RADIOACTIVE DECAY OF Ac^{227} *AND EXCITED LEVELS OF* Fr^{223} *AND* Th^{227}

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The radioactive decay of Ac^{227} was investigated. Alpha decay was studied by means of an alpha-particle spectrometer. A beta-ray spectrometer in conjunction with an alpha-beta coincidence circuit was employed to study beta decay and the spectra of conversion electrons accompanying the alpha and beta decays of Ac^{227}. Fine structure was detected in the Ac^{227} alpha-particle spectrum. The energies of seven new lines and the intensities of the corresponding transitions were determined. A number of gamma transitions between levels of the Fr^{223} daughter nucleus were detected. A level scheme for Fr^{223} is proposed. Three levels were detected in the spectrum of Th^{227} that results from the beta decay of Ac^{227}. The energies of the levels and the intensities of beta decay to them were measured.

1. INTRODUCTION

ESSENTIAL differences are known to exist between nuclear level schemes depending upon the relative completion of nucleon shells in different nuclei. Nuclei possessing a large number of nucleons not included in closed shells are considerably deformed in their equilibrium states. Low-lying levels of such nuclei are, as a rule, associated with collective rotational motion. Because of the simple laws that govern the rotational motion of deformed nuclei it is relatively easy to detect rotational levels and to determine their quantum characteristics (spin, parity etc.). A large number of such nuclei have already been studied. In the case of nuclei with closed shells or with a small number of additional nucleons the stable equilibrium shape is a sphere, and rotational levels cannot appear. The single-particle and vibrational levels are relatively high-lying. As the number of nucleons in unfilled shells increases the energies of these levels are lowered.

Nuclei in the vicinity of closed shells with Z = 82, N = 126 and nuclei with a large number of nucleons in addition to these shells have been studied relatively thoroughly, but the intermediate region is still relatively unexplored. It is usually assumed that the boundary between spherical and deformed heavy nuclei is near A = 225. Even-even nuclei in this region reveal levels that are characterized by the vibrational properties of the nuclear surface. However, the level schemes of odd nuclei near this value of A (in the few instances when they have been studied) are much more complicated and unclear than those of the adjacent even-even nuclei. For example, the alpha-particle spectrum of Th^{227} contains 15 groups of alpha particles,[1] but quantum numbers have not been determined for even a single excited level of the daughter nucleus.

We have studied the radioactive decay of Ac^{227}, which is also in the intermediate region and contains an odd number (7) of protons outside of the shell Z = 82. Ac^{227} decays with 1.2% emission of alpha particles and ~ 99% emission of beta rays, with a half-life of 21.6 years. Although this isotope has been known for a long time its radioactive decay has remained practically uninvestigated. Concerning the alpha decay of Ac^{227} there are only unpublished studies (mentioned in reference 1) which indicate that in its alpha-particle emission Ac^{227} decays only to the ground level of Fr^{223}. Papers concerning the beta decay of Ac^{227} contain conflicting information about the levels of Th^{227}. Reference 2 states that 85% of the decays go to the ground level of Th^{227} and 15% to a 37-kev excited level. More recently[3] the measured end-point energy of the Ac^{227} beta-ray spectrum was found to be 45.5 ± 1.0 kev, but the existence of excited Th^{227} levels was not confirmed. Reference 4 contains indications that the beta decay of Ac^{227} is accompanied by gamma-ray emission with energy > 16 kev.

