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COMPLEX ALPHA SPECTRA OF RADIOTHORIUM (TH228) AND THORIUM-X (Ra224)

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ABSTRACT

The alpha and gamma spectra of Th^{228} have been studied with an alpha particle spectrograph and gamma-ray scintillation counters. Thorium 228 has alpha groups of 5.421 (71%), 5.338_5 (28%), 5.208 (0.4%), and 5.173 Mev (0.2%), and gamma rays of 89 (1.6%), 137 (0.26%), 169 (0.09%), and 212 kev (0.27%). Spins and parities are assigned to the energy levels defined by the alpha groups and the results are evaluated with respect to the developing theory and systematics of complex alpha spectra.

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I. INTRODUCTION

In recent communications[1, 2] we have shown that the energy levels reached by the decay of even-even alpha emitters fall into a regular pattern with respect to (1) energy level spacing, (2) spectroscopic designation of the states, and (3) the relative population of the states. Starting with the heaviest nuclei the energy levels of the low-lying states increase progressively with decrease in charge and neutron number. The low-lying states seem to be even spin-even parity states and in a limited region the relative energy spacings conform well with their assignments to even rotational states as suggested by Bohr and Mottelson.[3] It was also shown[2] that the second even excited state is populated to a much lower degree than the demands of simple alpha decay theory and that the degree of departure from expectations increases progressively with atomic number. As a possible explanation it was suggested that there is a progressive change in the charge asymmetry of the nuclei giving a spherically non-symmetrical potential barrier and that the alpha group in question is emitted in a direction in which it encounters a greater potential barrier than do the groups leading to the first excited state and the ground state.

Evidence already mentioned, as well as analysis of gamma ray conversion coefficients, all point to the rule that for the heaviest even-even nuclides the low-lying states all have even spin and even parity. This situation was analyzed[4] in detail for Pu^{238} (alpha decay product of Cm^{242})

and it was also shown that the first odd spin-odd parity state probably lies about 1 Mev above the ground state. The present paper on Th^{228} alpha decay shows the existence of the low-lying even spin states, but in addition, an odd spin state has dropped down so that it falls within the even spin states.

The two nuclides considered here (Th^{228} and Ra^{224}) are members of the thorium decay series and as such have been examined a number of times in the past in other laboratories. The points of similarity and differences between the present work and earlier measurements will be brought out in the discussion of the results.

II. METHODS

The Th^{228} and Ra^{224} were obtained from a mesothorium source of ~70 mg radium gamma ray equivalent. It will be seen that six alpha groups so far found are ascribable to these two isotopes and since the energies are close to each other, fairly rigorous chemical separations are required to assign the groups, particularly those in low intensity. Furthermore, possible interfering groups are at hand from the RaC(Bi^{212}) of this decay series as well as from members of the radium (Ra^{226}) series present as minor contaminants in the mesothorium. The problem of analysis hinges on the growth rates of the members of the series. Th^{228} (1.9 y half-life) can be obtained pure initially but Ra^{224} grows in with its 3.64-day half-life. Other members rapidly follow the Ra^{224} growth except Bi^{212} (60 min) which is held up slightly by the growth of its beta-emitting parent, 10.6 hr Pb^{212}. As may be visualized from these lifetimes, it is possible to make an alpha spectrum analysis of Th^{228} relatively free of decay products and of Ra^{224} with its products free of Th^{228}, but it is not possible to analyze Ra^{224} free of its decay products. If Ra^{226} is present in the mesothorium, its decay products, Em^{222} and Po^{210}, will produce alpha groups in the energy range of

interest; Bi^{214} is excluded from consideration because of its extremely low alpha branching.

The method of chemical separation made use of Dowex 50 ion exchange resin upon which the radium and thorium fractions were adsorbed and eluted selectively. The conditions differed considerably for the different preparations and will be mentioned when the separate determinations are discussed.

After chemical separation of a sample, its solution was evaporated on a tungsten filament and then vacuum sublimed onto a platinum plate masked to approximate a line source of alpha activity. The sample was then measured in the magnetic spectrograph. The techniques of source preparation and spectrograph measurements were described in earlier publications.[5, 4] An analysis was also made of the gamma ray spectrum of Th^{228} using a sodium iodide (thallium-activated) crystal detector and a 50-channel pulse analyzer.

