

Directional Distribution of Alpha Emission in Th^{228} Decay Series

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Dedicated to Professor Dr. W. GENTNER on the occasion of his 60th birthday

Directionality of alpha emission in terms of alpha-alpha angular correlation distributions between the consecutive alpha particles in the serial decay of Th^{228} is experimentally studied from Thorium stars produced in emulsion plate. The angular correlations fit a $\sin \theta$ -distribution.

Alpha emission in terms of angular momentum has been given by ROSE¹ in a theoretical expression specifying the angular distribution of alpha particles in terms of nuclear matrix elements. HILL and WHEELER² without considering the angular momentum, have shown that the nonspherical shape of a nucleus has an effect on alpha emission. They pointed out that in the case of a prolate spheroidal nucleus the potential barrier along the nuclear poles would be lower and thinner than that along the nuclear equators and naturally a strong preferential emission of alpha would take place along the nuclear poles. In particular, in even-even nuclei with total angular momentum zero in the ground state but with intrinsic quadrupole moment Q_0 , alpha emission would be isotropic in the laboratory fixed reference but anisotropic with reference to the intrinsic nuclear symmetry axis. HANAUER et al.³ measured the angular distribution of alpha particles emitted by oriented Np^{237} nuclei. They concluded from their measurements on Np^{237} that alpha-particles are emitted preferentially along the nuclear angular momentum vector. This conclusion is independent of any assumption about nuclear shape or the mechanism of alpha particle emission. Under the assumption that the nuclear angular momentum vector is oriented along the nuclear symmetry axis, it appears from their experimental results that the alpha particles are emitted preferentially along the nuclear symmetry axis. For a prolate nucleus, the preferential emission is from the polar regions which is in agreement with the prediction of HILL and WHEELER. A rigorous theoretical treatment of alpha

emission considering both angular momentum and nuclear shape have been presented by RASMUSSEN and SEGALL⁴, BRUSSAARD and TOLHOEK⁵, STEENBERG and SHARMA⁶, and FRÖMAN⁷. It is generally expected that the potential barrier is the dominating factor in the anisotropy of alpha emission from nonspherical nuclei.

It seems that nuclei with mass number greater than ~ 220 have a stable spheroidal deformation and give rise to a rotational band. In this respect Th^{228} has a stable spheroidal deformation of the prolate type. It decays through a series of α and β emissions as shown below to Pb^{208} which is a closed shell spherical nucleus. For convenience alphas emitted from each step are named in general as α_1 , α_2 , α_3 etc. without consideration of the emission of certain percentages of D-wave or G-wave as in Th^{228} and in Ra^{224} instead of an abundant S-wave. (In Th^{228} : 71% 0^+ , 28% 2^+ , 0.2% 4^+ ; Ra^{224} : 95% 0^+ , 4.8% 2^+ ; Rn^{220} : 99.7% 0^+ , 0.3% 2^+ ; Po^{216} : 99.86% 0^+ ; Po^{212} : 99.97% 0^+).

It can be thought that α_1 and α_2 are emitted one after the other from the nucleus Th^{228} at a very long interval of time depending on its half life. If an alpha particle has the possibility of being emitted from a nonspherical even-even nucleus having intrinsic quadrupole moment Q_0 at an angle θ_1 with respect to the intrinsic nuclear symmetry axis, and the next alpha with an angle θ_2 with respect to the same axis, then a correlation distribution between the two alphas might arise. The angular correlation distribution between the successive alpha particles in such a cascade decay would be rather interesting in order

¹ M. E. ROSE, Elementary Theory of Angular Momentum, John Wiley & Sons, Inc., New York 1957, pp. 176—186.

² D. L. HILL and J. A. WHEELER, Phys. Rev. **89**, 1102 [1953].

³ S. H. HANAUER, J. W. T. DABBS, L. D. ROBERTS, and G. W. PARKER, Phys. Rev. **124**, 1512 [1961].

⁴ J. O. RASMUSSEN and B. SEGALL, Phys. Rev. **103**, 1298 [1956].

