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β–γ angular correlations using sources of thorium active deposit

R. D. SCOTT† and J. KYLES
Department of Natural Philosophy, University of Edinburgh
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Abstract. β–γ angular correlations have been studied for the following cascades occurring in the decay chain of the thorium active deposit:

(i) 350 kev β–239 kev γ in the ThB → ThC decay.
(ii) 1·52 Mev β–729 kev γ in the ThC → ThC′ decay.
(iii) 1·80 Mev β–583 kev γ in the ThC″ → ThD decay.

In all three cases, a null result was obtained. The β decays in question are of the first forbidden type according to the accepted decay schemes and might, therefore, have been expected to show some anisotropy in the β–γ angular correlation. The β transitions in cascades (i) and (iii) above have rather low (< 6) lg *ft* values, more in keeping with an allowed assignment, but it is concluded that the lack of a measurable anisotropy is not sufficient evidence to warrant the adoption of a possible alternative decay scheme which would make all three decays of allowed type.

1. Introduction

In an attempt to establish the degree of forbiddenness of some β transitions occurring in the neighbourhood of the doubly closed shell at ^{208}Pb, β–γ angular correlations have been performed using sources of the active deposit of thorium. The relevant parts of the presently accepted decay schemes of this chain are shown in figure 1. The decay scheme

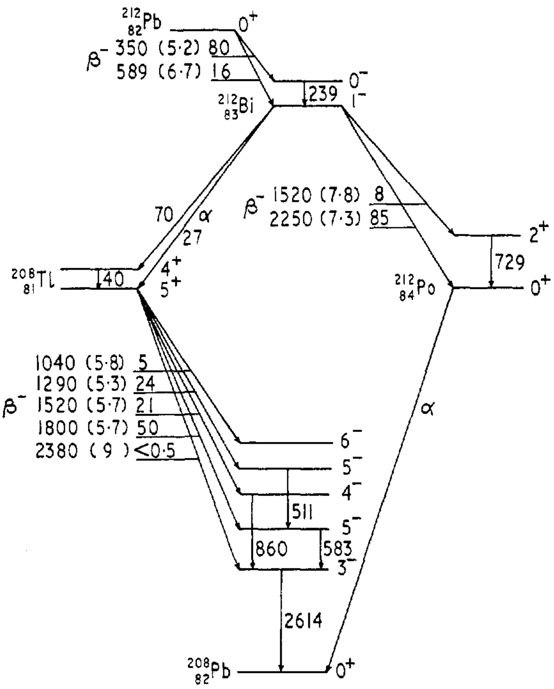

Figure 1. Partial decay scheme of the thorium active deposit. Energies are in kev. The numbers in brackets are lg *ft* values and the β transition intensities are shown.

† Now at Scottish Research Reactor Centre, East Kilbride, Scotland.

shows that, with a single exception, the β transitions are of the first forbidden type. However, the lg *ft* values of some of the transitions are more in keeping with an allowed, rather than a first forbidden assignment. Thus, the $0^+ \rightarrow 0^-$ transition of ThB (^{212}Pb) and all but the very weak, highest-energy transition of ThC″ (^{208}Tl) have lg *ft* values less than 6. On the other hand, the ground state to ground state ThB transition and those of ThC (^{212}Bi) have lg *ft* values appropriate to their first forbidden assignment.

The decay scheme of figure 1 is based upon the following experimental evidence. Doubly even nuclei have a 0^+ ground state. Conversion coefficient measurements (Martin and Richardson 1950, Martin and Parry 1955, Sokolowski *et al.* 1956, Krisyouk *et al.* 1957, Emery and Kane 1960, Roetling *et al.* 1960) have shown the 239 kev γ-ray of ThC to be pure M1 and the 729 kev γ-ray of ThC′ (^{212}Po) to be pure E2 in nature. Thus, either the ground or first excited state of ThC is likely to have zero spin and the first excited state of ThC′ is 2^+, an assignment which is consistent with the observation of long-range α-particles from that level. The 40 kev γ-ray of ThC″ is of M1 type (Horton 1955) so that the ground and first excited states of ThC″ have the same parity and either the same spin, or spins differing by one unit. The level scheme of ThD (^{208}Pb) has been well established by γ–γ angular correlation studies (Elliot *et al.* 1954).

