A. Fage and R. F. Sargent

DESCRIPTION OF PLATES 1 and 2

Plate 1

Figure 5. (a) to (b) Töpler striation photographs; (c) to (i) direct shadow photographs.

(a) $M_1 = 1.407, \chi = 2.11$.  
(b) $M_1 = 1.283, \chi = 1.76$.  
(c) $M_1 = 1.407, \chi = 2.11$.  
(d) $M_1 = 1.443, \chi = 2.22$.  
(e) $M_1 = 1.392, \chi = 2.10$.  
(f) $M_1 = 1.346, \chi = 1.95$.  
(g) $M_1 = 1.336, \chi = 1.87$.  
(h) $M_1 = 1.310, \chi = 1.84$.  
(i) $M_1 = 1.206, \chi = 1.52$.

Plate 2

Figure 6. (l), (o), (p) Töpler striation photographs; (j), (k), (m), (n), (q) direct shadow photographs.

(j) $M_1 = 1.08, \chi = 1.19$.  
(k) $M_1 = 1.472, \chi = 2.33$.  
(l) $M_1 = 1.472, \chi = 2.33$.  
(m) $M_1 = 1.439, \chi = 2.25$.  
(n) $M_1 = 1.439, \chi = 1.76$.  
(o) $M_1 = 1.439, \chi = 2.25$.  
(p) $M_1 = 1.439, \chi = 1.76$.  
(q) $M_1 = 1.469, \chi = 1.39$.

The assignment of the slow-neutron-produced activities of thallium and the dual disintegration of radium E

By E. Broda and N. Feather, F.R.S.

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Recoil experiments with a strong source of RaE (21 mC initial activity) electrolytically deposited on platinum show that an activity of half-value period 4-2 min. is obtained. This observation indicates that Ra E undergoes dual disintegration with an $\alpha:\beta$ branching ratio of the order of $5 \times 10^{-7}$. Rough absorption measurements favour the identification of the activity with the 4-2-min. activity produced in the $(n, \gamma)$ and $(d, p)$ reactions with thallium, and therefore require assignment of the latter to $206^{\text{Tl}}$, contrary to earlier suggestions. Consideration of the energies involved shows that the postulated $\alpha:\beta$ branching ratio has roughly the value which would be predicted on the basis of accepted regularities.

Irradiation of thallium by slow neutrons, as well as its transformation in the $(d, p)$ reaction (Fajans & Voigt 1941), leads to the production of $\beta$-activities of half-value periods 4-2 min. and 3-5 years. On the basis of the observation of Heyn (1937), that the 4-2-min. activity is also obtained as the result of fast-neutron irradiation, it has generally been assumed (see, for example, Seaborg 1944) that the 4-2-min. activity is to be attributed to $206^{\text{Tl}}$ and the activity of 3-5 years half-value period to $204^{\text{Tl}}$. The arbitrary nature of this assignment has been pointed out by Fajans & Voigt (1941), and it is the object of the present paper to indicate how the matter may be further investigated from a different starting-point and to describe some experiments, in which it was possible for $206^{\text{Tl}}$ but not $204^{\text{Tl}}$ to be produced, which show
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conclusively that the currently accepted assignment is incorrect. In these experiments the 4.2-min. activity was in fact obtained, but not the other; thus as a result of them it must be concluded that the short-period activity definitely belongs to the heavier, and the long-period activity presumably to the lighter, of the two thallium isotopes in question. Heyn's observation must then be explained either as an early example of the detection of fast-neutron capture, or on the basis of his failure completely to exclude slow neutrons in his experimental arrangement.

Briefly, the new experimental approach is by way of a close reinvestigation of the natural radioactivity of Ra E ($^{210}_{83}$). Obviously, if $\alpha : \beta$ branching occurs with Ra E, as with the other known $\beta$-active isotopes of bismuth, then $^{208}_{81}$Tl will be formed in the rare mode of disintegration. Energetically, $\alpha : \beta$ branching is possible so long as the energy liberated in the $\beta$-disintegration of $^{206}$Tl is less than 6.57 MeV, the sum of the disintegration energies of Ra E and Ra F; practically, its observation is likely only if the $^{206}$Tl $\beta$-disintegration energy lies within fairly narrow limits. We shall inquire, therefore, what the branching ratio would be, first on the assumption that the 3.5-year body is to be identified as $^{206}$Tl and then on the assumption that the 4.2-min. activity is to be assigned to this species. The accepted values for the $\beta$-disintegration energies are 0.87 MeV in the former case (Fajans & Voigt 1941) and $1.70 \pm 0.05$ MeV (Fajans & Voigt 1940, 1941; Krishnan & Nahum 1940) in the latter; thus the energy released in the hypothetical $\alpha$-disintegration of Ra E is 5.70 MeV according to the first and $4.87 \pm 0.05$ MeV according to the second assumption. Figure 1 provides the basis for an estimate of partial disintegration constants once