The absence of reliable experimental data on the decay of Ac^{227} results from the great difficulties encountered in the study of this nucleus. The beta decay of Ac^{227} results in the rapid accumulation of a number of short-lived isotopes: Th^{227} ($T_{1/2}$ = 18.6 days), Ra^{223} ($T_{1/2}$ = 11.2 days), Em^{219} ($T_{1/2}$ = 3.92 sec), Po^{215} ($T_{1/2} = 1.8 \times 10^{-3}$ sec), Bi^{211} ($T_{1/2}$ = 2.6 min), etc. The existence of short-lived emanation and its daughters among the products of Ac^{227} beta decay results in a strong background of random alpha particles, which greatly hinders the

study of the relatively weak Ac^{227} alpha activity. The study of Ac^{227} beta decay is hampered by the large number of conversion electrons emitted by the daughter nuclei; it is quite impossible to distinguish conversion lines associated with Ac^{227} alpha decay against this background. The first problem in the investigation of Ac^{227} decay was therefore the thorough chemical removal of Th^{227} and Ra^{223}. The other products of Ac^{227} decay possess very short half-lives and accumulate only in proportion to the accumulation of Th^{227} and Ra^{223}.

2. CHEMICAL PURIFICATION OF ACTINIUM

Th^{227} and Ra^{223} were separated chromatographically from Ac^{227} by means of a procedure which Peterson[5] and Hagemann[6] had used to separate actinium from irradiated Ra^{226}. In reference 5 Ac was separated from Ra using the Amberlite IR-1 ion-exchange resin. The eluant for Ac was 0.25 M ammonium citrate, while the eluant for Ra was 3N HCl. In reference 6 Ra was eluted from the Dowex-50 cation-exchange resin by means of 2N nitric acid, while 4N nitric acid was used for the Ac. In the latter reference the procedure was indicated very briefly without a description of the conditions for separating these elements.

We separated Ac^{227} from Th^{227} and Ra^{223} using ion-exchange resin KU-2 in hydrogenous form. Nitric acid in different concentrations was the eluant. The resin was first finely divided and shaken together with water. For column filling we required a fraction that would settle at the rate of 0.2 — 0.5 cm/min. 5N HNO_3 and 5% NH_4OH were passed through the columns successively in order to remove the impurities in the resin.

The actinium used as the starting material contained no appreciable amount of radioactive impurities other than its own decay products. The weight of inactive impurities in the actinium sample did not exceed the weight of actinium oxide. Doubly-distilled HNO_3 with a specific gravity of 1.38 was used to prepare the nitric acid eluant. All solutions were kept in quartz flasks.

In developing our technique we had two purposes: 1) to determine HNO_3 concentrations for eluting Ra^{223}, Ac^{227}, and Th^{227}; 2) to determine the conditions for separating these elements which would enable us to reduce the volume of eluant to a minimum.

The first trials showed that the bulk of the Ra^{223} is eluted with 2N HNO_3, Ac^{227} with 4N HNO_3, and Th^{227} with 5N HNO_3. In these experiments an actinium fraction amounting to 50 cm^3 contained ~ 80% of the original amount of Ac together with 3% of the equilibrium amount of Ra^{223} and 1 to 2% of the equilibrium amount of Th^{227}.

The following optimum experimental conditions were determined: column length $l = 8 - 10$ cm, diameter d = 0.2 cm and elution rate v = 0.15 cm^3/min. These conditions enabled us to reduce to 15 or 20 cm^3 the volume of the actinium fraction containing the bulk of the Ac, together with 2 or 3% of the equilibrium amounts of Ra^{223} and Th^{227}. The use of thoroughly purified ion-exchange resin and solutions insured the required purity of the final solution; evaporation and further heating of a 25 cm^3 actinium fraction left a residue weighing no more than 0.01 mg.

The final trials to obtain an actinium solution free of Ra^{223} and Th^{227}, which would be used in preparing sources, were conducted under these same conditions. In trials 1, 2, and 3 the original solution contained 31, 19, and 570 microcuries of Ac^{227} respectively; the results are shown in Table I.

TABLE I

Number of sample	HNO_3 concentration	volume of eluate, cm^3	Ac^{227} content in sample, microcuries		
			Trial 1	Trial 2	Trial 3
1	2 N	50	—	—	—
2	4 N	1	0.7	0.7	8.0
3	4 N	20	20.0	14.3	430
4	4 N	15	7.5	1.4	35
5	4 N	15	0.1	0.1	7.5
6	5 N	50	2.5	1.7	74

For our subsequent work we used samples 3 and 4, which in all three trials contained the bulk of Ac^{227}. The solutions were boiled down to a small volume in quartz dishes and were then transferred to conical quartz test tubes in which they were evaporated almost to dryness before being diluted with 1N HNO_3 to a volume of 0.05 cm^3.

