The Alpha-Gamma Angular Correlation in Radiothorium*

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The alpha-decay of radiothorium is followed by an 83.3-kev gamma-ray whose angular correlation with the alpha-particle was investigated, using scintillation counters and fast coincidence circuits. The correlation curve does not agree with any of the theoretical ones derived under the assumption that the angular momenta of the three states involved are $0-J-0$. Possible reasons for this somewhat unexpected result are discussed.

I. INTRODUCTION

THE alpha-gamma correlation occurring in the decay of radiothorium has been examined by two different groups of workers.[1,2] The earlier experiment cannot be regarded as reliable, since no attempt was made to separate the radiothorium from its daughters. This present work was under way at the time the second measurement was reported. Inasmuch as the results of the latter experiment were rather surprising, it seemed worth while to carry this present work to its conclusion.

For some time it has been supposed that the energy levels of ThX consisted of an 86.8-kev and a 3.5-kev excited state in addition to the ground state. This was based on the report of two gamma-rays by Surugue and Tsien-San-Tsiang[3] and by Riou,[4] and also from the study of the alpha-fine structure by Rosenblum,[5] who indicated a possible slight broadening of the ground state alpha-group consistent with the existence of a 3.5-kev excited state.

However, a more recent study of the gamma-rays by Riou[6] based on a more careful separation of the decay products of radiothorium indicates that only the 83.3-kev gamma-ray is to be attributed to radiothorium decay, the 86.8-kev gamma occurring in the derivatives. The results of the present work tend to confirm this contention. It is then assumed that only one excited state of ThX occurs with an appreciable intensity in the alpha-emission of radiothorium, and that this 83.3-kev excited state decays to the ground state by the emission of one gamma-ray. It is hoped that the study of the angular correlation between the short-range alpha and the 83.3-kev gamma will shed some light on the angular momentum of the nuclear levels involved.

II. PREPARATION OF SOURCES

Radiothorium was separated from its decay products by alternate hydroxide precipitations of radiothorium, which left ThX in solution, and sulfide precipitations which removed ThB and ThC. The purified radiothorium plus the carrier was made into a suitable alpha-source by a modification of a technique described by Tödt,[7] in which radiothorium and thorium are electrolytically deposited onto a rhodium foil from an acid solution at high current density. In the region of the cathode where the solution has become alkaline due to a vigorous evolution of hydrogen, the thorium forms a hydroxide and is deposited as such on the cathode. A rhodium foil of one-half mil thickness was selected as a cathode since it does not absorb the gamma-radiation too severely and serves as a suitable backing material.

III. APPARATUS

The source was placed in a vacuum chamber. The alphas were detected by a three-mil thick anthracene flake mounted in the chamber with the source and observed by the phototube through a Lucite window. The gammas were detected with a sodium iodide crystal. Both detectors showed good energy resolution. A pulse-height spectrum of the alpha- and gamma-counters showed 20 percent and 16 percent resolution, respectively, at half-maximum for the radiations observed.

The pulses from both phototubes were sent through fast amplifiers to a fast coincidence circuit (resolution about 1.5×10^{-8} sec), and also through linear amplifiers to channel discriminators. A relatively slow coincidence circuit (7 μsec resolution) was fed from the output of the channel discriminators. And finally, a coincidence between the output of the fast coincidence circuit and the coincidence between the channel discriminators was recorded. In this way an energy criterion was imposed on both radiations while good time resolution was maintained. The accidental coincidence rate was determined by the introduction of sufficient lengths of delay line in the fast circuit to remove all true coincidences and provided a background correction.