![Figure 1](http://rspa.royalsocietypublishing.org/)
these energies are given. In this figure total disintegration energies and partial disintegration constants are plotted (the latter logarithmically) for all the α-particle groups belonging to the known modes of disintegration of Ac C (\(^{234}\)\(_{95}\)), Th C (\(^{232}\)\(_{90}\)) and Ra C (\(^{214}\)\(_{88}\)). The full curve bounding the points on the side of \(\lambda\) greatest (drawn and extrapolated with reference to corresponding curves for \(\alpha\)-bodies having other values of \(Z\) (Feather 1946))\(^{*}\) is assumed to represent the relation between \(\log_{10} \lambda\) and \(E^\uparrow\) for disintegrations in which there is no change of spin—and the distribution of points below the curve indicates the extent to which spin changes are likely to result in smaller values of \(\lambda\) (by a factor of roughly 10 in the extreme cases illustrated) than would otherwise apply. From this figure, using the energies already quoted, table 1 has been prepared. From the values of the \(\alpha:\beta\) branching ratios given in the table it is evident at once that the assignment of the 3·5-year activity to \(^{206}\)Tl runs entirely contrary to the crudely determined fact that there is no very obvious \(\alpha\)-component in the total radiation from Ra E—at least so long as our general views on the probabilities of radioactive change remain unchallenged. But it is also clear from the table that, if the alternative assignment were to be adopted, and the \(\beta\)-particles of 1·70 MeV limiting energy were to be attributed to the disintegration \(^{206}\)Tl (4·2 m.) \(\beta\). \(^{206}\)Pb, then there would be nothing very surprising in the fact that the \(\alpha:\beta\) branching of Ra E has not hitherto been discovered—or in the fact that the feeble higher-energy component due to the presence of \(^{206}\)Tl in equilibrium in Ra E sources has so far remained undetected. The experiments of Bastings (1924) on the decay of strong sources, as measured by the ionization produced by the \(\beta\)-particles of highest energy only, would not have been sensitive enough to detect any initially anomalous decay, even if his sources had been rapidly prepared and examined, and the most careful investigations on the upper limit of the \(\beta\)-spectrum by the magnetic method (such as those of Alichanian, Alichanov & Dželepov (1938)) would fail, on account of background effects due to scattered \(\beta\)-particles, to indicate the presence of the 1·70 MeV component if its intensity were less than \(10^{-3}\) with respect to the main spectrum having its end-point at 1·17 MeV. It would, in fact, appear that, of the experiments reported up to date, those of Gray & Henderson (1936) by the simple absorption method set the lowest limit to the possible admixture of higher-energy particles with the main radiation from Ra E. These authors claim to have shown that not more than one \(\beta\)-particle in \(2 \times 10^6\) has sufficient energy to penetrate 0·400 g./cm.\(^2\) aluminium plus 0·212 g./cm.\(^2\) lead. Accepting this figure, it can readily be granted that the degree of admixture of a 1·70 MeV component with the main radiation cannot be greater than 1 in \(10^4\). Summing up the position, then, one would say that the negative evidence from previous experiments is overwhelmingly in favour of the attribution of the 4·2-min. thallium activity to \(^{206}\)Tl and the 3·5-year activity to \(^{204}\)Tl, rather than the reverse, which, as already stated, is the currently

\(^{*}\) From back numbers of periodicals recently received, it appears that Berthelot (1942) pointed out the regularities which are evident in Geiger-Nuttall diagrams plotted for individual values of \(Z\) before this point was stressed by one of the present writers.