III. RESULTS

The standard of reference for energy was the principal alpha group (α_o) of Ra^{224} which had been determined as 5.681 Mev by Briggs.[6] In some instances energies were determined more conveniently relative to Em^{222} (Rn) and Po^{210} after these were identified in the samples. Also other abundant groups of Ra^{224} and Th^{228} served as secondary standards. The data concerning the six alpha groups attributed to Th^{228} and Ra^{224} are shown in Tables 1-3. In these, the experiment number refers to the spectrograph exposure and the sample number identifies the particular alpha particle source whose preparation is described in the Appendix. Pertinent data on the exposures are also summarized there.

Alpha decay of Ra^{224}. -- The main alpha group of Ra^{224} (5.681 Mev) leading to the ground state of Em^{220} is readily identified and, as mentioned, has served as the energy standard for the present series of measurements. Rosenblum, Valadares, Perey, and Vial[7] have found an alpha group of Ra^{224} at 5.448 Mev in 4.6% abundance. Table 1 contains data identifying this same group. It is seen that its abundance relative to α_0 of Ra^{224} is reasonably constant in the different preparations and it bears no relation to the amount of Th^{228} present. The energy of this group is 5.445 Mev and its abundance (of total Ra^{224} alpha particles) is 4.9%; both values are in agreement with those of Rosenblum and co-workers.[7]

The energy difference between the two alpha groups of Ra^{224} as found here is 236 kev, therefore the second group leads to an excited state of 240 kev. No measurement of the gamma ray transition was made by us, but Rosenblum, Valadares, and Guillot[8] have observed K and L conversion lines of a 241 kev gamma ray and from the conversion coefficient deduced that the transition is of the E2 type. This conclusion designates the 240 kev state as 2+, the first even spin state. The corresponding excited state apparently appears in each even-even nucleus and the energies of these states follow a distinct pattern from the region of lead to the heaviest elements.[1, 9]

From the discussion of the Th^{228} spectrum which follows, it will be seen that no other alpha groups of Ra^{224} could be identified down to energies 884 kev below the main alpha group. This observation does not agree with the work of others.[7] On the basis of our measurements, the alpha decay scheme of Ra^{224} consists simply of the ground state transition in 95% abundance and the transition to the 240 kev 2+ state in 5% abundance. Any other alpha group within the range of our measurement is

in <0.1% abundance.

Table 1

Low energy alpha-group of Ra^{224} (5.445 Mev)
(Main alpha-group (α_0) at 5.681 Mev)

Experiment and Sample Numbers	Energy Reference	Energy (Mev)	Abundance Relative Ra^{224} α_0	Abundance Relative Th^{228} α's
Exp. 174, Sample 1	Ra^{224} (α_0)	5.441 ± 0.005	5.6%	>600%
	Em^{222}	5.445 ± 0.005		
Exp. 226, Sample 2	Th^{228} (α_{84})	5.445 ± 0.001	4.8%	1.6%
Exp. 241, Sample 3	Th^{228} (α_{84})	5.447 ± 0.002		
Exp. 258, Sample 4	Ra^{224} (α_0)	5.442 ± 0.002	5.2_5%	0.31%
Best Value		5.445	5.2%	

Alpha spectrum of Th^{228}. -- The principal alpha group of Th^{228} was measured as 5.421 ± 0.001 Mev, using Ra^{224} α_0 as the standard. The measurement was made on Sample 2, Exp. 225. In the same experiment, the group leading to the first excited state was observed and its energy was found to be 5.388_5 ± 0.001 Mev and its abundance 28%. The energy difference defines the first excited state at 84.3 kev. In another run (Sample 4, Exp. 259) the abundance of the group was found to be 26% and the energy level 84.9 kev. The data on the two principal groups of Th^{228} are in excellent agreement with those of Rosenblum, Valadares, and Perey[10] who reported energies of 5.423 and 5.338 Mev in exactly the same relative abundances as in our Exp. 225.

