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Berkeley, California

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F. Asaro and I. Perlman

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NUCLEAR SPECTROSCOPIC STUDIES OF Am^{257*}

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Chemistry Department
University of California
Berkeley, California

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ABSTRACT

The nuclear radiations associated with the alpha 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±0.2)%?, 6.696±0.003 (3.2±0.3)%, 6.519±0.002 (94±1)%, 6.443±0.003 (2.2±0.3)% and ~6.35 MeV (~0.5)%? Gamma-ray transitions were seen in coincidence with alpha particles with energies of 242, 180, and 62 keV.

The ground state of Fm^{257} is given the Nilsson assignment $9/2+9/2[615\downarrow]$ as is the 242 keV state of Cf^{253}. The ground state of Cf^{253} is given the assignment $7/2+7/2[613\uparrow]$. For these bands in the daughter nucleus the rotational constants, $\hbar^2/2\Im$, 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 alpha-and gamma-ray transitions are interpreted in terms of a Coriolis interaction between the two assigned Nilsson levels in the daughter nucleus.

I. INTRODUCTION

The alpha-emitter Fm^{257} is of some interest because the study of its alpha 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 studies[1,2,3] 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 alpha-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 procedure[4] 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" nickel plate covering an area about 1/8" in diameter.[5] Four fermium sources were prepared in this fashion. Source I contained after chemistry considerable 20 hour Fm^{255}, ~2.3 dis/min of Fm^{257}, ~1.2 dis/min of Es^{253} (impurity), 0.35 dis/min of Cf^{252} (impurity) and ~0.25 dis/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 dis/min of Fm^{257}, source III contained 28 dis/min of Fm^{257} and source IV contained 8.4 dis/min of Fm^{257} after chemistry.

III. EXPERIMENTAL

A. Alpha 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 alpha groups are partially registered in this type of measurement.[3]

The alpha 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 hour 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 alpha 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 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} alpha peak is also very likely caused by intense conversion electron coincidences with the main alpha group. In order to check this possibility the alpha spectrum was measured at solid angles of 7.5, 4, 3, and 1.75 percent with surface-barrier Au-Si detectors coupled to appropriate electronic circuitry. Figure 2 shows the alpha spectra of Fm^{257} taken at a solid angle of 3 percent. The various alpha 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 alpha 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±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±0.2)% after the coincidence effects due to electrons are subtracted. There is a group at 6.441 MeV with an intensity of 2.0% and a rather dubious group at ~6.35 MeV with an intensity of ~0.5%. Neither of these latter two groups could be caused by α-e^- coincidences as their energies are smaller than the main α peak.

B. Alpha Particle-L x-ray Coincidences

The alpha 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 μsec 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 ... electron vacancies per Fm^{257} alpha particle. The true value would be somewhat larger as coincidences are lost when K x-rays, gamma rays, or other L x-rays enter the L x-ray detector simultaneously with L x-rays. The large amount of vacancies per alpha particles shows in itself that the principal alpha group or groups are in coincidence with more than one highly converted gamma ray.

C. Alpha Particle—Gamma Ray Coincidences

The L x-ray detector in the apparatus described above was replaced with a 3" × 3" NaI detector. The gamma-ray spectrum in coincidence with alpha particles is shown in Fig. 3. Copious K x-rays of Cf were observed (51% per Fm^{257} alpha particle) and gamma rays of ~185, (8.5±1)% and 243 keV, (10±1)%. The K x-rays were also measured with a Ge detector (resolution ~1.1 keV) and the energies of the K_{α_1} and K_{α_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 alpha group.

D. Alpha Particle—Conversion Electron Coincidences

The electrons were detected with a Li-drifted gold-surfaced silicon detector, 0.3 cm thick and 1.2 cm in diameter. The alpha particles were detected through the 0.0001" 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 alpha-particle gamma-ray coincidence experiments. The electron spectra in coincidence with alpha particles were investigated with sources I and II. The 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 alpha 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 gamma ray confirm this assignment.