IV. RESULTS

The experiments performed were originally based on the older assumption of the presence of two gamma-rays

* This investigation was supported by the AEC.
[1] Kilchitsky, Latishev, and Buliginsky, Doklady Akad. Nauk, S.S.S.R. **64**, 57 (1949).
[2] Beling, Feld, and Halpern, Phys. Rev. **84**, 155 (1951).
[3] J. Surugue and Tsien-San-Tsiang, Compt. rend. **213**, 172 (1941)
[4] M. Riou, J. phys. et radium **11**, 185 (1950).
[5] Rosenblum, Valadares, and Perey, Compt. rend. **228**, 385 (1949).
[6] M. Riou, private communication.
[7] F. Tödt, Z. physik. Chem. **113**, 331 (1924).

of 86.8 kev and 83.3 kev, each in coincidence with the same short range alpha. Angular correlation data were taken with the source alone, and then approximately equal amounts of data were taken with a thallium absorber interposed in the gamma-ray beam. An 86.8-kev gamma has sufficient energy to eject the K electrons of thallium and is severely absorbed, whereas an 83.3-kev gamma does not have sufficient energy for this process and is not so severely absorbed. Thus, two correlation curves are obtained which would have the two alpha-gamma processes present in appreciably different proportions. Hence the knowledge of the attenuation of the two gamma-rays in the thallium absorber is sufficient to isolate the two alpha-gamma-processes.

Inasmuch as the geometry involved in an angular correlation is far from ideal for the measurement of absorption coefficients, it was decided that an experimental measurement of the attenuation of the two gamma-rays was preferable to a computed attenuation based on tabulated absorption coefficients. An 86.8-kev gamma-ray was simulated by an 87.8-kev gamma-ray occurring in the K capture in Cd^{109}, and an 83.3-kev gamma by an 84.1-kev gamma occurring in the beta-active $thulium^{170}$, where appropriate corrections were made for the bremsstrahlung continuum from the beta-decay.

Considerable care was expended in lapping the thallium absorber to a thickness of 0.241 g/cm² uniform to 2 percent, so that the measurements would be independent of the exact area of the absorber used. The absorber was made somewhat larger than the minimum size necessary to cover the detector as insurance against imperfect alignment.

Values for the attenuation of the assumed radiothorium gamma-rays in the thallium absorber were then obtained by appropriate corrections for the small differences in energy between the radiothorium gamma-rays and the gamma-rays used for calibration. The attenuation for an 83.3-kev gamma-ray was found to be 0.622 ± 0.009. This corresponds to an absorption coefficient that is somewhat smaller than the tabulated value, due primarily to imperfect geometry. The attenuation for an 86.8-kev gamma-ray was found to be 0.279 ± 0.004. This corresponds to an absorption coefficient that is appreciably smaller than the tabulated value, due primarily to the detection of the fluorescent K-radiation from the thallium absorber, again because of the poor geometry.

Assuming that there are two gamma-rays following the short-range alpha of radiothorium, the relative intensities of these two transitions can be directly obtained from the correlation data with and without absorber and from the values of the gamma-ray attenuations. The experimental value of the attenuation of the coincidence rate, integrated over all angles, is 0.646 ± 0.017. This coincides with the value for the 83.3-kev gamma-ray of 0.622 ± 0.009, within the experimental error. This is consistent with Riou's result that there is only one gamma-ray present in the decay of radiothorium, that is, the 83.3-kev gamma-ray.