Besides these two high abundance groups, two others in low intensity were observed which we attribute to Th^{228}. Fig. 1 shows one spectrum taken covering the energy range from 5.15 to 5.71 Mev (Exp. 226, Sample 2). The solid curve applies to the 19 hour exposure and on this the groups α_0 and α_{84} of Th^{228} and α_0 of Ra^{224} were too intense to be shown on the same scale. The peaks for these groups shown as broken lines in Fig. 1 were obtained by making partial scans of the photographic plate and represent about 2.4% of the total tracks registered. It will be noted that groups due to Po^{210} and Em^{222} are on the plate. Other exposures on samples from which Po^{210} was absent and Em^{222} present in altered abundance helped confirm their assignments. An alpha group of Bi^{212} (5.603 Mev) falls within the energy range of these measurements but its estimated intensity would have been below our limit of detection.

Tables 2 and 3 summarize the data on the two low intensity groups of Th^{228}. These groups at 5.173 and 5.208 Mev bear constant relationship to the Th^{228} content of the sample and not to the amount of Ra^{224}. This observation is somewhat at variance with the results of Rosenblum and co-workers[7] who reported no groups of Th^{228} in this energy range but one at 5.194 Mev with Ra^{224} in 0.4% abundance. It will be seen that these two groups of Th^{228} fit in well with a proposed decay scheme which includes the gamma ray transitions. Aside from the Th^{228} groups already mentioned, an upper limit of 0.04% could be set for any in the energy range 4.80 → 5.67 Mev.

It will be noted (Table 3, Exp. 248) that $Ra^{226}\alpha_0$ was the standard for the energy determination of the 5.173 Mev group (α_{253}) of Th^{228}. The agreement of this energy with that using other standards was obtained using the revised energy for Ra^{226}, 4.777 Mev.[11]

Table 2

Assignment of 5.208 Mev group as α_{217} of Th^{228}

Experiment and Sample Number	Energy Reference	Energy (Mev)	Abundance Relative Th^{228} α_0 and α_{84}	Abundance Relative Ra^{224} α_0
Exp. 226, Sample 2	Th^{228} α_{84}	5.208 ± 0.001	0.45%	1.3%
Exp. 238, Sample 3	Th^{228} α_{84}	5.21 ± 0.10	≤0.5%	≤0.2%
Exp. 241, Sample 3	Th^{228} α_{84}	5.21	≤0.65%	≤0.6%
Exp. 258, Sample 4	Ra^{224} α_0	5.206 ± 0.001	0.42%	7.0%
Exp. 230, Sample 2	Po^{210}	5.209 ± 0.001		
Best value		5.208	0.4%	

Table 3

Assignment of 5.173 Mev group to α_{253} of Th^{228}

Experiment and Sample Number	Energy Reference	Energy (Mev)	Abundance Relative Th^{228} α_0 and α_{84}	Abundance Relative Ra^{224} α_0
Exp. 226, Sample 2	Th^{228} α_{84}	5.173 ± 0.001	0.22%	0.65%
Exp. 238, Sample 3	Th^{228} α_{84}	5.17 ± 0.02	≤0.2%	≤0.1%
Exp. 241, Sample 3	Th^{228} α_{84}	5.17	≤0.5%	≤0.4%
Exp. 258, Sample 4	Ra^{224} α_0	5.171 ± 0.002	0.19%	3.1%
Exp. 230, Sample 2	Po^{210}	5.174 ± 0.001		
Exp. 248, Sample 2	Ra^{226} α_0	5.174 ± 0.004		
Best value		5.173	0.2%	

Gamma rays and decay scheme of Th^{228}. -- In order to examine the gamma rays of Th^{228} free of Ra^{224} and its decay products, a quick chemical separation was devised. The mixture in dilute nitric acid solution was placed on a Dowex 50 resin column jacketed to allow operation at an elevated temperature (in this case 87° C) which permits rapid equilibration. The radium, lead and bismuth fractions were eluted with 4 M nitric acid after which the thorium was stripped with a 50 volume percent solution of lactic acid at pH 3. The thorium solution was evaporated to dryness, the alpha activity measured and then the gamma rays were analyzed with a scintillation counter coupled to a 50-channel pulse analyzer. The gamma ray spectrum was measured within one hour of the time that the decay products of Th^{228} were separated.