\(^\uparrow\) \(\lambda\) in sec.\(^{-1}\), and \(E\) in MeV.
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accepted attribution. Further, it may be repeated in anticipation, the new experiments now to be described provide positive evidence for the correctness of this conclusion.

| Table 1 |
|-----------------|-----------------|-----------------|
| assumed half-value | energy of α-disintegration | α;β branching ratio for Ra E |
| period of 206Tl | of 210Ra E | 37:63 to 3-7:963 |
| 3-5 years | 5-70 MeV | 9 × 10^{-6} to 2 × 10^{-7} |
| 4-2 min. | 4-87 ± 0-05 MeV | |

Our first experiments were designed to test whether the 3-5-year body is in fact produced from Ra E, unprejudiced by the consideration that a positive result would be extremely unlikely for the reasons already set forth. We had at our disposal a solution containing about 10 mC Ra D which had not been treated chemically for at least 8 years. This solution was evaporated to dryness, and the residue, which contained a few milligrams of inactive lead, was dissolved in HCl and small quantities of thallous and didymium salts were added as carriers. Didymium hydroxide was precipitated with ammonia and this precipitate carried down Ra D, Ra E and polonium. The process of addition of didymium and precipitation with ammonia was repeated until the filtrate was considered to be sufficiently free from Ra E for the purposes of the test. Ammonium iodide was then added to precipitate the thallium. The mixture was filtered and the precipitate of thallous iodide was tested for activity using a G.M. counter having a mica window of about 3-5 mg./cm.² thickness covering one end. A small activity was observed initially, but when the precipitate was dissolved and reprecipitated, after traces of lead and bismuth salts had been added to the solution, the residual counting rate due to the thallium was reduced to 0-8 ± 0-4 min.⁻¹ under the geometrical conditions obtaining in the experiment. In order to make sure that the negative result of this test was not due to the fact that the active thallium had been preferentially adsorbed on the glass of the vessel in which the Ra D solution had been standing, an acid solution of thallium salt was brought to the boil and allowed to stand in this vessel overnight and a similar test was made on the iodide precipitate obtained from this solution. A similarly negative result was again found. Taking count of the possibility of incomplete radioactive equilibrium in the parent solution, and of losses in the chemical processes involved, these results set an upper limit to the α;β branching ratio corresponding to the formation of the 3-5-year thallium from Ra E at a value certainly not greater than 10⁻⁸:1. This conclusion having been reached, attention was concentrated on testing for the formation of the 4-2-min. active thallium, as described below.

For this second stage of the investigation, chemical methods of separation were tried first. A procedure was evolved which allowed of the separation of thallium with a reasonable yield (25 to 50 %), even from a very large excess of bismuth (with Ra E), and the production of a sample in a form ready for counting, in 7 or 8 min. Before the method could be applied the polonium content of the main solution had first to be reduced as far as possible. To this end the Ra (D + E + F) was taken up in
$\frac{n}{10}$ HCl and three copper foils were rotated for successive periods of 1 hr. each in the solution. The last traces of polonium were removed by adding about 1 g. Bi$_2$O$_3$ dissolved in HCl, some crystals of telluric acid, and finally a solution containing about 200 mg. SnCl$_2$. The polonium and tellurium, which precipitate quantitatively in the cold, were removed by filtration. The filtrate was evaporated to dryness, dissolved in 1 : 1 HCl, and extracted with ether repeatedly until no further solid was recovered. The solution, now ready for the thallium separation, contained, at the time of the first experiment, about 7 mC Ra E and 0.1 mC polonium. The method of separation was as follows. To the solution of bismuth (and Ra E) chloride, about 12 mg. TINO$_3$ was added, together with excess bromine to oxidize the thallium, and the whole was heated on a water bath. After the TINO$_3$ had dissolved and the colour of the bromine had more or less disappeared, the solution was chilled with ice and stirred quickly with 2 c.c. of ether for about 10 sec. The end of the stirring was taken as the effective time of separation of the radiothallium. The ether, containing the extracted thallium chloride was then pipetted off, washed four times with 1 : 1 HCl in a separating funnel, and evaporated drop by drop on a warm glass dish. The solid residue so obtained was transferred to the counter and tested. After the method had been perfected the initial activity of the thallium residue was generally of the order of 10 to 15 counts per minute (counter efficiency $\sim 3\%$). With such a small counting rate it was naturally difficult to investigate a possible short-period decay, but even to attribute the whole observed effect to $^{206}$Tl is already to set quite a low upper limit to the branching ratio concerned (not greater than $10^{-6}$ : 1, assuming 25 % efficiency for the chemical processes involved). As to the decay of the effect, the conclusion from a large number of extractions was definitely that a decrease in counting rate took place during the first few minutes’ observation, but it was evident from control experiments that part of this decrease at least was associated with changes in temperature (the sample being presented to the counter on a dish which had just been heated) and the final result was that the experiments, though suggestive, were at best inconclusive.