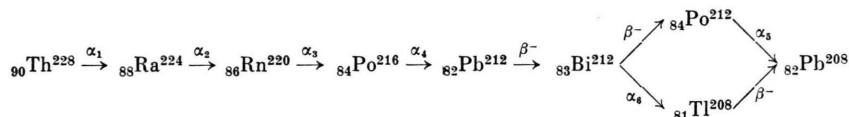
⁵ P. J. BRUSSAARD and H. A. TOLHOEK, Physica **24**, 233 [1958].

⁶ N. R. STEENBERG and R. C. SHARMA, Can. J. Phys. **38**, 290 [1960].

⁷ P. O. FRÖMAN, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Skrifter **I**, (No. 3) [1957].



to see how the spheroidal shape of the Th^{228} -nucleus attains the spherical one. In 1936 WILKINS⁸ measured



Measurement of angles

the angles between pairs of alpha tracks in Radium and Thorium stars produced in emulsion plates. He observed that in radium-plate Radium C' of length 30 grains makes a characteristic angle of 110° with an accompanying track of length 20 grains. A pair each of length 13 grains making an angle of about 170° seems to occur frequently. In the thorium plate characteristic groups have also been found by him, but the typical angle between the pairs was not mentioned in his work. At that time the technique of measurement in emulsion plates was not highly developed. Under such circumstances, an investigation is being carried out to measure the angles between the successive alpha particles emitted in the serial decay of Th^{228} . That is the angles $\alpha_1 \alpha_2$, $\alpha_2 \alpha_3$, $\alpha_3 \alpha_4$ and $\alpha_4 \alpha_5$ are measured. The angle $\alpha_4 \alpha_6$ is also measured, though α_6 transition is not an even-even case. In this report details of the experimental technique are given and results of preliminary nature are represented in a qualitative way, deferring the final conclusion at a later time.

Experiment

Production of alpha stars of thorium in emulsion plates

In order to record the alpha stars produced by decay of Th^{228} and its daughter members, K0-plates (Ilford) are exposed to thorium nitrate solution containing 0.982% of thorium in (N) HNO_3 solution. The p_H value of the solution is adjusted to nearly 5–6 by adding ammonium acetate to it. The plates are soaked with the solution at $\sim 68^\circ\text{F}$ for 10 minutes and rinsed in distilled water and then left in a light-tight box for 13 days for the production of latent images of the stars. The plates are then processed by the metol-hydroquinone developer recommended by M/s Ilford Ltd. The processing steps are conducted at $\sim 68^\circ\text{F}$ as follows: presoaking for 10 min, developing for 25 min, rinsing for 2 min, keeping in stop bath (2% acetic acid) for 12 min, rinsing for 2 min, fixing in hypobath (30%) for 2 hrs and final washing in distilled water for 3 hrs. The procedure is carried out in a room illuminated by orange-coloured safelight. After washing the plates are kept for 12 hrs in a dust-free closed chamber for drying.

Stars which originated from a common centre, all prongs of which ended within the emulsion, are selected. The tracks of most of the stars originated from a common centre as observed within the limitations of microscopic view, the recoil of the daughter nucleus being too small to produce any observable displacement in the emulsion. One prong track to five prong stars have been obtained. In some cases, due to diffusion of gaseous radon (thoron) its subsequent decay-alphas are found to be displaced from the previous origin and to form another centre. All such stars are discarded. As far as possible only stars free from distortions are selected. Ranges of the track of the selected stars are measured in a Leitz microscope (type Ortholux) with a total magnification $850\times$ in a combination of an eyepiece micrometer ($12.5\times$) and an oil immersion objective ($53\times$). The alpha particles from each decaying nucleus are identified from the range-energy relation. The distribution of the ranges of the alpha particles emitted from each nucleus are first assumed as given by POWELL et al.⁹. On this basis several hundreds of particles emitted from each nucleus are measured and histograms of 600 particles for each group are drawn. From these histograms, the range distributions of alphas from each nucleus are obtained in these measurements. The ranges are given in Table 1.

