

Nuclear Spectroscopic Studies of $\text{Fm}^{257}\dagger$

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The nuclear radiations associated with the α decay of Fm^{257} have been investigated by a variety of spectroscopic techniques. Alpha groups were observed with energies and intensities of ~ 6.75 , $(0.4 \pm 0.2)\%$; 6.696 ± 0.003 , $(3.2 \pm 0.3)\%$; 6.519 ± 0.002 , $(94 \pm 1)\%$; 6.441 ± 0.003 , $(2.0 \pm 0.4)\%$; and ~ 6.35 MeV, $(\sim 0.5)\%$. Gamma-ray transitions were seen in coincidence with α particles with energies of 242, 180, and 62 keV. The ground state of Fm^{257} is given the Nilsson assignment $\frac{5}{2} + \frac{3}{2}[615\downarrow]$ as is the 242-keV state of Cf^{253} . The ground state of Cf^{253} is given the assignment $\frac{3}{2} + \frac{3}{2}[613\uparrow]$. For these bands in the daughter nucleus the rotational constants $\hbar^2/2\mathcal{I}$ have the respective values of 7.1 ± 0.4 and 7.0 ± 0.1 keV, the largest values found yet in the very heavy elements. Some of the α - and γ -ray transitions are interpreted in terms of a Coriolis interaction between the two assigned Nilsson levels in the daughter nucleus.

I. INTRODUCTION

THE α emitter Fm^{257} is of some interest because the study of its α decay could reveal the spectroscopic assignment for neutron number 157 and the low-lying states associated with neutron number 155 in the Cf^{253} daughter. The ground state for neutron number 155 is known from Fm^{255} but nothing is known about neutron number 157. Fm^{257} has a half-life of something under 100 days^{1,2} and grows successively the β^- emitter Cf^{253} and the α emitter Es^{253} , each with a half-life of approximately 20 days. The difficulty of extracting information from Fm^{257} decay lies in the fact that it has only been made in very minute amounts. The earlier studies¹⁻³ employed sources of only a fraction of a disintegration per minute up to about one disintegration per minute. In the present study, sources several times more intense were available as a result of the irradiation of curium isotopes in a high-flux reactor at the Savannah River Plant. The amounts prepared were still not enough for a detailed study of the spectrum but the favored α transition was clearly delineated as leading to a state at 242 keV, and much was learned about the ground-state band by measuring γ transitions from the favored state.

II. SOURCE PREPARATIONS

Mixtures of curium isotopes were irradiated in the Materials Testing Reactor (MTR) and the Savannah River Reactor (SRR). The irradiated material was put through the usual chemistry procedure⁴ by the Berkeley or Livermore heavy-element production groups and a fermium fraction was extracted. These operations were

followed by many separations in ion-exchange columns to remove plutonium, separate the fermium from other actinides and rare earths, and make the fermium nearly mass free. After purification the activities were dissolved in 0.004 *M* HNO_3 and electroplated onto a 0.0001-in. nickel plate covering an area about $\frac{1}{8}$ in. diam.⁵ Four fermium sources were prepared in this fashion. Source I contained, after chemistry, considerable 20-h Fm^{255} , ~ 2.3 disintegrations/min of Fm^{257} , ~ 1.2 disintegrations/min of Es^{253} (impurity), 0.35 disintegrations/min of Cf^{252} (impurity), and ~ 0.25 disintegrations/min of Cf^{251} from the decay of Fm^{255} . The other sources were not processed as quickly and did not contain as many impurities. Source II contained 1.9 α disintegrations/min of Fm^{257} , source III contained 28 disintegrations/min of Fm^{257} , and source IV contained 8.4 disintegrations/min of Fm^{257} after chemistry.