The attempt was then made to separate an active thallium from Ra E by the method of recoil. To this end it was necessary to prepare a strong source of Ra E of the minimum thickness, and containing as little polonium as possible in order to reduce aggregate recoil effects to tolerable proportions. A solution of Ra (D + E + F) of about 50 mC strength was freed from most of the polonium by the rotation of six copper foils. Thereafter two nickel foils were rotated in the solution for 45 min. each, the acidity being kept at $\frac{n}{10}$ HCl and the temperature that of the water-bath. The nickel foils were dissolved in nitric acid, about 200 mg. didymium nitrate was added, and didymium and Ra E were precipitated as hydroxides with ammonia. The precipitate was then washed with ammonia, to remove all traces of nickel, and was dissolved in trichloracetic acid. This solution was evaporated to dryness and the residue again taken up in trichloracetic acid ($2\%$) and electrolyzed for 12 hr., using platinum electrodes, each of area about 7 cm.$^2$, at 1.9 V and 1 mA. About 21 mC of Ra E was found on the cathode after electrolysis. The weight of the deposit was
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certainly not more than 0.1 mg. and may have been considerably less, the surface
of the platinum showing no discoloration.

Recoil experiments were made with this source over a period of 8 days. Throughout
these experiments the source remained in position in the recoil vessel, and its decay
was followed by removing the ebonite plug from the vessel, replacing it by a brass
plug bored with a 0.25 in. hole and carrying an absorber of aluminium of 125 mg./cm.²
thickness, and bringing the whole vessel (with source) into a standard position below
a β-ray electroscope. The activity of the source, measured in this way, was found to
decrease accurately according to an exponential law with a half-value period of
5.0 days. Exposures were made using a brass button of 2.0 cm. diameter as collector,
the button being held at a distance of 6 mm. from the platinum disk carrying the
Ra E, and the collecting voltage being 450 V. Both for the investigation of the pos-
sible short-period decay of the activity collected by recoil, and, when this had been
established, for the study of the absorption of the radiations from the short-lived
body so formed, as well as for the investigation of the variation of the amount of
short-period activity collected with the age of the primary source, sets of six ex-
posures of 12 min. each were made, with just sufficiently long intervals in between
for a study of the initial decay of the activity to be made. Table 2 is a summary of
the results obtained with the first set of six exposures, which established the fact
that a short-lived body of half-value period about 4.2 min. was in fact collected.
The figures quoted in the table are the total numbers of counts recorded (the
‘natural’ of the counter being included) during the periods of counting given in the
left-hand column, for each of the six sources belonging to the set in question. That
the ‘residual’ count (19 to 27 min. from the end of the exposure) is systematically
greater for sources 2, 4 and 6 than for 1, 3 and 5 is simply to be explained by the fact
that the exposure button was newly cleaned only before exposures numbers 1, 3
and 5; for the alternate exposures it was put back to expose with the decayed active
deposit from the previous exposure still adhering to it. The totals given in the extreme
right-hand column of the table were treated numerically on the assumption that a
simple admixture of a constant background counting rate (counter ‘natural’ plus
the activity due to aggregate recoil) with an exponentially decaying rate was in-
volved. In order to determine the background rate it was assumed that the short
decay period was in fact 4.2 min. On this basis the total of 1740 counts for the set of
six observation periods 19 to 27 min. after the end of the exposure was assumed to
be made up of $81 \pm 4$ counts due to the short-period product which had almost
decayed and $1659 \pm 28$ due to background. Of the mean rate due to this back-
ground ($34.6 \pm 0.6$ min$^{-1}$) more than two-thirds (about 24 min$^{-1}$) was known
to be due to counter ‘natural’). The excess (about 10 min$^{-1}$) represents the
activity collected by aggregate recoil—effectively during 18 min. exposure.* The
nature of this activity was examined by longer exposures as will be described
below.

* Taking count of the fact that the button was cleaned for re-exposure on three occasions
only (vide supra).
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Having determined the background rate as just described, appropriate deductions were made from the totals for the counts belonging to the eight successive sets of 2 min. periods of observation to which the entries in the main body of table 2 refer. In figure 2 these corrected totals, with probable errors, are plotted logarithmically against the mean times of observation, referred to the end of exposure as $t = 0$.

