, and the aluminium sheet (which had previously been covered with a very thin film of wax on a hot plate) pressed down until the wax set.

Each wire was inserted in the following way. A piece of wire was cut, about four times the length of the counter. One end was passed through the counter, the anode assemblies were threaded on piece by piece, finally cleaned with acetone, and screwed into the end plates. There was then a length of wire outside the counter which had received no handling at all in the above process. This length was inspected to make sure there were no kinks in it, cleaned with acetone and then pulled through so that it was inside the counter. The ends were then anchored to the bolts and the central holes waxed over with the aid of a hot glass rod.

**Filling.** To test the counter, it was filled with 8 cm. methane and 70 cm. argon.
The filling system was of glass and rubber, with the necessary taps and clips, mercury manometer, and liquid nitrogen vapour trap. The gas cylinders were equipped with the usual regulating valve (one is seen in the photograph).

If one pumps out a system connected to such a regulating valve, there is a danger that air will be drawn in through leaks in the screwed connections. (Oxygen is fatal to a proportional counter.) This danger was avoided by proceeding in the following (so far as is known, original) way. Taps A and B (fig. 29) were closed and the regulators opened until the gauges read 1.5 atmospheres. Taps A and B were then opened in turn, gas was allowed to flow out for several seconds, and the taps closed. The regulators and lead-tubes from the cylinders were then oxygen-free and at pressure, so that no inward leak could occur (provided that there is no excessive drain, the regulator keeps the pressure at its pre-set value).

The system, including the counter, was then evacuated, and the pump cut off by closing tap C. The counter was filled with argon, (by slightly opening tap A) and again evacuated, the cycle being repeated several times to flush out traces of air from the counter. Finally the counter was filled with the required amounts of methane and argon.

Testing. The electronic equipment consisted of two
nitro3e, selfIe

FIGURE 29
pulse amplifiers, one for each counter, a single-channel pulse analyser to follow the amplifier of counter A, an anti-coincidence unit (A.E.R.E. type 1036C), and a scaler. The E.H.T., up to 4,000 volts, was supplied by a type 200 power unit.

The working of the electronic equipment was checked by means of a double pulse-generator with variable attenuation. The linearity of the amplifiers was checked, and the adjustable delay times and resolving times of the anti-coincidence unit checked against the variable delay time between two pulses from the generator. After a few minor adjustments, the apparatus was found to be in order.

The proportional counter was tested, in a preliminary way, with γ-rays from a Co\(^{60}\) source. The performance seemed to be satisfactory, with no appreciable deterioration over a fortnight after each filling. With the amplifier of A at full gain, pulses became visible (on a C.R.O. screen - about 3 volts per mm deflection) for a counter voltage of 1,000 volts. At 2,800 volts, the amplified pulses reached 70 volts, the maximum output-pulse voltage of the amplifier. At higher multiplication (higher E.H.T.), the amplifier overloads, the amplification becomes non-linear, the tail of the amplified pulse overshoots, and the negative-going overshoot pulse upsets the bias of the pulse analyser.
For Co$^{60}$ $\gamma$-rays, both counters worked satisfactorily at a common voltage of 3,000 volts, when the gains of the amplifiers of the large and small counter were attenuated by 16 db. and 36 db. respectively. These optimum values would not necessarily be best for a RaD source. The optimum combination can be found rapidly with a double beam oscilloscope.

The resolution of the large counter could not be tested since sources emitting suitable $\gamma$-rays or X-rays (10 - 70 keV) were not available up to the time of writing. Attempts to use characteristic X-rays excited by radiation from an X-ray tube failed because of pick-up from the tube.

Conclusions.

It is hoped that the ideas outlined in this chapter, and the preparation of the experiment, will be a stimulus to further work on the subject.

There is no doubt that, given the RaD tetramethyl, some method such as method (4) (page 126) would be preferable to the one described above.