3. EXPERIMENTAL PROCEDURE

The alpha-particle spectrum of Ac^{227} was investigated by means of an alpha-particle spectrometer.[7] The sources were prepared by vacuum evaporation of actinium (that had been purified of decay products) from a tantalum filament. Additional separation of Ac and its daughter isotopes during evaporation was achieved through the proper choice of filament temperature. The chemical purification combined with purification during sublimation yielded sources that were practically free of Ra^{223}, Em^{219}, and all decay products of Em^{219}. At the beginning of the readings the Th^{227} content of the sources amounted to 3 — 6% of the Ac^{227}, as determined from the total activity. With such small

Th^{227} content the ratio of the number of all alpha particles to the number of Ac^{227} alpha particles per unit time varied from 3:1 to 6:1 and became larger in the course of time. Therefore no source could be used for longer than 100 hours, after which period the background of random alpha particles became too large. The actinium activity of the sources amounted to 1 — 3 microcuries.

The spectrum of conversion electrons accompanying the alpha and beta decays of Ac^{227} was investigated by means of an alpha-beta coincidence spectrometer.[8] Even though the sources were carefully purified of Ac^{227} decay products the background of Th^{227} and Ra^{223} conversion electrons accumulating during readings was still too large. Therefore in order to distinguish the conversion electrons accompanying Ac alpha decay we introduced an additional channel for coincidences between conversion electrons and alpha particles discriminated according to their energies. We thus simultaneously registered three curves: 1) the total beta-ray count, 2) the count of alpha-beta coincidences and 3) the count of β-α_{discr} coincidences.

In the β-α_{discr} channel alpha particles were detected by a scintillation counter with a thin CsI(Tl) crystal. The counter resolution was 8%. Alpha particles were discriminated by means of a single-channel pulse-height analyzer with a window of variable width. The transmission of the apparatus was ~ 2% for the recording of β-α coincidences and 0.5% in the β-α_{discr} channel.

A shortcoming of the technique used to study beta-ray and coincidence spectra was the use of a scintillation counter with a stilbene crystal to detect electrons. The sensitivity of such counters declines rapidly as the energy rises. To determine the sensitivity the Ac^{227} spectrum was recorded with accelerating, zero and retarding potentials applied to the source. The sensitivity curve was established according to the variation of relative peak intensities. With calibration performed in this manner the errors in determining conversion line intensities in the soft part of the spectrum may amount to 30%.

4. MEASUREMENT RESULTS

Figures 1 and 2 show the alpha-particle spectrum of Ac^{227} with the ordinate axis representing n_{av}, the average number of alpha-particle tracks in a strip of photographic emulsion. Figure 1 shows alpha lines representing transitions to the ground state and to three low-lying excited levels of Fr^{223}, which were registered during a period of 11 hours and 45 minutes. Figure 2 shows alpha lines corresponding to higher excited levels of Fr^{223}, which were registered in 45 hours and 20 minutes. A