Name of nuclei emitting particular alpha particles	Ranges in microns	Mean range in microns
$\text{Th}^{228} (\alpha_1)$	19.75 ± 0.07 to 25.00 ± 0.09	23.25 ± 0.08
$\text{Ra}^{224} (\alpha_2)$	21.50 ± 0.08 to 27.25 ± 0.10	25.25 ± 0.09
$\text{Rn}^{220} (\alpha_3)$	27.50 ± 0.10 to 30.75 ± 0.11	29.10 ± 0.10
$\text{Po}^{216} (\alpha_4)$	30.25 ± 0.11 to 35.20 ± 0.13	32.30 ± 0.12
$\text{Po}^{212} (\alpha_5)$	44.75 ± 0.16 to 51.20 ± 0.18	47.80 ± 0.17
$\text{Bi}^{212} (\alpha_6)$	25.50 ± 0.09 to 28.80 ± 0.10	27.95 ± 0.10

Table 1. Observed ranges in emulsion of alphas emitted from different nuclei.

⁸ T. R. WILKINS, Phys. Rev. **49**, 639 [1936].

⁹ C. F. POWELL, P. H. FOWLER, and D. H. PERKINS, The Study of Elementary Particles by the Photographic Method, Pergamon Press, London 1959, p. 138.

Angles between the consecutive alpha particles such as $\hat{\alpha}_1\alpha_2$, $\hat{\alpha}_2\alpha_3$, $\hat{\alpha}_3\alpha_4$, $\hat{\alpha}_4\alpha_5$ and $\hat{\alpha}_4\alpha_6$ are measured with the help of an eyepiece goniometer ($12.5\times$) capable of measuring up to one minute. The angles are converted into space angles with the help of a WULF's stereographic projection net graduated at two degree interval.

Results

The ranges of α_3 and α_6 , though each having a different mean value, overlap each other in certain portions. To avoid this confusion the angles $\alpha_2\alpha_3$ and $\alpha_3\alpha_4$ measured from the stars produced through the branch Po^{212} and through the branch Tl^{208} are separately analysed. A total of 3346 angles has so far been measured. 2300 angles are utilised for angular distributions taking $\hat{\alpha}_2\alpha_3$ and $\hat{\alpha}_3\alpha_4$ from the stars produced only through the branch Po^{212} . The correlations of the angles $\hat{\alpha}_1\alpha_2$ and $\hat{\alpha}_4\alpha_6$ are excluded in the present report. The distributions of $\hat{\alpha}_2\alpha_3$, $\hat{\alpha}_3\alpha_4$ and $\hat{\alpha}_4\alpha_5$ are normalised to unity at 90° and are shown in Fig. 1, 2, 3 respectively with singlefold statistical fluctuations applied to the experimental points. The experimental distributions $\hat{\alpha}_2\alpha_3$, $\hat{\alpha}_3\alpha_4$

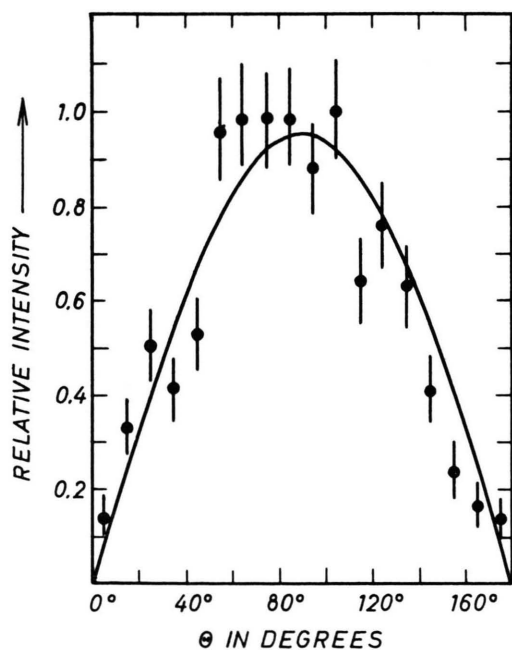


Fig. 1. α_2 — α_3 angular correlation distribution. For α_2 , α_3 see decay scheme. Continuous curve is $0.95 \sin \theta$ distribution.