Fig. 2 shows the gamma ray spectrum of Th^{228} in which is found a prominent peak at 89 kev and lesser peaks at 137, 169 and 212 kev. The assignment of these gamma rays to Th^{228} seems certain since the growth of the 238 kev gamma ray of Pb^{212} and those of Bi^{212} were readily followed and extrapolation of their abundances to time of initial measurement showed that separation of the decay products of Th^{228} was complete. The small peak on the low energy side of the 89 kev peak is probably its escape peak and each of the others will also have an escape peak hidden under the next lowest one. Similarly, K x-rays from the internal conversion of the higher energy gamma rays will be under the 89 kev peak. For calibrating the instrument, the 60 kev gamma ray of Am^{241} and the 184 kev gamma ray of U^{235} were employed. The standards could not be run concurrently with the Th^{228}; as a result there are uncertainties in energy due to drift of photomultiplier voltage which has been found in practice to be less than

one channel corresponding to 8 kev. Probably the gamma energies are good to about 5 kev.

The intensities of the gamma rays were determined by correcting the observed intensities for counter efficiency and for the escape peaks. The corrected values as percentages of the total Th^{228} alpha particles are shown in the decay scheme, Fig. 3. The energy levels shown are those determined from the alpha spectrum while the measured gamma ray energies are indicated along with their abundances by the vertical arrows designating the transitions.

The 89 kev gamma ray is almost surely the same as that assigned to a pair of close-lying gamma rays[12] or more recently to a single gamma ray of 84 kev[13] or 83 kev[14]. The energy level of this state from our alpha decay data is 84 kev, which is closer to the above cited values[13, 14] for the gamma ray energy. Riou[14] measured the intensity of the gamma ray (relative to total alpha particles) as 2.1 percent and relating this to the 28 percent abundance of α_{84} the conversion coefficient was found to be twelve[15]. From our measurements (1.6 percent intensity and 28 percent population by α_{84}) the conversion coefficient is 16. These conversion coefficients as well as the $L_I/L_{II}/L_{III}$ ratios correspond with the assignment to E2 radiation (from calculated conversion coefficients of Gellman, Griffith and Stanley[16]) as already pointed out by Rosenblum, Valadares and Guillot[13]. Attempts have been made[17, 18, 19] to assign the radiation uniquely by alpha-gamma angular correlations but although there is strong evidence[18, 19] for an electric quadrupole component the correlation is not exact. Nevertheless, the preponderance of evidence in this case, as well as for even-even nuclei in general, points to 2+ for the first excited state.

We shall consider next the 169 kev gamma ray. This energy agrees well with the difference in energy between the states populated by α_{253} and α_{84}. From the population of the 253 kev state (0.2%) and the intensity of the 169 kev gamma ray (0.09%) a total conversion coefficient of 1.2 is calculated. The sum of $K^{(20)}$ and $L^{(16)}$ theoretical conversion coefficients for E1, E2 and M1 radiation are estimated respectively to be 0.1, 1.2 and 4.9. The radiation is therefore probably E2 and the spectroscopic state designation is 0, 2+, or 4+. Because of the absence of the crossover transition and the apparent absence of any M1 admixture, the most probable designation is 4+, although 0+ is not ruled out from these considerations alone.

The picture so far discussed for Th^{228} corresponds precisely with the spectrum of another even-even nuclide, Cm^{242}, namely, the appearance of two excited even-spin states (2+ and 4+) whose energies are in the ratio of about three.[4] The generality of this condition for the even-even nuclides of the heavy elements has been discussed[2] and shown to conform with the expectations for rotational levels for the even-spin states.[3] In the case of the excited states of Pu^{238} from alpha decay of Cm^{242} and beta decay of Np^{238}, it was deduced[4] that the low-lying states have even parity and that the first odd parity state lies at about 1 Mev. This takes into account the above mentioned alpha decay picture and, since Np^{238} should have odd parity, the observed division into an allowed low energy beta transition and one or more high energy beta transitions. To explain the other gamma rays found in Th^{228} decay we are assuming that the first odd parity state of Ra^{224} has decreased in energy so that it lies among the even parity states according to the following arguments.