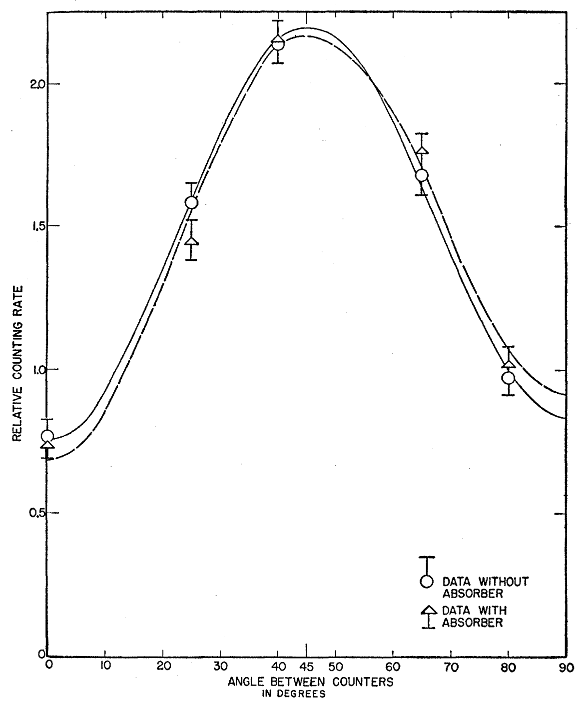

FIG. 1. Experimental data with and without absorber. The solid curve represents the least square fit to the experimental angular correlation data without absorber, the dotted curve with absorber. The solid angles of both counters are a 3×10^{-3} fraction of the total sphere. The curves are normalized to the same area, where the correlation with absorber is $W(\theta)=0.91+5.37\cos^2\theta-5.60\cos^4\theta$, and the correlation without absorber $W(\theta)=0.83+5.55\cos^2\theta-5.63\cos^4\theta$.

A separate experiment designed to detect gamma-gamma coincidences in the 80- to 90-kev range was also performed with a circuit of 2 μsec resolution. No coincidences were detected, in agreement with the findings of Beling *et al.* This result, however, is also in agreement with the assumption of only one gamma-ray.

The growth of the decay products of radiothorium is controlled by the 3.64-day half-life of its first daughter, ThX. The angular correlation data were taken with radiothorium sources that were not more than sixteen-hours old. Although the single channel rates increased continuously throughout the experiment, the coincidence rate did not, indicating that the decay products were not contributing any coincidences to the correlation. The data are shown in Fig. 1. These were taken with the counter solid angles equal to a 3×10^{-3} fraction of the total sphere. The curves represent a least square fit to the data. Within the experimental error, the two curves may be regarded as representing the same correlation, which is again consistent with the assumption of only one gamma-ray.

As Beling *et al.* point out, lifetime arguments exclude the possibility of the gamma-radiation being of higher

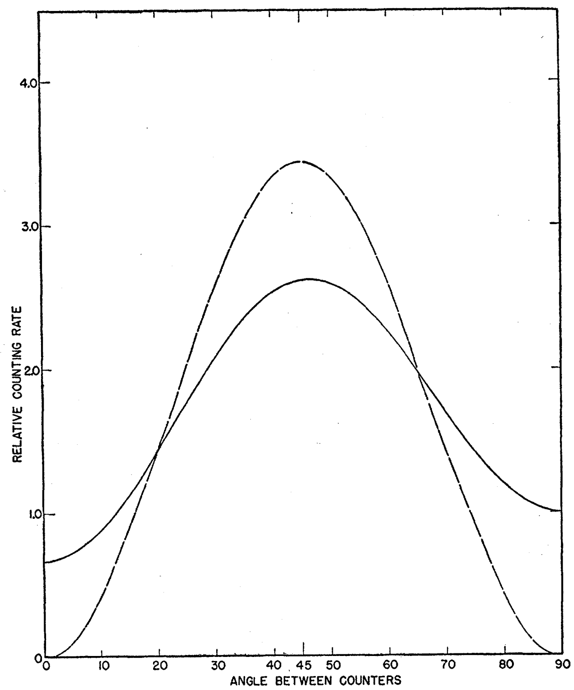

FIG. 2. The solid angular correlation curve represents the average, after reduction to zero solid angle, of the least square fit to the data with and without absorber. The dotted curve corresponds to a $0-2-0$ sequence and is plotted for comparison. The curves are normalized to the same area.

multipolarity than quadrupole. This in turn limits the complexity of the distribution to $\cos^4\theta$. The argument is even stronger with our shorter resolving time of 1.5×10^{-8} sec. The presence of the large $\cos^4\theta$ term indicates that the gamma-radiation is at least quadrupole.