III. EXPERIMENTAL

A. α Spectra

The earlier measurements of the α particles of Fm^{257} using ionization chambers showed a group at 6.525 ± 0.005^3 or 6.56 ± 0.04 MeV.¹ Other groups of somewhat higher energy were unresolved and could have been fictitious because conversion electrons in coincidence with α groups are partially registered in this type of measurement.³

The α spectrum of source I was measured with a Frisch grid chamber at a geometry of about 40%. A spectrum taken about two weeks after the initial Es-Fm separation and about 9 days after the last separation is shown in Fig. 1. The 20-h Fm^{255} originating from Es^{255} is still prominent. This peak is quite broad because of copious coincidences with conversion electrons. Also prominent is a smear of α -particle energies from 6.53–6.76 MeV which has been previously identified^{1,3} as Fm^{257} . A portion of the smear in our spectrum is due to Es^{253} which was present as an impurity. The peak

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¹ E. K. Hulet, R. W. Hoff, J. E. Evans, and R. W. Loughheed, *Phys. Rev. Letters* **13**, 343 (1964).

² Combined Radiochemistry Group, *Phys. Rev.* **148**, 1192 (1966).

³ T. Sikkeland, A. Ghiroso, R. Latimer, and A. E. Larsh, *Phys. Rev.* **140**, B277 (1965).

⁴ R. M. Latimer and J. T. Haley, University of California Lawrence Radiation Laboratory Report No. UCRL-16191, 1965 (unpublished).

⁵ This electroplating technique was devised by Dr. R. M. Latimer of the Lawrence Radiation Laboratory in Berkeley.

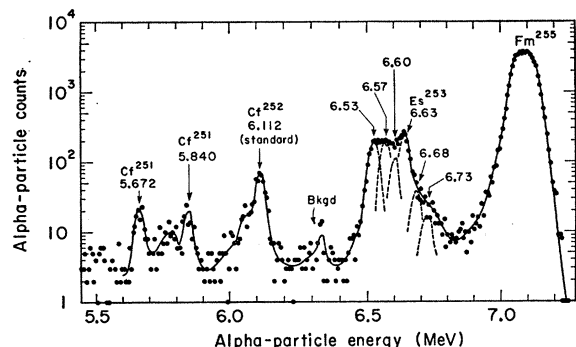


FIG. 1. Fm^{257} α spectrum taken with a Frisch grid chamber ($\Omega=40\%$).

at 6.11 MeV is due to Cf^{252} which was incompletely removed in the chemistry, and the peaks at 5.84 and 5.67 MeV are due to Cf^{251} which grows into Fm^{255} . The smearing of the Fm^{257} α peak is also very likely caused by intense conversion electron coincidences with the main α group. In order to check this possibility, the α spectrum was measured at solid angles of 7.5, 4, 3, and 1.75% with surface-barrier Au-Si detectors coupled to appropriate electronic circuitry. Figure 2 shows the α spectra of Fm^{257} taken at a solid angle of 3%. The various α -particle energies and intensities are summarized in Table I. At the lowest solid angle most of the Fm^{257} alpha radiation is in a single peak at 6.519 MeV. A high-energy shoulder on this peak becomes relatively more intense as the solid angle becomes larger. This type of behavior would be caused by coincidences between the main α group and conversion electrons, Auger electrons, and L x rays. A peak at 6.696 MeV represents a true alpha group of Fm^{257} as its intensity, $(3.2 \pm 0.3)\%$, did not change drastically in the various runs. A peak at 6.75 MeV seems to be due to Fm^{257} and has an intensity of $(0.4 \pm 0.2)\%$ after the coincidence effects due to electrons are subtracted. There is a group of 6.441 MeV with an intensity of 2.0% and a rather dubious group at ~ 6.35 MeV with an intensity of $\sim 0.5\%$. Neither of these last two groups could be caused by α - e^- coincidences because their energies are smaller than the main α peak.

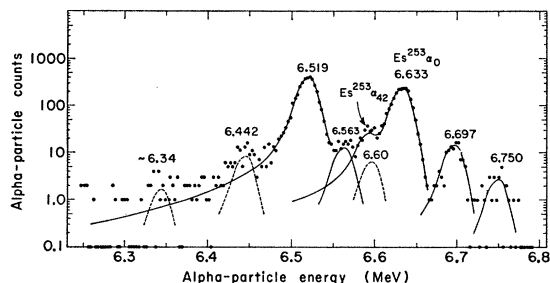


FIG. 2. Fm^{257} α spectrum taken with a Au-Si surface-barrier detector ($\Omega=3\%$).