The energies of the 137 kev and 212 kev gamma rays agree within experimental error with transitions from the state populated by α_{217} to the first even-spin state and to the ground state. The sum of the gamma ray intensities is the same as the population of the 217 kev by alpha particles within experimental error (actually larger by measurement) so that the conversion coefficients for both gamma rays must be considerably less than unity. This can only be fulfilled by assigning both to El transitions which makes the 217 kev level a (1-) state. All of these data are consistent with the decay scheme of Fig. 3.

Alpha decay theory.--It has been shown by a number of independent calculations that the half-life-energy relationships of the ground state transitions of both Th^{228} and Ra^{224} conform substantially with simple alpha decay theory. Similarly, the first even spin states (2+) of both conform with the theory for their respective energies or, in our terminology, they are unhindered. However, the transitions of Th^{228} to the 1- and the 4+ states are hindered by about a factor of ten. The reason for this behavior is not yet clear although in another paper[2] a suggestion is advanced.

The reason alpha groups of Ra^{224} other than that to the ground state and to the 2+ state are not seen must be because of the low energy which such groups would have. We can estimate[2] that the energy of the 4+ state would be about 600 kev above the ground state, and even if the alpha transition is unhindered its abundance would be less than 0.05%. A group of this intensity would not have been measurable even if it were not obscured by other alpha groups in the samples. Since we cannot

estimate the position of the first odd parity state, no definite statement can be made regarding the absence of the corresponding alpha transition.

IV. SUMMARY

The present paper aims to present a link in the accumulating evidence for a high degree of regularity in the spacing of energy levels and their quantum states for even-even nuclei in the heavy element region. Starting with the heaviest elements we have as a basis a series of low-lying states 0+, 2+, 4+, 6+(?) with the first odd parity state apparently lying at an elevated level. As one descends to lower elements the same structure (at least for the lowest states) persists, but the energy spacings increase. Furthermore, the ratios of these energy levels for each nucleus follow a $J(J+1)$ dependence up to a certain point. With Th^{228}, and presumably others in this region, an odd parity state appears among the even parity low-lying states. These considerations should be of value in interpreting beta spectra of the odd-odd nuclides in this region.

Alpha decay theory for transitions to the ground state and the 2+ state is obeyed in first approximations by the two alpha emitters considered here. However, for transitions to other states, an extension must be created. The factors to be considered are probably different than those for odd-nucleon nuclides for which alpha decay theory also does not apply.

V. ACKNOWLEDGMENTS

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VI. APPENDIX

Preparation of Samples for Alpha-Particle Spectrograph Exposures

Sample 1.--About 10^8 disintegrations per minute of a dilute acid solution of Th^{228} were added to the top of a Dowex-50 cation-exchange resin column upon which the Th^{228} and most decay products remained. Upon passing 4 M nitric acid through the column, the thorium fraction remained and the radium washed through. The radium fraction was concentrated and then evaporated to dryness on a tungsten filament which could function as a heater.

By heating the filament at relatively low temperature (dull red heat) any polonium present was removed, as was the lead (ThB). The Ra^{224} was then sublimed in vacuum onto a platinum plate by raising the filament temperature. The final sample contained 2×10^5 alpha disintegrations per minute of Ra^{224} with no detectable Th^{228}.

Sample 2.--About 10^{11} disintegrations per minute of Ra^{228} (MsThI) in dilute nitric acid were placed on a Dowex-50 resin column and the radium fraction stripped as above with 4 M nitric acid. The thorium was then removed with 16 M nitric acid. A portion of this eluate was concentrated and evaporated to dryness on a tungsten filament. The thorium and decay products which had grown in were vacuum-sublimed onto a platinum plate. The sample so prepared contained 1.4×10^8 disintegrations per minute of total alpha particles, about half of which belonged to Th^{228} and the rest to its decay products.

Sample 3.--The Th^{228} residue on the filament from which Sample 2 was prepared was allowed to remain for 13 days to equilibrate with its

decay products. A relatively low temperature vacuum sublimation was carried out to concentrate the Ra^{224} on the collector plate relative to the Th^{228}. About one-sixth of the 5×10^7 alpha disintegrations caught belonged to Th^{228} and the remainder to Ra^{224} and its decay products.