If the least square curves are corrected to zero solid angle, the angular correlation without absorber becomes $W(\theta)=1.00+7.27\cos^2\theta-7.48\cos^4\theta$, and the angular correlation with absorber becomes $W(\theta)=1+6.39\cos^2\theta-6.76\cos^4\theta$. The average of these two curves weighted equally is $W(\theta)=1+6.83\cos^2\theta-7.12\cos^4\theta$. This agrees rather closely with the results of Beling *et al.* for their correlation without absorber, that is $W(\theta)=1+6.90\cos^2\theta-7.07\cos^4\theta$. Our curve is shown in Fig. 2.

CONCLUSIONS

Inasmuch as the nuclei involved have an even number of neutrons and protons, it might be expected that the ground states would have zero angular momentum. If the gamma-radiation is quadrupole, then the sequence of angular momentum states would be $0-2-0$. However, the experimental data do not fit $W(\theta)=\cos^2\theta-\cos^4\theta$, the correlation shown in Fig. 2, which corresponds to this sequence, nor does it fit any other $0-J-0$ sequence. Some general remarks should be made in this connection.

1. The possibility of impurities from other disintegration series in the radiothorium sources, that could contribute false coincidences, was rejected as highly improbable, because the radiothorium supply was separated from MsTh I some months in advance of taking any correlation data. The only contaminant that is usually found with MsTh I is radium, and this would be removed from the radiothorium with MsTh I. The decay products of radium which would give coincidences would have died out before the correlation data were taken. On the other hand, if the decay products of radiothorium are changing the correlation, then the coincidence rate should increase and the shape of the correlation change with time. Neither of these things occurred.

2. It is possible that an atomic magnetic field could alter the population of the magnetic substates of the intermediate nucleus and thus change the correlation. However, it is difficult to believe that there are magnetic fields of electronic origin in the solids used, which are strong enough to reorient the nucleus in 10^{-8} sec. The value 10^{-8} sec is used as an upper limit for the lifetime, since no coincidences above the accidental rate were observed when a delay line of 2×10^{-8} sec was introduced in one of the channels. It is also interesting to note that our source was composed of thorium hydroxide, and the source prepared by Beling *et al.* was iron chloride as a carrier for thorium. Hence, it appears that similar results were obtained with different solid compounds, which provides an additional argument against reorientation.

3. There still remains the possibility that the proposed disintegration scheme is not correct. However, at the present time there is no direct evidence for other modes of decay. It would therefore be of considerable interest to determine whether other gamma-rays whose energy is very close to 83 kev exist and are in coincidence with the short-range alpha.

4. If it is assumed that the disintegration scheme is simple, that is, having one gamma-ray, that no impurities are contributing coincidences, and that the intermediate state is not being reoriented, then the assumption that the angular momenta of both ground states are zero must be rejected. However, none of the angular momentum assignments in which the selection rules operate to limit the alpha-particles to one angular momentum group followed by quadrupole gamma-radiation seem to fit the data very well. An example which is not too remote from a fit but nevertheless outside the probable error, is the sequence $1-2-1$ with $L=2$ in both transitions. This gives $W(\theta)=1+5\cos^2\theta-5.33\cos^4\theta$. In computing alpha-gamma correlations, mixtures of the different alpha-angular momentum groups that are consistent with angular momentum and parity selection rules must be considered.[8] This introduces the parameters of the relative intensities of the different alpha-angular momentum groups and the phase angles between their matrix

[8] S. P. Lloyd, Phys. Rev. 85, 904 (1952).

elements into the correlation expression. The systematic investigation of assignments using mixtures of alpha-angular momentum groups involves much computation. None of the cases that involve mixtures of angular momentum groups for the alpha-particle have been computed here; however, Beling[9] has considered a few without finding a convincing fit with the data.

[9] J. K. Beling, dissertation, Massachusetts Institute of Technology (1951), unpublished.

We are indebted to Professor I. Halpern and his colleagues for communicating to us the results of their investigation. We also wish to thank Professor M. Riou for sending us his most recent unpublished results on the gamma-radiation from the decay of radiothorium.[10]

[10] *Note added in proof.*—A recent paper by Rosenblum, Valadares, and Guillot (Compt. rend. **235**, 238 (1952)) reports measurements of the internal conversion lines of radiothorium, which confirm the existence of one gamma-ray of 83.4 kev as assumed in the interpretation of our data.