Again, the apparatus of method (4) could be adapted to work in a way identical to the working of the apparatus constructed. There would then be no cosmic ray background, and a guarantee of minimum wall effects. Also a quasi-permanent Ra(D + E) source could be used, since the great majority of the RaE
electron pulses would be cut out by the outer detector. As it is, $\beta$-particles from RaE growing in the source will give small pulses, corresponding to energies 0 - 15 keV, and the source will need to be changed for each run of observations.
Chapter VII.

Discussion of Theory.

The shell model theory of the nucleus is based on the assumption that any one nucleon can be considered to be moving in a static potential field produced by all the other nucleons (105). This assumption is particularly plausible when the nucleon considered is the only one, or one of two, outside closed shells. So in the disintegration chain

$$\text{RaD}_{82}^{126+2} \rightarrow \text{RaE}_{82+1}^{126+1} \rightarrow \text{RaF}_{82+2}^{126}$$

where 82 and 126 are respectively the proton and neutron "magic numbers", one expects the shell model to be particularly reliable, and this chapter will consist of an examination of the implications of this model.

The level scheme given by Klinkenberg (106) suggests that the proton(s) are in states $6h_{1/2}$, $5f_{7/2}$ or $5f_{5/2}$ (in probable ascending order of energy) and that the neutron(s) are in states $6g_{9/2}$, $7i_{11/2}$ or $5d_{5/2}$. The letter gives the orbital angular momentum $\ell$ and the suffix the total angular momentum $j$ of the state. Proceeding by analogy with the successful use of the theory applied to atomic electron states, it
was until recently assumed that each "configuration" consisted of a combination of pure states for each particle.

For instance, Lee-Whiting (107), fitting the predictions of the possible RaE configurations to the observed shape correction factor, the observed ground state spin (33) and the observed, extremely small, nuclear magnetic moment, concluded that the configuration of the ground state was \((h_{1/2}; 1\mu/2)_1\). The order here is (proton; neutron). If it were true that the configuration for each state is pure, it would be possible for a \(\beta\)-transition between given states to be virtually prohibited even in cases where the spin and parity changes indicate an allowed \((\Delta I = 0,1; \text{no})\) or 1st forbidden \((\Delta I = 0,1,2; \text{yes})\) transition; for the configurations of initial and final states might be such that a single particle jump would be ruled out. For example, Lee-Whiting thought that the RaD ground state might be pure \((\;g_{1/2})_0\). The RaD \(\beta^\circ\) transition would then involve a simultaneous 2-particle transition

\[
\begin{align*}
\text{neutron } g_{1/2} &\quad \text{proton } h_{1/2} \\
\text{neutron } g_{1/2} &\quad \text{neutron } 1\mu/2 
\end{align*}
\]

and would be of zero or, at any rate, unobservable intensity.

However, the atomic and nuclear cases are very
different. In the atomic case, the orbital electrons move in a static field produced by a compact nucleus. The perturbing effects of other electrons is comparatively small. In the case of the nucleus, the interactions between the nucleons which are outside closed shells produce appreciable perturbations in the state of any one nucleon, so that it becomes likely that any given state is a mixture of configurations.

Elliot and Flowers (108) in 1955 performed calculations on configuration mixing for nuclei beyond the $0^+_2$ doubly closed shell.

Bayman has since performed analogous calculations for RaD, RaS and RaF and he has kindly made his results available to the author before publication.

Using the available data for Pb $^{126+1}_2$ and Bi $^{124+1}_2$, he has calculated the interaction energy between each outer particle and the 82,126 closed shell. Then, following Pryce (109), he assumed that the interaction between the two outer particles was a contact interaction, with the singlet force strength (Pryce, page 276) 1.5 times the triplet force strength. To facilitate calculation, he used harmonic oscillator wave functions for the particle states. Square well wave functions would have been more plausible for these heavy nuclei, but the labour would have been greater and the results not greatly different. There remained one free parameter, $V_0$ in the Flowers notation, which
is a measure of the absolute strength of the interaction between the two nucleons outside the doubly closed shell. Bayman believes this to be between 30 and 50 MeV. His results for the configuration mixing, using these two values of \( V_0 \), are given in Table XVIII. The percentage contribution is the square of the calculated amplitude.