It is difficult, on the experimental evidence alone, to assign unambiguous spin and parity values to the ground states of the odd–odd nuclei ThC and ThC″. Spin 0 for the ThC ground state is unlikely from a consideration of the α intensities in the ThC $\rightarrow$ ThC″ transition, and β-spectrum shape measurements of the ThC $\rightarrow$ ThC′ decay also tend to rule out this possibility (Burde and Rozner 1957). Spin 2, or more, is also eliminated and the most likely value is therefore 1. Such an assignment is clearly consistent with the directly measured spin of 1 for RaE (^{210}Bi) provided that the additional neutrons couple to give zero spin. The α–γ angular correlation experiment of Horton (1955) using the 6·05 Mev α-particles from ThC and the succeeding 40 kev γ-ray gave the most likely spin sequence $1 \rightarrow 4 \rightarrow 5$ for the cascade. Such an experiment was, in theory, capable of deciding the relative parities of the ThC and ThC″ levels by distinguishing whether the particles carried off 4, or a mixture of 3 and 5, units of angular momentum. However, the experimental results were not sufficiently accurate to distinguish between these possibilities and appeal was made to the theory of α fine structure to rule out the case $l_\alpha = 4$. The parities of the ThC and ThC″ states were therefore deduced to be different. The assignment of positive and negative parities to the ThC″ and ThC states, respectively, has been made solely on the basis of the shell-model level sequence. Evidently, the reverse parity assignment would make all the β decays allowed, with the exception of the highest energy ThC″ transition which would become second forbidden.

The shell model definitely favours the accepted parity assignments, for the proton outside the closed shell is almost certainly in a negative parity state, the nearest positive parity state being $i_{13/2}$. Similarly, the excess neutrons seem certain to be in positive parity states. An $i_{13/2}$ proton assignment is very unlikely in view of the well-established $9/2^-$ ground state of ^{209}Bi. In order to obtain negative parity states of ThC″, the proton hole would have to be $h_{11/2}$ and the ^{207}Tl ground-state assignment of $1/2^+$ is a strong argument against this.

The recent review of Gove (1966) indicates that, as the heavier mass regions are approached, the lg *ft* values of allowed decays tend to increase, while those of first forbidden decays tend to decrease. Many anomalously low (< 6) first forbidden lg *ft* values occur in the region of doubly closed ^{208}Pb, but there are some cases far removed from this region, e.g. ^{244}Am. It is doubtful, therefore, if the lg *ft* value is a useful criterion in the determination of degree of forbiddenness of β decays as Z increases to values of the order of 82. On the basis of lg *ft* value, there is little to choose between the parity possibilities mentioned above. The accepted scheme gives some first forbidden decays a somewhat low value; the alternative gives some allowed decays a high value together with a low value for the second forbidden transition in ThC″.

First forbidden β decays are expected to show some anisotropy in the β–γ angular correlation and a non-zero result would therefore rule out the hypothesis that the decays might be allowed. The following correlations were measured:

(i) The 350 kev β–239 kev γ in the ThB $\rightarrow$ ThC decay.
(ii) The 1·52 Mev β–729 kev γ in the ThC $\rightarrow$ ThC′ decay.
(iii) The 1·80 Mev β–583 kev γ in the ThC″ $\rightarrow$ ThD decay.

In addition, an attempt was made to repeat a previously reported correlation (Demichelis and Ricci 1956) using the 2·38 Mev β group of ThC″ and the 2·614 Mev γ-ray of ThD, which has resulted in a 4^+ assignment to the ThC″ ground state, contrary to the accepted one.

2. Apparatus and experimental method

The angular correlation chamber was made from a 15 cm high, 15 cm diameter brass cylinder of 0·3 cm wall thickness with the central 5 cm portion of wall turned down to 0·15 cm thickness. The β detector, a cylindrical piece of NE 102A plastic scintillator of 2·2 cm diameter and 1 cm high, coupled to a photomultiplier, was set into the chamber wall by means of a flange. The γ detector, a 5·1 × 5·1 cm NaI(Tl) crystal, was mounted, together with the photomultiplier tube, on a collar surrounding the chamber. Measurements were performed at a pressure of approximately 10^{-3} torr, achieved by means of a backing pump.