Sample 4. --The objective in preparing this sample was to observe groups of Th^{228} in the absence of the equilibrium amount of Ra^{224}. Thorium 228 was separated from its decay products by the rapid method outlined in the text of this paper for preparing Th^{228} for gamma ray analysis. In this case the separation of Th^{228} from Ra^{224} was not quantitative because macroscopic impurities interfered with the column operation. After column separation the Th^{228} solution was concentrated and evaporated to dryness on a tungsten filament. The Th^{228} was then vacuum sublimed onto a platinum plate after a low temperature sublimation to remove decay products. About 4×10^8 alpha disintegrations per minute were collected, about 75 percent of which were from Th^{228}.

Table 4

Table showing characteristics of the various spectrograph exposures

Sample number	Source slit* width - inches	Exposure number	Length of exposure	Time between preparation of sample and start of exposure
1	0.13	174	94 hours	3 days
2	.018	225	30.0 min.	1 hour
	.018	226	19 hours	2 hours
	.018	230	41 hours	3 days
	.13	248	44 hours	64 days
3	.13	238	10 hours	1 day
	.018	241	63 hours	3 days
4	.018	258	3 1/4 hours	1 hour
	.018	259	10.0 min.	4 hours

*This defines the width of the masking slit which is placed immediately in front of the sample to simulate a line source. The length of the slit is about one inch.

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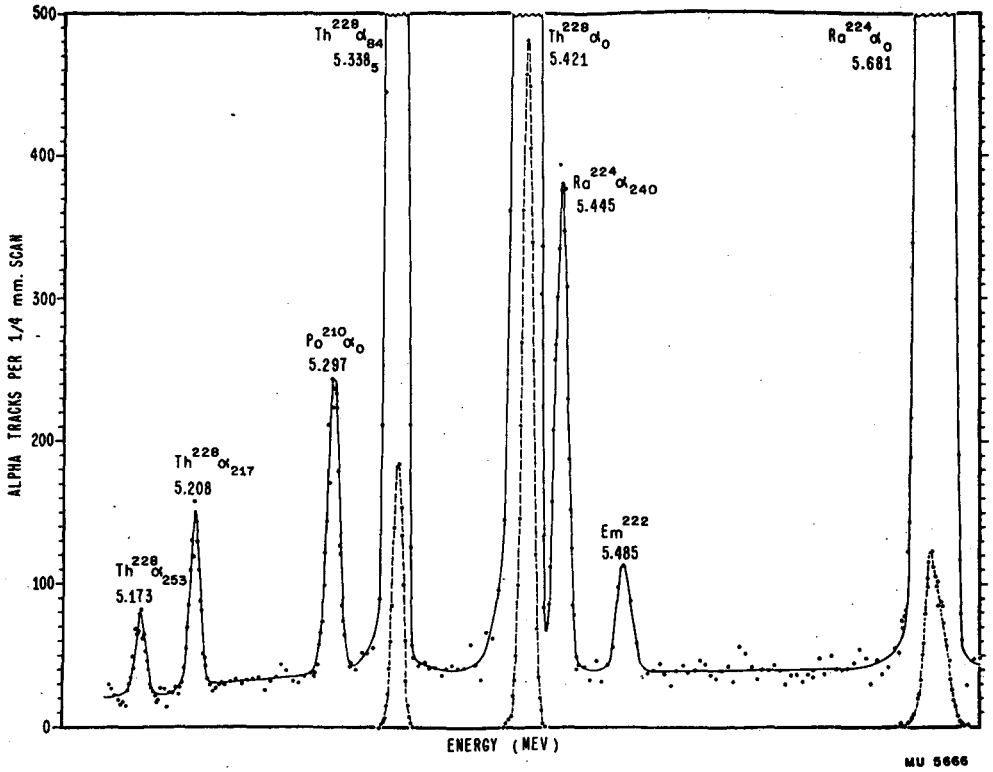