B. α -Particle- L X-Ray Coincidences

The α particles were detected with a ZnS screen coupled to a 5819 photomultiplier tube, and the L x rays were detected with a 3.8×0.1 -cm NaI crystal with a beryllium window. After amplification the output of the detectors was fed into a coincidence unit with 1- μ sec resolving time. The coincidence output operated a linear gate which controlled a parallel L x-ray input into a pulse-height analyzer. The observed intensity of coincident L x rays in source I corresponded to 1.3 $L+M \dots$ electron vacancies per Fm^{257} α particle. The true value would be somewhat larger as coincidences are lost when K x rays, γ rays, or other L x rays enter the L x-ray detector simultaneously with L x rays. The large amount of vacancies for each of the α particles shows in itself that the principal α group (or groups) are in coincidence with more than one highly converted γ ray.

C. α -Particle- γ -Ray Coincidences

The L x-ray detector in the apparatus described above was replaced with a 3-in. \times 3-in. NaI detector. The γ -ray spectrum in coincidence with α particles is shown in Fig. 3. Copious K x-rays of Cf were observed (51% per Fm^{257} α particle) and γ rays of ~ 185 keV, $(8.5 \pm 1)\%$ and 243 keV, $(10 \pm 1)\%$. The K x rays were also measured with a Ge detector (resolution ~ 1.1 keV) and the energies of the $K\alpha_1$ and $K\alpha_2$ x rays were 114.9 and 109.9 keV, respectively. All of these radiations are of sufficient intensity that they must originate from the level populated by the 6.519-MeV α group.

D. α -Particle-Conversion-Electron Coincidences

The electrons were detected with a Li-drifted gold-surfaced silicon detector, 0.3 cm thick and 1.2-cm diam. The α particles were detected through the 0.0001-in. Ni-backing plate with a surface-barrier Au-Si detector. The full width at half-maximum for an electron peak of 100-keV energy was 5 keV. The outputs of the detectors were fed through electronic circuitry similar to those used for the α -particle γ -ray coincidence experiments. The electron spectra in coincidence with α particles were investigated with sources I and II. The

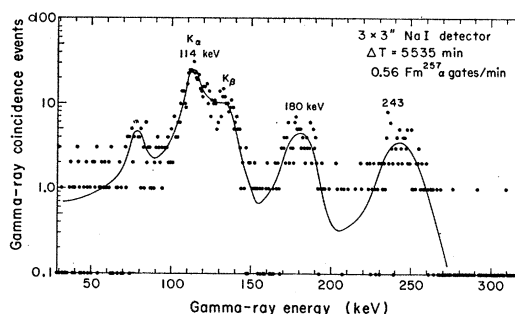


FIG. 3. Fm^{257} α spectrum in coincidence with α particles.

TABLE I. Fm^{257} alpha-spectra data. Energies are in MeV.

$\Omega=40\%$	$\Omega=7.5\%$	$\Omega=4\%$	$\Omega=3\%$	$\Omega=1.8\%$	Best value ^a	HF ^b
			~ 6.34 $\sim 0.4\%$	~ 6.35 $\sim 0.5\%$	$\sim 6.35?$ $\sim 0.5\%$	
	6.43 $\sim 1.4\%$	6.44 $\sim 1.8\%$	6.442 (1.9 ± 0.4)%	6.441 (2.0 ± 0.4)%	6.441 ± 0.004 (2.0 ± 0.3)%	15
~ 6.53 $\sim 35\%$	6.520 75%	6.519 87%	^c 89%	^c 93%	6.519 ± 0.002 94%	0.85
~ 6.57 $\sim 35\%$	6.56 $\sim 13\%$	6.55 $\sim 7\%$	6.56 3.5%	6.56 2%		
~ 6.60 $\sim 20\%$	~ 6.61 $\sim 4\%$	6.61 <3%	6.60 1.4%	6.60 <2%		
~ 6.68 $\sim 6\%$	6.700 (4.2 ± 0.8)%	6.702 (4.0 ± 0.5)%	6.697 (3.1 ± 0.3)%	6.696 (3.3 ± 0.4)%	6.696 ± 0.003 (3.2 ± 0.3)%	1.7×10^2
~ 6.73 $\sim 4\%$	~ 6.76 (1.0 ± 0.3)%	~ 6.75 (0.45 ± 0.2)%	6.75 (0.7 ± 0.15)%	~ 6.76 <1.4%	6.75 (0.4 ± 0.2)%?	$\sim 3.3 \times 10^3$