The electronics was of standard type. In each case, negative pulses were taken from the photomultiplier anode and fed through a cathode follower, amplifier and single-channel pulse-height analyser to a coincidence unit and scalers. The coincidence resolving time, measured by the independent source method, was $0·56 \pm 0·01$ μs.

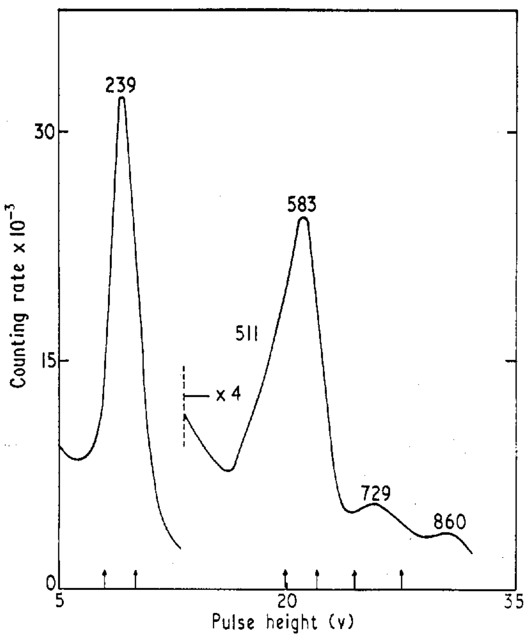

Figure 2. Low-energy γ spectrum of thorium active deposit. Energies are in kev.

Figures 2 and 3 show the γ spectrum of the active deposit together with an indication of the settings at which the various correlations were performed. A source of ^{207}Bi, which shows conversion peaks at 480 and 974 kev, was used to calibrate the β channel. A calibration was performed before and after every experimental run to check electronic stability. The drifts found were small and there was no need to correct for them.

The sources were collected electrostatically, from a dry emanating parent source of ^{228}Th, on 2 mm diameter, 1 mg cm^{-2} thickness, aluminium foils. These were then transferred to the centre of a backing sheet of 1 mg cm^{-2} aluminium foil clamped in a 5 cm diameter aluminium ring source holder suspended from the chamber lid. The sources so made were in 'sandwich' form and there was no danger of contamination of the chamber walls resulting from recoil nuclei following α decay. The sources decay (eventually) with a 10·6 hour half-life and were therefore made daily. The time, after removal from the source pot, which elapses before the sources begin to decay with the ThB half-life was calculated from the usual set of decay equations and sources were not used until this situation obtained.

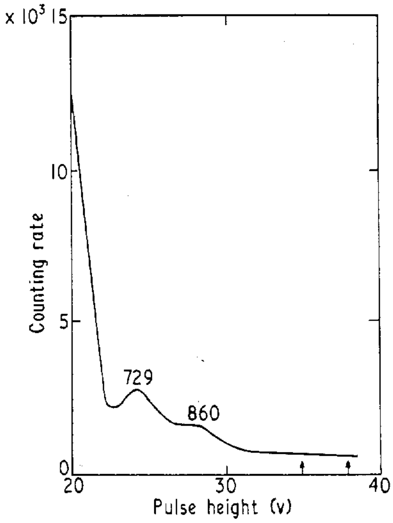

Figure 3. Detail of γ spectrum showing background from the 2·614 MeV γ-ray.

The sources were centred by trial and error using adjusting screws on the chamber lid. Counts were taken at relative counter angles of 90°, 180° and 270° and were of 5 minutes duration. The coincidence counts, after subtraction of chance coincidences, were normalized using ratios of γ single-channel counting rates, thus making correction for source decay and (to first order) for source misalignment. γ–γ coincidence rates were determined by placing an aluminium absorber of appropriate thickness between source and β detector.

The γ spectrum shows that there was a roughly constant background arising from the Compton distribution of the 2·614 MeV γ-ray of ThD. Since the entire β feed of ThC″ is in coincidence with this γ-ray, there are, unavoidably, coincidences of the type β–unobserved γ or γ cascade–2·614 MeV γ-ray. An angular correlation was therefore performed using a section of this background in order to correct for any anisotropy arising from this source. An anisotropy was found which was large enough to account for a slight anisotropy found in the β–729 keV γ correlation, but which was not large enough to influence, within the available statistical uncertainty, the other correlations performed. The data were corrected to account for finite detector size and the effect of finite source backing thickness (Yates 1965, Gimmi *et al.* 1956). The possibility of perturbation of the correlations was considered but was found to be unlikely on account of the level lifetimes involved.