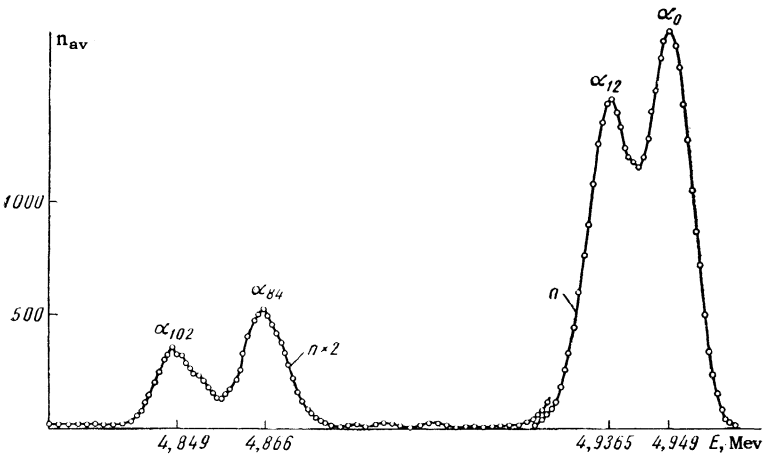

FIG. 1. Alpha-ray spectrum of Ac^{227} in the 4825–4975 kev region.

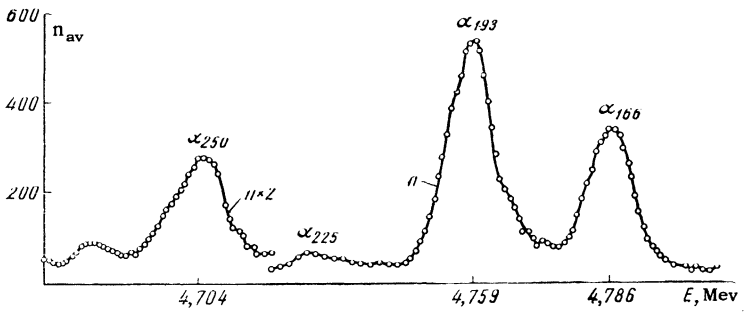

FIG. 2. Alpha-ray spectrum of Ac^{227} in the 4650–4800 kev region.

weak 225-kev line is observed, but the intensity of the corresponding transition is only 0.1% of the alpha activity and the existence of a 225-kev level cannot be regarded as proven. The energy intervals from 4950 to 5050 kev and from 4450 to 4650 kev were also investigated. The 4450-4650 kev region revealed a line with ~ 0.2% intensity corresponding to a 440-kev level, the existence of which also requires confirmation. No alpha line with intensity greater than 0.1% was detected in the 4950 — 5050 kev region.

The results of our investigation of the Ac^{227} alpha-particle spectrum are summarized in Table II.

TABLE II

Line	Alpha-particle energy, kev	Energy level, kev	Alpha-transition intensity, %	Hindrance factor η
α_0	4949±2	0	48.7±3	6.7
α_{12}	4936.5±3	12.7	36.1±3	8
α_{84}	4866±3	84.5	6.9±1	14
α_{102}	4849±3	102	5.5±1	14
α_{166}	4786±5	166	1.0±0.5	40
α_{193}	4759±5	193	1.8±0.5	17
α_{225}?	4728±8	225	~0.1	—
α_{250}	4704±8	250	0.4±0.2	38
α_{440}	4517±10	440	~0.2	—

The Ac^{227} spectra of beta rays and coincidences of conversion electrons with alpha particles are shown in Fig. 3. Curve 1 in the upper part represents the total beta-ray count, while curve 2 represents the β-α coincidence count. Curve 1 clearly shows 9.3-, 15.2-, and 24.5-kev conversion