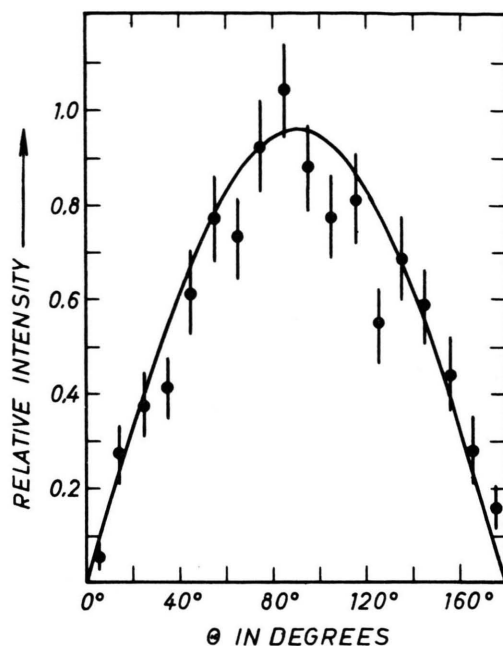


Fig. 2. α_3 — α_4 angular correlation distribution. For α_3 , α_4 see decay scheme. Continuous curve is $0.96 \sin \theta$ distribution.

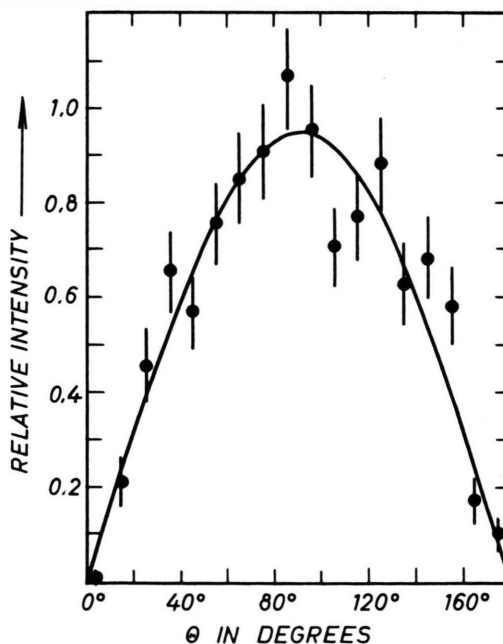


Fig. 3. α_4 — α_5 angular correlation distribution. For α_4 , α_5 see decay scheme. Continuous curve is $0.95 \sin \theta$ distribution.

and $\hat{\alpha}_4\alpha_5$ are fitted with theoretical curves $0.95 \sin \theta$, $0.96 \sin \theta$ and $0.95 \sin \theta$ respectively as shown in the diagrams.

Discussion

In this preliminary survey, it is only remarked that the angular correlations between the two groups of alpha particles α_2 and α_3 emitted from the nuclei Ra^{224} in succession, between α_3 and α_4 from Rn^{220} nuclei and between α_4 and α_5 from Po^{216} after two intermediate β -decays fit nearly a $\sin \Theta$ -distribution.

In $\alpha_2 \alpha_3$ distribution there is 4.8% D-wave admixture to the S-wave in α_2 in the decay of Ra^{224} . Rn^{224} has a stable prolate¹⁰ spheroidal deformation with $Q_0 = +11.6 \times 10^{-24} \text{ cm}^2$ giving rise to a rotational band. The last alpha-decaying nucleus Po^{212} is nearly spherical with¹¹ $Q_0 = 2 \times 10^{-24} \text{ cm}^2$. How far this observed anisotropy is in accord with the predic-

tion by HILL and WHEELER of the anisotropy in alpha emission from even-even nuclei due to intrinsic quadrupole moment awaits quantitative estimations. Once an alpha particle would be directionalised to some spot on the nuclear surface, its subsequent passage to the exterior region is equivalent to its potential scattering. It appears that after the emission of an alpha particle the next one would have the maximum possibility to be seen at an angle of 90° from the first one. It might possibly be conjectured if a correlation of the emission zones on the surface of such nuclei in these decay series could be found from such information. A deeper understanding into the consequences of such results shall be reported later on.

¹⁰ Th^{228} is a prolate spheroidal nucleus (ref. 4). RASMUSSEN and SEGALL have pointed out that shell filling towards the equator takes place as A increases as in the case of Cm^{242} . It is accepted here that Ra^{224} is also a prolate nucleus.

¹¹ A. BOHR and B. R. MOTTelson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. 27, (16) 116 [1953].