^a The intensities have been corrected for losses caused by coincidences between α particles and electrons in the α detector. The α -energy standard was Es^{253} (6.633 MeV).

^b The α -decay hindrance factors were calculated with the spin-independent equations of M. A. Preston [Phys. Rev. 71, 865 (1947)] and a Fm^{257} half-life of 96 days.

^c This group was used as an energy standard (6.519 MeV).

TABLE II. Fm^{257} electron lines.

Energy (keV)	Intensity (%)	Shell	Binding energy (keV)	Gamma-ray energy (keV)	Conversion coefficients					Gamma-ray multipolarity
					Experimental	E1	Theoretical ^a E2	M1	M2	
36.8 ± 0.5	31 ± 6^b	L_{II} (L_{II})	26.0 (25.1)	62.8 (61.9)	>19	0.37	152	26	746	($M1, E2, \text{or } M2$)
44.7 ± 0.9	31 ± 6^b	K	134.8	179.8	4.1 ± 0.9	0.11	0.15	5.7	18.5	$M1 (E2?)^c$
54.8 ± 1.7	12 ± 3	M_I	6.8	61.6						
(59?) ^d	~ 6	N_I	1.8	~ 61						
(71?) ^d	~ 3	M_I	6.7	~ 78						
(~ 76) ^d	~ 5	L_I	26	~ 106						
97.0 ± 1.7	2.8 ± 1.4	M_I	6.7	103.7						
106.4 ± 0.7	14 ± 3	K	134.8	241.2	1.4 ± 0.4	0.057	0.11	2.5	7.1	$M1 (E2?)^c$
153.8 ± 0.9	5.0 ± 1.2^b	L_I	26.0	179.8	0.7 ± 0.2	0.026	1.2	1.3	9.1	
171.4 ± 1.6	3 ± 1	M_I	6.7	178						
216 ± 1.6	2.4 ± 1	L_I	26.0	242	0.25 ± 0.1	0.013	0.35	0.55	2.9	
235 ± 1.6	2.6 ± 1	M_I	6.7	242						

^a The theoretical conversion coefficients were taken from the tables of Sliv and Band (Ref. 8). The value listed after the L_I subshells are actually the sum of conversion in the L_I , L_{II} , and L_{III} subshells.

^b In determining the conversion coefficients these intensities were increased 10% to compensate for losses due to coincidences between the 63- and 180-keV transitions.

^c These are considered the most likely assignments, although $E1 - M2$ admixtures would also be consistent with the electron and γ -ray coincidence data.

^d The values are dubious and represent one of a number of possible resolutions of the electron energy groups.

electron spectrum for source II is shown in Fig. 4 and the energies and intensities are tabulated in Table II.

The three most intense electron groups are relatively well defined. The 106-keV group must be due to the K conversion of a 241-keV transition by the process of elimination. If it were an L -conversion line, the M -conversion line (which was not observed) should have been seen. If on the other hand it were an M -conversion line, the L -conversion line (which was not observed) should have been seen. This assignment was confirmed by finding groups (216 and 235 keV) corresponding to the L - and M -conversion lines of a 242-keV transition (Table II).

Similar arguments indicate the 45-keV line must be due to K conversion of a 180-keV transition. The 37-keV electron group cannot be a K -conversion line as there are no corresponding L -conversion lines in sufficient intensity. It could possibly be an M -conversion line, but then the total intensity for the transition would be somewhat larger than 100%, and we would have expected an appreciably larger value than our

measured one of 1.3 for the electron vacancies per α particle. The best assignment for the 37-keV electron group appears to be as an L -conversion line of a 63-keV transition. The observation of groups (59 and 55 keV) corresponding in energy and intensity with the expected values for M and N conversion of a 61-keV γ ray confirms this assignment.