### Table XVIII.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Configuration</th>
<th>( V_0 = 30 \text{ MeV} )</th>
<th>( V_0 = 50 \text{ MeV} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>RaD</td>
<td>( 1; g_{1/2} )</td>
<td>0.93, 87%</td>
<td>0.87, 76%</td>
</tr>
<tr>
<td></td>
<td>( 1; i_{1/2} )</td>
<td>0.36, 13%</td>
<td>0.49, 24%</td>
</tr>
<tr>
<td>RaE</td>
<td>(h_{1/2}; 1; i_{1/2} )</td>
<td>0.25, 6.5%</td>
<td>0.88, 77%</td>
</tr>
<tr>
<td></td>
<td>(h_{1/2}; g_{1/2})</td>
<td>0.96, 93%</td>
<td>-0.44, 19%</td>
</tr>
<tr>
<td></td>
<td>(f_{1/2}; g_{1/2})</td>
<td>0.07, 0.5%</td>
<td>-0.19, 1%</td>
</tr>
<tr>
<td>RaF</td>
<td>(h_{3/2}; )</td>
<td>0.97, 93%</td>
<td>0.93, 87%</td>
</tr>
<tr>
<td></td>
<td>(f_{1/2}; )</td>
<td>0.26, 7%</td>
<td>0.36, 13%</td>
</tr>
</tbody>
</table>

It is seen from the table, on the safe assumption that the percentage contributions vary monotonically as \( V_0 \) varies from 30 MeV to 50 MeV, that there is no possibility that the \( \beta_0 \) transition is prohibited.

By combining these results with the work of
Plaasman and Langer (83), it is possible to derive some information about the $\beta_0$ shape factor.

Using the shape formula, 8, given on page 8c, Plaasman and Langer found the best pairs of values for $x$ and $\xi$ to give the observed shape factor for RaE $\rightarrow$ RaF. Their pairs of values lie on a straight line ($x$ plotted against $\xi$).

Bayman selects one of these pairs by the following method. First he finds a value of $\Lambda$ by means of a formula derived by Pursey (110). (All the parameters used here are defined on pages 85, 86.)

$$\frac{\alpha Z}{2R} \Lambda = (W_0 - 1) - (M_N - M_p) + \frac{\alpha Z}{R}$$

$$- \frac{15(N - Z)(1 - 3A^{-2/3})}{A^3} - 3 \text{e} \frac{1 - A^{-2/3}}{A^3}.$$

$M_N - M_p$ is the neutron-proton mass difference.

$\text{e}$ is a function of $V_o$ (see below). The results are

**Table XIX.**

<table>
<thead>
<tr>
<th>Transition</th>
<th>$\frac{\alpha Z}{R}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$(V_o = 30 \text{ MeV})$</td>
</tr>
<tr>
<td>RaE $\rightarrow$ RaF</td>
<td>30.9</td>
</tr>
<tr>
<td>RaD $\rightarrow$ RaE</td>
<td>26.3</td>
</tr>
</tbody>
</table>

Then, since
it follows that for RaE,

\[ x \approx 1.42 - 16.7 \xi \]

for both values of \( V_0 \).

Thus \( x \) and \( \xi \) satisfy two linear relations, the one above, and the empirical one plotted by Plasman and Langer. The intersect gives

\[
\begin{align*}
    x & = 3.6 \\
    \xi & = 0.63
\end{align*}
\]

for RaE.

These values cannot be used directly for the \( \beta_0 \) transition since the initial and final states are different in the two cases. However the ratio of the values of \( \xi \) appropriate to the two transitions can be found as follows.