Using a source of ^{152}Eu identical in size to the thorium sources, a β–γ angular correlation was performed using the 1·47 MeV β group and the 344 keV γ-ray of ^{152}Gd. β particles of energy greater than 1 MeV were used and the value of the $P_2(\cos\theta)$ coefficient was $-0{\cdot}32 \pm 0{\cdot}03$. This lies within the range of values obtained by other workers (Frauenfelder and Steffen 1965), and was accepted as evidence of the satisfactory performance of the apparatus.

3. Results and discussion

The angular correlation may be written as

$$\begin{aligned} W(\theta) &= 1 + a\cos^2\theta \\ &= 1 + \epsilon\, P_2(\cos\theta) \end{aligned}$$

where a and ϵ are related by

$$\epsilon = \frac{2a}{3+a}$$

and a is given in terms of the corrected coincidence totals by

$$a = \frac{N(180)-\{N(90)+N(270)\}}{N(90)+N(270)}$$

in an obvious notation. (A count was performed in the 180° position for each of those in the 90° and 270° positions). The values of ϵ in the various correlations are shown in table 1.

Table 1

γ-ray energy (kev)	β energy range (kev)	ϵ
239	250 → 350	$-0{\cdot}002 \pm 0{\cdot}01$
729	> 800	$0{\cdot}007 \pm 0{\cdot}02$
583	> 800	$-0{\cdot}007 \pm 0{\cdot}01$
583	>1000	$-0{\cdot}005 \pm 0{\cdot}03$

The attempt to repeat the correlation of Demichelis and Ricci (1956) was a failure. A completely negligible number of genuine coincidences was found when the β channel was set to record β particles of energy greater than 2·25 Mev and the γ channel was set on the 2·614 Mev photopeak. The experiment was not considered feasible with the present source strength of approximately $\frac{1}{2}$ mCi. Since the source strength quoted by Demichelis is only 45 μCi, it is difficult to see what was being measured. It is unlikely that the different β energy selection (> 2·25 Mev as against > 1·80 Mev by Demichelis) can explain the discrepancy in coincidence rates.

The null result of the β–239 kev γ correlation is acceptable as a corroborative piece of evidence in favour of the zero spin assignment to the 239 kev level of ThC. The result is in agreement with an earlier β- conversion electron angular correlation of Siegbahn (1952).

The β–729 kev γ correlation was of integral, rather than differential, type because the β feed to the 729 kev level of ThC′ is weak. The β energy discriminator setting of 800 kev was selected because the partial β spectrum leading to the next higher level of ThC′ has an end point energy of 750 kev. Thus, cascades of the type β–unobserved $\gamma-729$ kev γ did not contribute to the correlation. There will be, unavoidably, a few coincidences from the 860 kev γ-ray Compton distribution but the γ channel setting encompassed a local minimum in this distribution.

The problem of competing cascades arose again, but in a much more severe form, in the β–583 kev γ correlation. The most important competing cascade, namely the β–unobserved 511 kev γ–583 γ, could have been eliminated by setting the β channel to record β particles of energy greater than 1·3 Mev. This was attempted, but the number of coincidences was greatly reduced and the further possibility of contributions from the β–unobserved 277 kev γ–583 kev γ cascade still remained. It is possible to correct for the effect of competing cascades (Frauenfelder and Steffen 1965) but the method requires measurements of the β–511 kev γ correlation, which were not possible using the present γ detector. The best that could be done was to vary the β energy, so altering the contributions of competing cascades, and see if the result varied. No variation was found, within the statistical accuracy.

The present set of null results, although admitting the possibility that the β decays in question are allowed, cannot rule out the possibility that the decays are first forbidden. The lack of correlation could be a result of the integral nature of the measurements, for the correlation is energy dependent, in general increasing with β-particle energy. The lower-energy portions of the spectrum necessarily receive the greatest weights in an integral measurement and a possible small correlation can be obscured. The comprehensive summary of experimental results given by Frauenfelder and Steffen (1965) shows cases where this effect has occurred. This summary also reveals that the differential correlation sometimes shows no anisotropy in well-established cases of first forbidden decay. It is worth remarking that two such cases (^{203}Hg and ^{199}Au) have high Z values and low lg ft values.

Acknowledgments

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