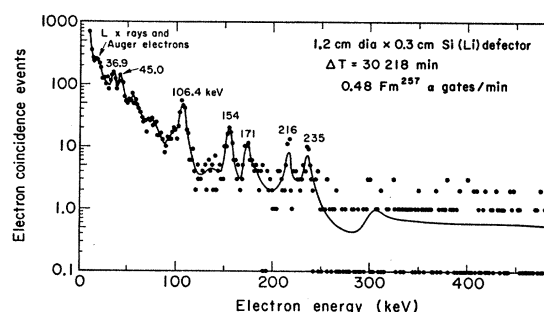


FIG. 4. Fm^{257} electron spectrum in coincidence with α particles.

The other radiations listed in Table II are rather dubious, but they are consistent with transitions of 78 and 103 keV each in about 8% abundance. There is also evidence for coincidence stack-up radiation between the 37- and 45-keV electron groups.

E. Transition Multipolarities

All transitions indicated in this paper were measured with a coincidence resolving time of $\sim 1 \mu\text{sec}$. Therefore $E3$, $M3$, and higher multiplicities can be excluded as their lifetimes would be expected to be much larger than $1 \mu\text{sec}$. From the K -conversion coefficients of the 180- and 242-keV transitions, as shown in Table II, they very likely have $M1$ multipolarity with possible $E2$ admixtures. Our data are not sufficiently precise, however, to rule out $E1$ – $M2$ admixtures. The multiplicities will be discussed further in the theoretical analysis of the decay scheme.

F. Fissions

The fissions in source II were counted over a period of $4\frac{1}{2}$ months. The fissions-to- α ratio was found to be $(2.3 \pm 0.4) \times 10^{-3}$ in good agreement with previous values.¹⁻³ The half-life as measured by the fission decay was found to be 85 ± 25 days.

IV. DISCUSSION

A. Fm^{257} Decay Scheme

The intensities of the 62, 180, and 242 keV transitions are sufficiently large that these transitions all originate with the 6.519-MeV α group. The most reasonable decay scheme consistent with all of our data is shown in Fig. 5. The ground state of Cf^{253} would be expected to have the same Nilsson quantum numbers,⁶ $K\Pi I [Nn_z \Lambda \Sigma]$, as Fm^{255} , $\frac{7}{2} + \frac{7}{2} [613 \uparrow]$.⁷ The ground state and the excited states at 62 and possibly 140 keV can be interpreted as rotational members of this $K = \frac{7}{2}$ band with spins of $\frac{7}{2}$, $\frac{9}{2}$, and $11/2$, respectively. The 62-keV γ ray should then be a rotational transition having $M1$ – $E2$ multipolarity which is consistent with the experimental limitations of either $M1$, $E2$, or $M2$

multipolarity. The rotational constant $\hbar^2/2\mathcal{I}$ deduced from this energy is 7.0 ± 0.1 , which is the largest observed in this region. The 6.75-MeV α group may populate the ground state, but better data than those obtainable from the sources available would be necessary to establish this conclusively.

The 242-keV state which gives rise to transitions with $M1$ components to the spin- $\frac{7}{2}$ and $\frac{9}{2}$ members of the ground-state band would then have positive parity and a spin (and K) of $\frac{7}{2}$ or $\frac{9}{2}$. The only Nilsson state in this region satisfying these requirements would have the quantum numbers $\frac{9}{2} + \frac{9}{2} [615 \downarrow]$. The level at 321 keV would be the first rotational state of this band with spin and parity of $11/2 +$ and the dubious state at ~ 414 could be the second rotational state with spin and parity $13/2 +$. The rotational constant $\hbar^2/2\mathcal{I}$ for this band is 7.1 ± 0.4 , which is also one of the largest in this region.