According to Pursey (110), the shell model predicts that

\[ \xi = k_1 - k_2 \left( \frac{c_T}{v_g} \xi^{-1} \right) \]

where

\[
\begin{align*}
    k & = - (j + \frac{1}{2}) \text{ for } j = \ell + \frac{1}{2} \\
    k & = + (j + \frac{1}{2}) \text{ for } j = \ell - \frac{1}{2}
\end{align*}
\]

and the suffixes refer to the initial and final states of the decaying neutron.
Using the mixing coefficients given in Table XVIII the results of Table XX are then calculated.

Table XX.

<table>
<thead>
<tr>
<th>Transition</th>
<th>$\epsilon (V_c = 30 \text{ MeV})$</th>
<th>$\epsilon (V_c = 50 \text{ MeV})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>RaE $\rightarrow$ RaP</td>
<td>3.143</td>
<td>1.13</td>
</tr>
<tr>
<td>RaD $\rightarrow$ RaE</td>
<td>5.66</td>
<td>0.88</td>
</tr>
</tbody>
</table>

The ratios of the values of $\frac{C}{\epsilon} = C_S/\epsilon C_T$ for the two transitions are given by the inverse ratios of the corresponding values of $\epsilon$, since the ratio $C_S/C_T$, whose precise value is at present unknown, is certainly the same for the two transitions (indeed, for all transitions).

So the value of $\frac{C}{\epsilon}$, and hence $\chi$, for the $\beta_0$ transition can be derived from the corresponding value for the RaE $\rightarrow$ RaP transition:

$$(V_c = 30 \text{ MeV}) \begin{cases} \chi = 3.6 \\ \frac{C}{\epsilon} = 0.38 \end{cases} \text{ for } \beta_0$$

$$(V_c = 50 \text{ MeV}) \begin{cases} \chi = -1.3 \\ \frac{C}{\epsilon} = 0.81 \end{cases} \text{ for } \beta_0$$

It has been mentioned in Chapter V that the extreme of energy dependence for the shape factor $S$ is produced when there is complete cancellation of the
145.

energy independent factor, i.e., when \( x = 0 \). Since \( x \) goes through zero in the region of uncertainty of \( V_C \), it is possible that this happens with the \( \beta_0 \) transition. Linear interpolation gives \( f = 0.7 \) at \( x = 0 \), and these values were used to calculate \( S_{EXT} \).

Given a definite value for \( V_C \), the shape factor for the \( \beta_0 \) spectrum could be calculated. Conversely, if the \( \beta_0 \) spectrum shape could be measured, it might be possible to find the value of \( V_C \). The fitting of the formula \( S \) to the observed shape factor would yield a series of pairs of values for \( x \) and \( f \). But, as has been seen, calculation for different values of \( V_C \) also yields a series of pairs of values. Comparison of the two sets would fix \( x, f \) and \( V_C \) uniquely. (The method would break down if, by chance, the two \( x-f \) relationships nearly coincided.) With \( V_C \) known, it would then be possible to calculate the configuration mixing ratios and hence the value of \( \epsilon \). Combining this with the corresponding value of \( f \) would yield the ratio \( C_S/C_T \). Thus it would seem that measurement of the shape of the \( \beta_0 \) spectrum might result in a determination of two of the fundamental constants in \( \beta \)-decay theory. The determination would, of course, be subject to the uncertainties inherent in the calculation (the validity of the shell model, the ratio of singlet to triplet force strengths etc.) but it would be an important contribution to the body of
knowledge needed for the elucidation of the theory.

The value found for the comparative half-life \((\log f_0t = 7.84 \pm 0.20)\) of the \(\beta_o\) transition is not, in itself, of any great significance, except perhaps that its closeness to that for RaE \((\log f_0t = 8.0)\) indicates that a similar cancellation occurs in the shape factors of both. However, in other paired (even-even \(\rightarrow\) odd-odd \(\rightarrow\) even-even) transitions, the \(f_0t\) values show no marked tendency towards equality. There is no systematic trend from which conclusions might be drawn.
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