Fig. 1

Alpha spectra of Th^{228} and Ra^{224}

————Full scan across width of photographic plate

- - - - - - -About 2.4% of full scan

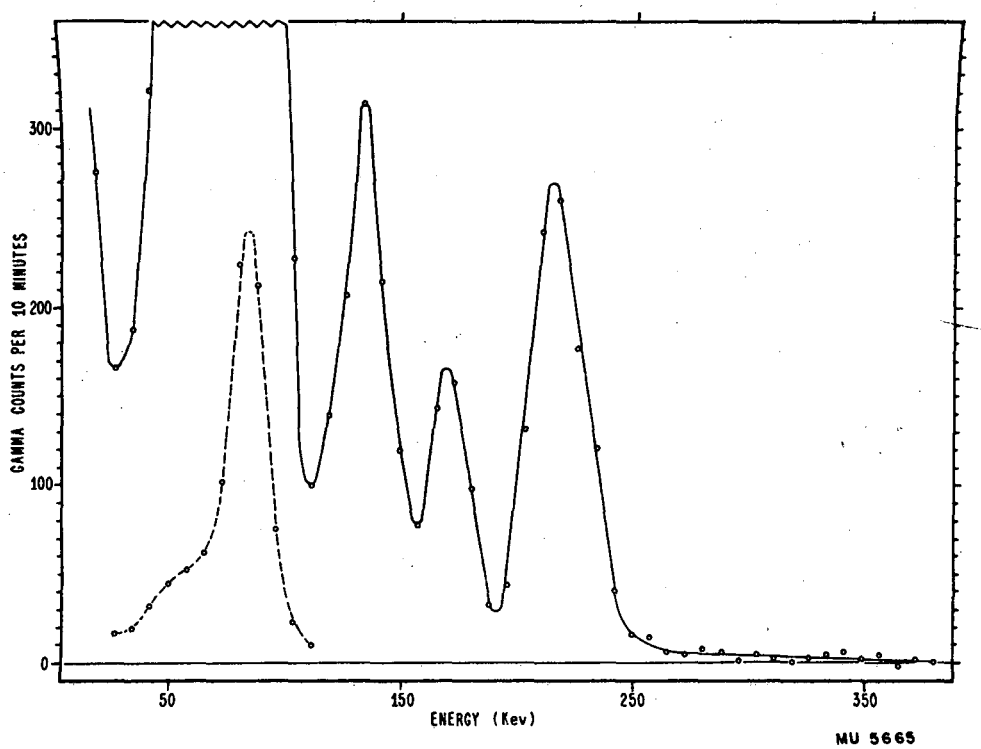

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Fig. 2

Gamma ray spectrum of Th^{228} -

———Gamma ray counting rate indicated by ordinate scale

- - - - - - -Gamma ray counting rate equals 10 times the value indicated by ordinate scale

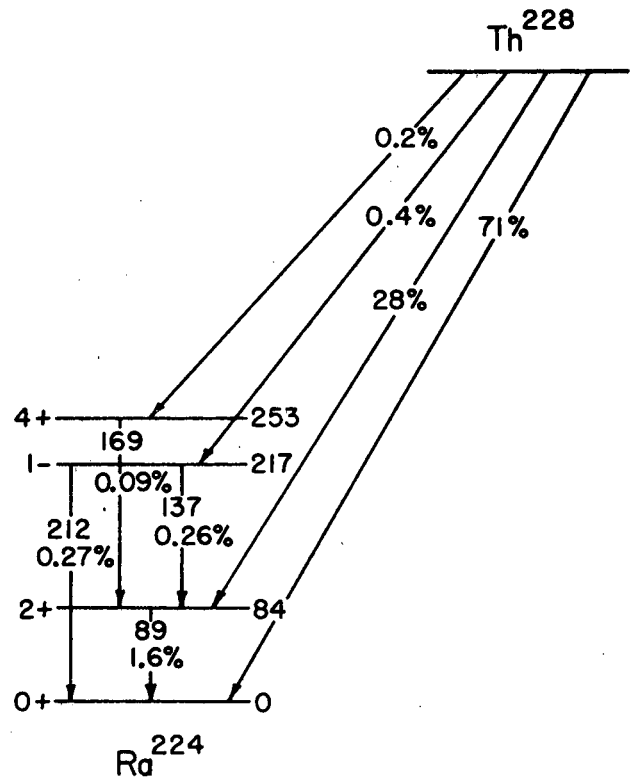

Fig. 3

Th^{228} decay scheme
(Energies given in kev)