B. γ -Ray Transition Probabilities

The reduced transition probabilities for γ rays from one state de-exciting to various members of a rotational band should be proportional simply to the square of appropriate Clebsch-Gordan coefficient,

$$\langle I_i L K_i K_f - K_i | I_i L I_f K_f \rangle^2,$$

for states with pure K . The indices i and f refer to the initial and final states, L is the angular momentum associated with the γ ray, I is the nuclear spin, and K is its projection on the nuclear symmetry axis. In Table III we show the calculated relative photon intensities for 241-, 180-, and 103-keV $M1$ transitions de-exciting a $K\Pi = \frac{9}{2} +$ state to the $\frac{7}{2}$, $\frac{9}{2}$, and $11/2$ members, respectively, of a $K\Pi = \frac{7}{2} +$ band. Also shown in Table III are the experimental values including a photon intensity for a 103-keV transition obtained from the electron intensity and the theoretical conversion coefficients⁸ for an $M1$ transition of this energy. It is seen that the transition probability to the spin- $\frac{9}{2}$ state relative to the $\frac{7}{2}$ state is about an order of magnitude smaller than the experimental value. In addition the transition probability to the spin- $11/2$ state is nearly two orders of magnitude smaller than our rough experimental value. Thus the $M1$ transitions cannot

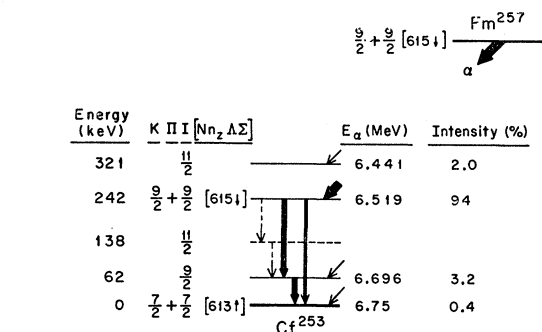


FIG. 5. Decay scheme of Fm^{257} .

⁶ S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 29, No. 16 (1965).

⁷ F. Asaro, S. Bjørnholm, and I. Perlman, Phys. Rev. 133, B291 (1964).

TABLE III. Theoretical intensities of γ rays de-exciting the 242-keV state of Cf^{253} .

Spin of daughter state	γ -ray energy (keV)	Experimental intensity ratios	$M1$ intrinsic transitions between pure states	Theoretical intensity ratios Transitions between the same intrinsic states (Coriolis admixtures)		
				$M1$ ($g_R = 0.30$) ($g_R = 0.6g_I$)	$M1$ ($g_R = 0.20$) ($g_R = 0.6g_I$)	$E2$ (collective) ($g_R = 0.11$)
11/2	103	0.11	0.0018	0.085	0.102	0.0022
9/2	180	0.85	0.092	0.59	0.794	0.11
7/2	241	1	1	1	0.675	0.68

⁸ L. A. Sliv and I. M. Band, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (North-Holland Publishing Company, Amsterdam, 1965), Vol. 2, p. 1939.

proceed simply between states with pure K values but must be caused by admixtures of other K values. One such expected admixture would be caused by the Coriolis interaction between the $K=\frac{7}{2}$ and $K=\frac{9}{2}$ bands. The 242-keV state, which has principally $K=\frac{9}{2}$, will contain a small $K=\frac{7}{2}$ component which can decay by $M1$ (and collective $E2$) transitions to the principal $K=\frac{7}{2}$ component in the ground-state band. In addition, the $K=\frac{9}{2}$ component in the 242-keV band can decay by $M1$ (and collective $E2$) transitions to a small $K=\frac{9}{2}$ component in the ground-state band. These two effects will interfere with each other and with the intrinsic decay between the principal K components. The strength of the interaction is strongly dependent upon the nature of the particular Nilsson orbitals, and, if we ignore pairing, can be calculated from Nilsson wave functions and the energy spacing between the bands. This will be discussed in more detail in the section on alpha decay. The pertinent point found is that the $M1$ transition probabilities due to the Coriolis interaction are not only comparable to the intrinsic values but can be much larger depending on the values chosen for the gyromagnetic ratio of the neutron, g_n , and the collective gyromagnetic ratio, g_R . In Table III we show the $M1$ transition probabilities expected from only the Coriolis-induced transitions without any intrinsic contributions for $g_n = 0.6$ and g_f and $g_R = 0.2$ and 0.3 at a deformation of $\eta = 6$. g_f is the free-nucleon value of the neutron gyromagnetic ratio, -3.82 . We have also calculated the $E2$ transition probabilities due to the Coriolis admixtures assuming a collective enhancement of 200. These values are also included in Table III. With the admixtures deduced in the later section on α decay, the $E2$ transition probabilities between the different intrinsic states are negligible. It is seen from Table II that the experimental γ -ray intensity ratios can be roughly reproduced by considering only transitions due to Coriolis admixtures. In addition it is seen that substantial amounts of $E2$ admixtures are expected, especially in the 242-keV transition. This is consistent with the K conversion coefficients observed for the 242- and 180-keV transitions. With the present rather crude experimental data, speculations on the effect of admixing the intrinsic and Coriolis-induced $M1$ transitions and the best values of g_n and g_R do not seem worthwhile.