<table>
<thead>
<tr>
<th>time from end of 12 min. exposure (min.)</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>1 – 3</td>
<td>183</td>
<td>175</td>
<td>175</td>
<td>162</td>
<td>148</td>
<td>159</td>
<td>1002</td>
</tr>
<tr>
<td>3½–5½</td>
<td>145</td>
<td>141</td>
<td>129</td>
<td>129</td>
<td>117</td>
<td>122</td>
<td>783</td>
</tr>
<tr>
<td>5½–7½</td>
<td>122</td>
<td>129</td>
<td>107</td>
<td>136</td>
<td>101</td>
<td>113</td>
<td>708</td>
</tr>
<tr>
<td>7½–9½</td>
<td>100</td>
<td>133</td>
<td>94</td>
<td>82</td>
<td>83</td>
<td>96</td>
<td>588</td>
</tr>
<tr>
<td>10–12</td>
<td>101</td>
<td>110</td>
<td>92</td>
<td>89</td>
<td>88</td>
<td>112</td>
<td>592</td>
</tr>
<tr>
<td>12½–14½</td>
<td>92</td>
<td>79</td>
<td>98</td>
<td>79</td>
<td>84</td>
<td>513</td>
<td></td>
</tr>
<tr>
<td>14½–16½</td>
<td>94</td>
<td>105</td>
<td>78</td>
<td>82</td>
<td>62</td>
<td>77</td>
<td>498</td>
</tr>
<tr>
<td>16½–18½</td>
<td>89</td>
<td>80</td>
<td>69</td>
<td>87</td>
<td>72</td>
<td>88</td>
<td>485</td>
</tr>
<tr>
<td>19–27</td>
<td>260</td>
<td>338</td>
<td>263</td>
<td>297</td>
<td>263</td>
<td>319</td>
<td>1740</td>
</tr>
</tbody>
</table>

**Figure 2**

The straight line in the figure represents an activity with half-value period 4·2 min. and with most probable initial strength as deduced from the numerical analysis already carried out. The lie of this line amongst the points is sufficient proof that the whole basis of interpretation is essentially correct. A short-period activity of 4·2-min. period is very obviously in question. At the mean time at which the set of six ex-
posures here dealt with was made, the calculated counting rate due to the amount of this short-lived body which was collected by recoil in 12 min. under our standard conditions was \((69.7 \pm 3.1) \text{ min}^{-1}\) at \(t = 0\).

The results just analysed refer to the first set of six exposures made when the primary Ra E source was at its strongest. Table 3 gives also the final result obtained from later sets, again expressed in terms of the calculated counting rate under standard conditions at \(t = 0\), deduced by similar numerical analysis of the other observations. The first row of the table indicates the number of the experiment, the second row shows the mean activity of the primary source in electroscope divisions per minute, determined as already described, whilst the third row gives the corresponding calculated initial rate due to the collected activity in counts per minute. Relative values of the yield per atom of Ra E disintegrating are then given (row 4) as the quotients obtained by dividing the numbers in row 3 by the corresponding numbers in row 2 of the table. It will be observed that, within the limits of error of the comparison, the yield is constant when referred to unit quantity of Ra E in the primary source. This result is clearly an unambiguous proof that the activity collected is that of an immediate daughter product of Ra E (in a very rare mode of disintegration) and that its production is in no way explicable by the initial presence or later growth of polonium in the source (production of an unidentified radioelement by \(\alpha\)-particle-induced transformation in the air or the electrode material—or formation of \(^{206}\text{Pb}\) in a metastable state in a rare mode of disintegration of polonium itself).