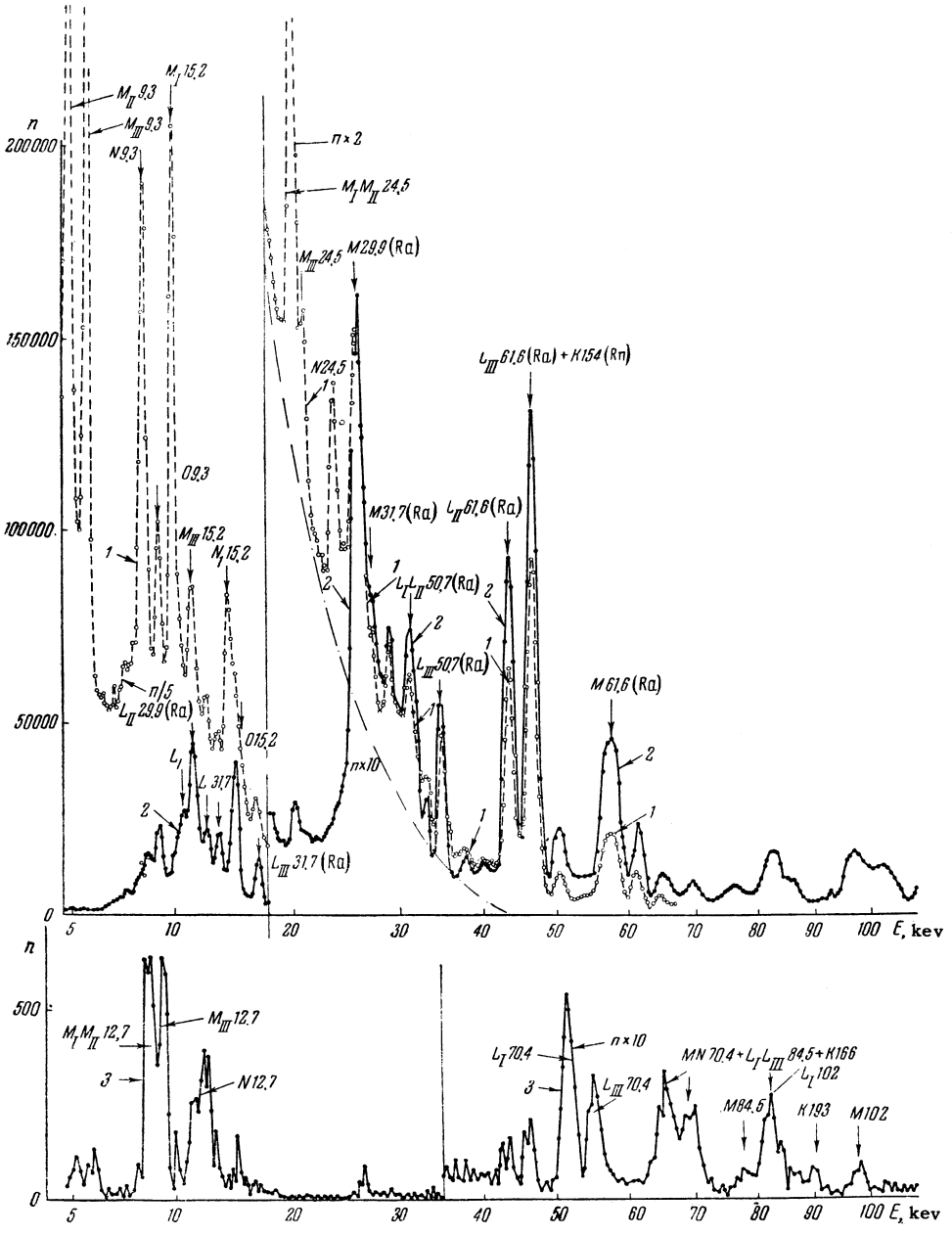

FIG. 3. Beta-ray spectrum of Ac^{227} and spectrum of conversion electrons accompanying the alpha and beta decays of Ac^{227}. 1) total count of electron detector; 2) e—α coincidence count; 3) e—α coincidence count with discrimination in the α-particle channel. The ordinate axis represents the number of beta-counter pulses. Each curve is plotted on two scales as indicated in the figure. The beta line near the end-point is plotted with a dot-dashed line.

transitions, which are not present on the coincidence curve (curve 2). The intensities of these lines do not increase as Ac^{227} decay products accumulate in the source. These energies are therefore associated with the beta decay of Ac and represent transitions between levels of the Th^{227} daughter nucleus. The dashed line below the total-count curve represents the part of the continuous beta-ray spectrum near the end-point.