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Entropies of Activation in Metallic Diffusion*

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The various contributions to the entropy of activation for metallic diffusion are examined theoretically. In particular, the entropy contribution arising from the altered vibrational frequencies around the saddle point is estimated. It is concluded that negative over-all entropies of activation are theoretically permissible and are likely to arise when the over-all activation energy is small.

I. INTRODUCTION

The temperature dependence of volume diffusion is accurately described by the simple exponential relation

$$D = D_0 e^{-E/RT}, \quad (1)$$

where D = diffusion constant; D_0 = constant; E = activation energy; R = gas constant; T = temperature in °K. The activation energy E may be estimated theoretically in simple cases.[1] It is much more difficult to give a sound theoretical interpretation to D_0, the so-called frequency factor, although many attempts have been made in the past to derive theoretical expressions for this quantity.[2]

This problem was attacked again in the last few years with clarification of some of the puzzling features. The writer proposed an empirical correlation which related D_0 to the quantity E/T_m, where T_m is the melting point in °K, and suggested that E/T_m be identified with the entropy of activation.[3] Zener[4] proposed a theory for D_0, which seemed to account at least partly for the above correlation, in which only positive experimental entropies of activation ΔS, calculated as

$$\Delta S/k = \ln(D_0/\nu a^2), \quad (2)$$

were permitted (ν = Debye frequency, a = lattice parameter). In Zener's theory, the origin of the entropy contribution is traced to the temperature coefficient of the work required to produce a given distortion within the lattice. Finally, this was traced to the temperature coefficient of the elastic moduli, which is negative, yielding a positive ΔS. Zener suggested that experimentally observed negative entropies of activation are to be attributed to experimental inaccuracies or to the presence of short-circuiting diffusion paths.

The writer pointed out[5] that Zener's theory is unable to account for the large negative entropies of activation for self-diffusion in tin as calculated from Fensham's excellent experimental data on single crystals.[6] At about the same time Nowick[7] published an article in which intermetallic diffusion experiments were reinterpreted on the basis of Zener's theoretical expression for D_0. Nowick argues that this theoretical D_0 is correct at least to order of magnitude and suggests that many experimental activation energies are greatly in error at any but the highest temperatures because of diffusion along internal surfaces or "short circuiting paths" and that the writer's earlier correlation reflects essentially this inaccuracy. Nowick is certainly correct in calling attention again to the well-known poor quality of most diffusion data. However, he offers no explanation for the behavior of tin. Further, recent researches by Smoluchowski and co-workers[8,9] indicate strongly that dis-

* Research carried out under contract with the AEC.

[1] See for example the recent review by F. Seitz in *Phase Transformations in Solids* (John Wiley & Sons, Inc., New York, 1951), pp. 77–149.

[2] See for example A. D. LeClaire, section on "Diffusion of Metals" in *Progress in Metal Physics I* (Interscience Publishers, Inc., New York, 1949), pp. 306–379.

[3] G. J. Dienes, J. Appl. Phys. **21**, 1189 (1950).

[4] C. Zener, J. Appl. Phys. **22**, 372 (1951); *Theory of Diffusion*, "Imperfections in Nearly Perfect Crystals," (John Wiley and Sons, Inc., New York, 1952), pp. 289–314.

[5] G. J. Dienes, J. Appl. Phys. **22**, 848 (1951).

[6] P. J. Fensham, Australian J. Sci. Res. **A3**, 91 (1950); **A3**, 105 (1950).

[7] A. S. Nowick, J. Appl. Phys. **22**, 1182 (1951).

[8] R. Smoluchowski, Phys. Rev. **87**, 482 (1952).

[9] R. Flanagan and R. Smoluchowski, J. Appl. Phys. **23**, 785 (1952).