C. α Decay

The 6.519-MeV alpha group has a hindrance factor very close to unity. This indicates that the ground state of the parent and the daughter state populated by the α group have the same configuration. Thus Fm²⁵⁷ would be expected to have the same Nilsson quantum numbers as the 242-keV state, i.e., $\frac{9}{2} + \frac{9}{2}[615\downarrow]$. It would ordinarily be possible to calculate the relative population to the other members of the $\frac{9}{2} +$ band in the daughter from appropriate Clebsch-Gordan coefficients and the hindrance factors for various angular-momentum alpha waves determined from adjacent even-even nuclei. Unfortunately, the relative hindrance

factors for $L=2$ and 4α waves are not known for Fm²⁵⁶ and Fm²⁵⁸. We have used instead the values for Fm²⁵⁴ decay along with the equations given in Ref. 7 and have calculated a population of 3.6% for an alpha group to a spin-11/2 group at 319 keV. A theoretical calculation by Poggenburg⁹ predicts a value of 3.89%. These predictions do not compare well with the experimental value of 2.0%. The discrepancy with our calculation may be due to Fm²⁵⁶ and Fm²⁵⁸ having appreciably larger $L=2$ hindrance factors than Fm²⁵⁴.

In order to calculate the expected α -particle intensity to the 62-keV state which would be induced by the Coriolis interaction, it is necessary to know, in both the parent α emitter and the daughter, the matrix elements for the interaction and the energy spacings between the interacting levels. In addition it is necessary to know the magnitudes and the phases of the matrix elements for α decay for both of the interacting states. As the energy spacing between the interacting levels in the parent is not known, we shall only consider the effects due to the admixture of $K=\frac{9}{2}$ in the 62-keV state of the daughter. The ratio of the hindrance factor for favored alpha decay to the hindrance factor for a similar decay to the 62-keV state is then simply the square of the admixture a of $K=\frac{9}{2}$ in the 62-keV state. For small values of $A_K/\Delta E$, then,

$$a^2 = (A_K/\Delta E)^2 (I-K)(I+K+1) = \text{HF}_{\text{fav}}/\text{HF},$$

where HF is the α -decay hindrance factor induced by the Coriolis interaction, HF_{fav} is the hindrance factor for Fm²⁵⁷-favored α decay, A_K is the Coriolis matrix element, ΔE is the spacing between the interacting levels, and $K=\frac{7}{2}$. With a value of $\hbar^2/2\mathcal{I}$ of 7.2 keV, the value of A_K is 5.4 keV as calculated from Nilsson's wave functions. The calculated HF is then $\sim 1.2 \times 10^2$. Considering the approximations mentioned above and the possible effects of pairing correlations on the Coriolis matrix element this is in fortuitous agreement with the experimental value; however, it qualitatively confirms the presumption of a Coriolis interaction between the states.

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⁹ J. K. Poggenburg, Jr., Ph.D. thesis, University of California Lawrence Radiation Laboratory Report No. UCRL-16187, 1965 (unpublished).