<table>
<thead>
<tr>
<th>experiment</th>
<th>1</th>
<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>mean activity Ra E</td>
<td>103.1</td>
<td>88.1</td>
<td>38.4</td>
</tr>
<tr>
<td>calculated initial activity of 4.2-min. body</td>
<td>(69.7 \pm 3.1)</td>
<td>(59.0 \pm 3.9)</td>
<td>(23.3 \pm 2.7)</td>
</tr>
<tr>
<td>relative yield</td>
<td>(0.676 \pm 0.030)</td>
<td>(0.670 \pm 0.044)</td>
<td>(0.606 \pm 0.070)</td>
</tr>
</tbody>
</table>

Absorption experiments, already briefly mentioned, provided further evidence for the identification of the short-lived body collected by recoil with \(^{206}\text{Tl}\)—or rather with the 4.2-min. active thallium of earlier investigators. In these experiments attention was concentrated on obtaining a fairly close estimate of the reduction in counting rate due to a single absorber, and for this purpose an aluminium foil of 78 mg./cm.\(^2\) thickness was chosen, as representing about one-tenth of the effective range of the \(\beta\)-particles from the short-lived thallium in question (Fajans & Voigt 1940; Krishnan & Nahum 1940). With each of a set of six sources obtained by recoil (12 min. collection) alternate periods of 2 min. counting were carried out with and without absorber, during the first 18\(\frac{3}{4}\) min. life of the source (cf. table 2), and then two successive periods of 8 min., with and without absorber, for the estimation of the background effect. Numerical analysis was carried out as before. The final result was that the reduction factor for the 78 mg./cm.\(^2\) foil in relation to the \(\beta\)-particles from the 4.2-min. body was found to be \(0.685 \pm 0.062\). By comparison with the absorption
of the primary $\beta$-particles of uranium $X_2$, this is just about what would be expected for the $\beta$-radiation from an ‘allowed’ transition using an absorber of thickness roughly one-tenth of the effective range of the particles.

It has been mentioned that, in addition to the 4-2-min. activity, collection by the method of recoil yielded a background activity which, in our numerical analyses, has been assumed constant over the period covered by the experiments with each short-exposure source. This point was checked by taking longer exposures, which showed that the amount of background activity was directly proportional to the time of exposure (for exposures made within a few hours of one another), and by decay and absorption measurements on the radiations from the stronger sources of background activity collected during exposures lasting overnight. These measurements showed that the substance responsible for the background activity had very closely the same properties as the Ra (E + F) mixture constituting the parent source at the time the exposure was made. To attribute the effect to aggregate recoil, therefore, was the natural assumption. Aggregate recoil in this case is certainly due almost entirely to the presence of polonium in the source, and it is in line with this assumption that it was found that the intensity of the background activity collected by overnight exposure did not decrease anything like so rapidly as the activity of the Ra $E$ contained in the parent source. Assuming that the aggregate recoil effect as observed in our conditions of counting, and for a constant exposure time, is proportional to the product of the mean activities of Ra $E$ and polonium in the parent source during the period of exposure, we have the result that the intensity of the effect for short exposures should decrease or increase initially with increasing age of the primary source as the number of atoms of polonium in the source initially is greater or less than $\lambda_1/(\lambda_1 + \lambda_2)$ of the initial number of Ra $E$ atoms present. Here $\lambda_1$ and $\lambda_2$ are the disintegration constants for Ra $E$ and polonium, respectively. In our case the amount of background activity collected per minute exposure time decreased during the first few days' life of the parent source. On these assumptions, therefore, the initial activity of the polonium in the source must have been greater than $\frac{1}{3}$ of that of the Ra $E$, that is greater than 0.75 mC. This is not entirely surprising in view of the time taken to work up the original source of 50 mC strength. At no time, however, was aggregate recoil a serious difficulty in our main experiment. The counting rate due to the background effect was never greater than 0.6 per min. for each minute of exposure—and as exposures in general were of 12 min. duration it will be seen that the overall result of aggregate recoil was effectively to increase the 'natural' of the counter by about 30%.

Returning to the question of branching ratio, since our standard exposure of 12 min. collected only 0.862 of the short-period activity which would have been collected during an ‘infinite’ exposure, we conclude that the saturation activity of the 4-2-min. body in the parent source at the time of experiment 1 of table 3 must have been at least $(2 \times 69.7)/0.862$ counts per minute in our counter arrangement (collection by the method of recoil cannot be more than 50% efficient). We determined the efficiency of the counter in this arrangement for the primary $\beta$-particles.
of Ra E, obtaining the value \((3.4 \pm 0.1)\%\). Taking the strength of the parent source at 17.5 mC at the time of the experiment in question, we obtain, therefore, as a lower limit to the branching ratio
\[
\frac{2 \times 69.7 \times 100}{0.862 \times 3.4 \times 60} \times \frac{1}{17.5 \times 3.7 \times 10^7} = 1.2 \times 10^{-7}.
\]