The β-α_{discr} coincidence curve is shown in the lower part of the figure. A comparison with curve 2 shows that the background of accidental coincidences is small and that all conversion lines can be assumed to represent transitions accompanying Ac^{227} alpha decay. An analysis of results obtained with the same source during different time intervals shows that the intensities of these lines do not increase. This provides additional proof that the corresponding transitions take place in the Fr^{223} nucleus which results from Ac^{227} alpha decay.

Table III summarizes the measurements of conversion electrons accompanying the alpha and beta decays of Ac^{227}.

5. LEVEL SCHEME OF Fr^{223}

The scheme of Ac^{227} alpha decay and Fr^{223} levels is shown in Fig. 4 together with the observed gamma transitions. The 12.7-kev transition (Table III) goes from the first excited level to the ground level; this is an E2 transition [as determined from the $(M_I + M_{II})/M_{III}$ ratio]. 70.4-kev gamma radiation results from a transition between the 84.5-

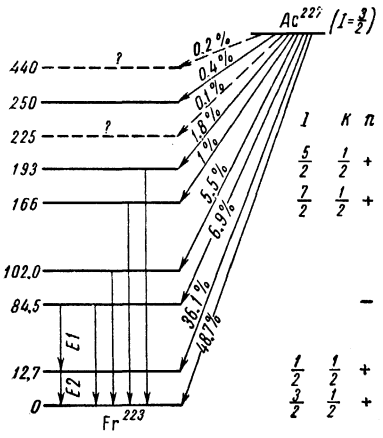

FIG. 4. Scheme of Ac^{227} alpha decay and Fr^{223} levels.

TABLE III

	Conversion-electron energy, kev	Relative line intensity, arb. units	Line origin	Gamma-ray energy, kev	Transition energy, kev
			Conversion electrons of Th^{227} (curve 1)		
1	4.46	1425	M_{II}	9.27	
2	5.24	1545	M_{III}	9.27	
3	8.28	720	N	~9.33	9.3±0.1
4	9.23	200	O	9.50	
5	10.04	400	M_I	15.20	
6	11.50	40	M_{III}	15.16	15.2±0.1
7	13.98	130	N_I	15.29	
8	15.02		O	15.29	
9	19.5	50	$M_I + M_{II}$	~24.5	
10	20.6	8	M_{III}	24.63	24.5±0.2
11	23.4	15	N	~24.45	
12	24.3		O	24.6	
			Conversion electrons of Fr^{223} (curve 3)		
1	8.47	130	$M_I + M_{II}$	~12.8	
2	9.04	130	M_{III}	12.70	12.7
3	11.90	80	N	~12.7	
4	51.76	11	L_I	70.39	70.4
5	55.38	6	L_{III}	70.40	
6	65.69		L_I	84.32	84.5
7	65.69	7	M	70.0	70.4
8	65.69		K	166.7	166
9	69.6	4	L_{III}	84.6	84.5
10	78.6	1	M	~83	
11	82.4	5	L_I	101.0	101
12	89	1	K	190	190
13	96.6	1.5	M	101	101

and 12.7-kev levels and may be of multipolarity E1 or M2, as shown by an analysis of L-conversion line intensities. In addition, the β-α_{discr} coincidence spectrum includes lines which may be attributed to 84.5-, 101-, 166-, and 190-kev gamma transitions, whose multipolarities could not be determined.

The Ac^{227} ground state has been shown experimentally to have spin $3/2$.[9] According to Nilsson's scheme[10] a nucleus with Z = 89 will have $K = 3/2$ and even parity (level [651]). It can therefore be assumed that the ground state of Ac^{227} has $K = 3/2$, $I = 3/2$ and even parity. The spin of Fr^{223} was not measured; if this is a spheroidal nucleus, according to Nilsson's scheme the ground state most probably has $K = 1/2$ and even parity (level [660]). The hindrance factor for decay to the ground state of Fr^{223} is small ($\eta = 6.7$), and it is reasonable to assume that the level scheme should contain rotational levels belonging to the ground-state band. An E2 ground-state transition takes place from the 12.7-kev level. If it is assumed that this level is the first rotational satellite of the ground level, it follows from their small separation that for these two levels $K = 1/2$, which agrees with the value of K for the Fr^{223} ground state in Nilsson's scheme. Higher levels of this band should, like the lower levels, form doublets. The Fr^{223} level scheme clearly shows two doublets in addition to the lower one. However the 84.5-kev level in the lower one of these additional doublets has parity opposite to that of the still lower levels. Moreover, the energies of the four lower levels are not given by the familiar formula[11] for rotational levels with $K = 1/2$:

$$E_{rot} = (\hbar^2/2J)\{I(I+1) - 3/4 - a + a(-1)^{I+1/2}(I+1/2)\}. \quad (1)$$

However this formula accurately represents the energies of the two lower levels and of the 166- and 193-kev levels (within experimental error). We here have $\hbar^2/2J = 17.4$, which agrees with the values that can be expected in this region. We therefore conclude that the 0-, 12.7-, 166-, and 193-kev levels are very probably rotational levels belonging to a single band with the spins given in Fig. 4. We have assigned spin $I = 3/2$ to the ground state and $I = 1/2$ to the first excited level. Inverted spin values have also been assigned to the two upper levels, because of the existing ground-state transition from the 193-kev level. This transition should not be observed if the 193-kev and ground levels have spins $7/2$ and $1/2$ respectively. For the given spin sequence the splitting factor in (1) is $a = -1.25$.

The existence of rotational levels in a nucleus belonging to the intermediate region between deformed and spherical nuclei is significant and merits more thorough discussion. It may be assumed that the nuclear shape and the positions and characteristics of the levels depend essentially on an odd nucleon. It is therefore interesting to compare the level schemes of Fr^{223} and Fr^{221}, which are in the same region of A and have the same number of odd nucleons outside of closed shells. The Fr^{221} levels are known from the alpha-decay spectrum of Ac^{225}, which contains nine alpha lines, while seven gamma transitions have been observed between levels of the Fr^{221} daughter nucleus.[1] As in the case of other investigations of nuclei in this region the literature contains no attempts to identify the observed levels. The lower levels of Fr^{221} are shown in Fig. 5 together with the known gamma transitions.

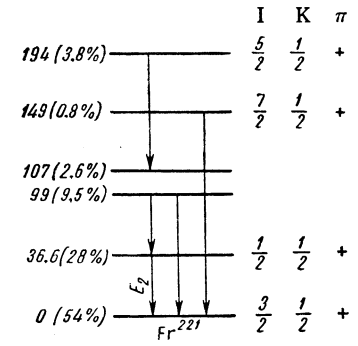

FIG. 5. Lower-lying level scheme of Fr^{221}

The conversion-line intensities associated with the ground-state transition from the first excited level indicate clearly that this is an E2 transition. This multipolarity as well as the relatively small level separation may, just as in the case of Fr^{223}, indicate that these two levels belong to a single band with $K = 1/2$. The next excited levels of the ground-state band may be the 107- and 194-kev levels, which together with the two lower levels are accurately represented by (1). We now have $\hbar^2/2J = 13.3$, which does not differ too much from the corresponding value for Fr^{223}.

The question as to whether the band formed by these four levels has a normal or inverted spin sequence cannot be answered unambiguously without additional investigation of the conversion electron spectrum. We select an inverted sequence by analogy with Fr^{223}. In this case $a = -1.93$, in satisfactory agreement with the value of a for Fr^{223}. Analysis thus shows that the structures of the Fr^{223} and Fr^{221} levels are similar, thus providing additional confirmation of the aforementioned view that the Fr^{223} and Fr^{221} nuclei are deformed. The (nearly) resolved character of the Ac^{227} ($K = 3/2$) alpha decay to the ground state of Fr^{223}

($K = 1/2$), in which K undergoes change, seems at first glance to be a contradiction. But it must be remembered that with diminished deformation we may expect a weakening of K-forbiddenness, and that we at present have no information concerning the magnitude of K-forbiddenness in the given region. It may be small, being represented by $\eta = 6.7$.

Fr^{223} and Fr^{221} may be regarded as deformed nuclei, if the foregoing reasoning is correct. The boundary between deformed and spherical nuclei is then somewhat lower than was previously supposed, at least for odd nuclei.

6. LEVEL SCHEME OF Th^{227}

Table III shows that Ac^{227} beta decay is accompanied by the emission of gamma rays with the energies 9.3 ± 0.1, 15.2 ± 0.1, and 24.5 ± 0.2 kev. The 9.3-kev transition is most likely of the electric quadrupole type, possibly mixed with a considerable amount of magnetic dipole radiation. The multipolarities of the 15.2- and 24.5-kev transitions were determined more accurately: 99.8% M1 + 0.2% E2 for 15.2 kev and 99% M1 + 1% E2 for 24.5 kev. The multipolarities of the transitions were determined from the ratio $(M_I + M_{II})/M_{III}$.

The fact that the sum of two transition energies practically equals the third transition energy indicates that the observed transitions take place between two excited levels at 9.3 and 24.5 kev respectively and the ground state of Th^{227}. Figure 6

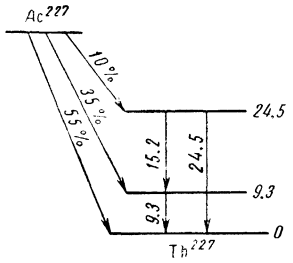

FIG. 6. Scheme of Ac^{227} beta decay and Th^{227} levels.

shows the scheme of Ac^{227} beta decay and Th^{227} levels. For the reasons mentioned above we were unable to determine the intensities of beta decays to Th^{227} levels; approximate intensities are indicated in Fig. 6. The beta-transition intensities were used in calculating log ft. For the ground-state transition log ft = 7.0; for the transition to the 9.3-kev level log ft = 6.9; for the transition to the 24.5-kev level log ft = 6.8. The striking similarity of these values results naturally from the identical parity of the levels. Gamma transitions between these levels are of types M1 and E2. We obtained 42 — 45 kev as the end-point energy of the beta spectrum (Fig. 3), which does not disagree with the result given in reference 3.

Our results are quite insufficient for determining the quantum characteristics of Th^{227} levels. For this purpose we would require information concerning higher levels, which may, of course, not be occupied through Ac^{227} beta decay because of the low end-point energy.

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[1] Strominger, Hollander, and Seaborg, Table of Isotopes, Revs. Modern Phys. **30**, 585 (1958).

[2] Lecoin, Perey, Riou, and Teillac, J. phys. radium **11**, 227 (1950).

[3] W. Beekmann, Z. Physik **142**, 585 (1955).

[4] Bouchez, Michalowicz, Riou, and Teillac, J. phys. radium **16**, 344 (1955).

[5] S. Peterson in The Transuranium Elements (Seaborg, Katz, and Manning, editors), McGraw-Hill Book Co., New York, 1949, Vol. 2, p. 1393.

[6] F. Hagemann in The Actinide Elements (G. T. Seaborg and J. J. Katz, editors), McGraw-Hill Book Co., New York, 1953; Russ Transl. IIL, 1955, p. 15.

[7] L. L. Gol'din and E. F. Tret'yakov, Izv. Akad. Nauk SSSR, Ser. Fiz. **20**, 859 (1956); Columbia Tech. Transl. p. 781.

[8] Tret'yakov, Gol'din and Grishuk, Приборы и техника эксперимента (Instruments and Measurement Engineering) **6**, 22 (1957).

[9] Tomkins, Fred, and Meggers, Phys. Rev. **84**, 168 (1951).

[10] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **29**, No. 16 (1955).

[11] Bohr, Froman, and Mottelson, Kgl. Danske Videnskab.Selskab, Mat.-fys. Medd. **29**, No. 10 (1955).

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