Remembering that the result of the chemical experiment led to a 'safe' upper limit of \(10^{-6}\) for this ratio, we may be satisfied that we now know the ratio within a factor of 5 and also that our strong source of Ra E prepared by electrolysis was reasonably 'clean' in relation to \(\alpha\)-recoil, that is that the mean surface density of radioactive deposit was less than 0.01 mg./cm.\(^2\) of 'heavy' elements or about 0.002 mg./cm.\(^2\) of 'light' elements. It will be noticed that the experimental value for the branching ratio is probably within the range of values given in table 1, though it is rather low in that range. If it is assumed that the disintegration energy of the 4.2 min. body is known to the accuracy supposed, two explanations are possible: either our extrapolation of the \((\log_{10} \lambda)/E\) curve is somewhat at fault or is not strictly relevant (which is by no means excluded) or the \(\alpha\)-disintegration of Ra E involves a change of nuclear spin. It might be remarked that there is every reason to suppose that the latter is in fact the case (the \(\beta\)-disintegration of Ra E is 'forbidden', that of the 4.2-min. body is 'allowed', and the \(\alpha\)-particle disintegration of polonium is unlikely to involve any spin change). On this supposition, then, the findings of experiment and the theoretical predictions are in full accord.

Regarded from this point of view, our discovery of the generic relation between Ra E and the 4.2-min. thallium is in effect a determination, within narrow limits, of the energy emitted in the disintegration
\[
\begin{pmatrix} 210 \\ 83 \end{pmatrix} \xrightarrow{\alpha} \begin{pmatrix} 206 \\ 81 \end{pmatrix}.
\]

This energy appears to be \(4.87 \pm 0.05\) MeV. It is interesting to compare this energy with the \(\alpha\)-disintegration energies of the other isotopes of bismuth \((Z = 83)\). Values are listed in table 4. As the mass number decreases the energy of \(\alpha\)-disintegration at first increases steadily and then, through \(A = 210\), falls abruptly until with \(^{209}_{83}\)Bi it is so small that the radioactivity of natural bismuth has never been detected. This variation of \(E\) with \(A\) for \(Z\) constant is reproduced exactly for \(Z = 84\). Disintegration energies are known for \(A = 218, 216, 215, 214\) and 212, and for this series there is once more a steady increase of \(E\) as \(A\) decreases, then, even more rapidly, \(E\) decreases again for \(A = 211\) and decreases still further for \(A = 210\) (polonium)*. For \(Z = 85\) information is less complete, but for this element the effect is again evident; the \(\alpha\)-disintegration energy for \(A = 211\) is definitely less than for \(A = 218\) and 216. Since for \(Z > 83\) no stable isotope is known, the rapid decrease in \(\alpha\)-

* This effect previously remarked by one of us (Feather 1946), and referred to as the polonium anomaly, had at an earlier date been commented on by Meyer (1936) whose paper containing these comments had escaped our notice.
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disintegration energy beyond a certain value of \( A \) (as \( A \) decreases in each case) can only be understood on the assumption that the lighter isotopes of these elements are unstable in relation to positron (or \( K \)-capture) disintegration—or that they undergo fission spontaneously at a relatively high rate. We already know, in fact, that the former suggestion is valid for \( ^{211}_{85}\) which exhibits \( \alpha:K \)-capture branching with a ratio of 40:60, but this does not necessarily invalidate the latter suggestion in other possible cases.

<table>
<thead>
<tr>
<th>mass number, ( A )</th>
<th>214</th>
<th>212</th>
<th>211</th>
<th>210</th>
<th>209</th>
</tr>
</thead>
<tbody>
<tr>
<td>energy of ( \alpha )-disintegration MeV</td>
<td>5.61</td>
<td>6.20</td>
<td>6.74</td>
<td>4.87</td>
<td>stable</td>
</tr>
</tbody>
</table>

Finally, in regard to nomenclature: we would suggest that since \( ^{206}\)Tl has now been shown to be a member of the uranium series of elements, its designation in that series should be Ra E"—bringing it into line with the other thallium isotopes which are derivatives of the ‘classical’ radioelements.

Throughout the whole of the work described here we have had the constant assistance of Miss P. K. Wright, both in the chemical work and in the routine counting of ‘aliquots’ for calibration purposes. Our best thanks are due to her for this help.

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**REFERENCES**


Bastings 1924 *Phil. Mag.* **48**, 1075.