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 ~1 μsec. Therefore E3, M3, and higher multipolarities can be excluded as their lifetimes would be expected to be much larger than 1 μsec. 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 multipolarities 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 4-1/2 months. The fission to α ratio was found to be $(2.3\pm0.4)\times10^{-3}$ in good agreement with previous values[1,2,3]. 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 alpha 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,[8] $K\,\Pi\,I\,[N\,N_z\,\Lambda\,\Sigma]$, as Fm^{255}, $7/2+7/2[613\uparrow]$.[9] The ground state and the excited states at 62 and possibly 140 keV can be interpreted as rotational members of this K=7/2 band with spins of 7/2, 9/2, and 11/2 respectively. The 62 keV gamma 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, $\hbar^2/2\mathfrak{I}$, deduced from this energy is 7.0±0.1 which is the largest observed in this region. The 6.75 MeV alpha group may populate the ground state, but better data than that obtainable from the sources available would be necessary to establish this conclusively.

The 242 keV state which drops transitions with M1 components to the spin 7/2 and 9/2 members of the ground state band would then have positive parity and a spin (and K) of 7/2 or 9/2. The only Nilsson state in this region satisfying these requirements would have the quantum numbers $9/2+9/2[615\downarrow]$. 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, $\hbar^2/2\mathfrak{I}$, for this band is 7.1±0.4, which is also one of the largest in this region.

B. Gamma Ray Transition Probabilities

The reduced transition probabilities for gamma rays from one state de-exciting to various members of a rotational band should be proportional simply to the square of the 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 gamma 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 $K\pi = 9/2+$ state to the 7/2, 9/2, and 11/2 members respectively of a $K\pi = 7/2+$ band. Also in Table III are shown the experimental values including a photon intensity for a 103 keV transition obtained from the electron intensity and the theoretical conversion coefficients[7] for an M1 transition of this energy. It is seen that the transition probability to the spin 9/2 state relative to the 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 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 = 7/2 and K = 9/2 bands. The 242 keV state, which has principally K = 9/2, will contain a small K = 7/2 component which can decay by M1 (and collective E2) transitions to the principal K = 7/2 component in the ground state band. In addition, the K = 9/2 component in the 242 keV band can decay by M1 (and collective E2) transitions to a small K = 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 $g_n = 0.6\ g_f$ 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 of alpha decay, the E2 transition probabilities between the different intrinsic states is negligible. It is seen from Table II that the experimental gamma 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. Alpha Decay

The 6.519 keV 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 alpha group have the same configuration. Thus Fm^{257} would be expected to have the same Nilsson quantum numbers as the 242 keV state, i.e., 9/2+9/2[615↑]. It would ordinarily be possible to calculate the relative population to the other members of the 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 4 alpha waves are not known for Fm^{256} and Fm^{258}. We have used instead the values for Fm^{254} decay along with the equations given in Ref. 9 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 K. Poggenburg[10] 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 Fm^{256} and Fm^{258} having appreciably larger $L = 2$ hindrance factors than Fm^{254}.

In order to calculate the expected alpha particle intensity to the 62 keV state which would be induced by the Coriolis interaction, it is necessary to know, in both the parent alpha 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 alpha 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 $K = 9/2$ 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 $K = 9/2$ in the 62 keV state. For small values of $A_K/\Delta E$ then:

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

where HF is the alpha decay hindrance factor induced by the Coriolis interaction, HF_{fav} is the hindrance factor for Fm^{257} favored alpha decay, A_K is the Coriolis matrix element, ΔE is the spacing between the interacting levels, and $K = 7/2$. With a value of $\hbar^2/2\mathscr{I}$ 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; but qualitatively confirms the presumption of a Coriolis interaction between the states.

V. Acknowledgments

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Footnotes and References

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Table I. Fm^{257} alpha spectra data.

Ω = 40%	Ω = 7.5%	Ω = 4%	Ω = 3%	Ω = 1.8%	Best values[a]	Hf[b]
			~6.34 ~0.4%	~6.35 ~0.5%	~6.35 ? ~0.5%	
	6.43 ~1.4%	6.44 ~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 MeV ~35%	6.520 MeV 75%	6.519 MeV 87%	c 89%	c 93%	6.519±0.002 MeV 94%	0.85
~6.57 ~35%	6.56 ~13%	6.55 ~7%	6.56 3.5%	6.56 ~2%		
~6.60 ~20%	~6.61 ~4%	6.61 <3%	6.60 1.4%	6.60 <2%		
~6.68 ~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 ~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)% ?	$\sim3.3\times10^{3}$

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

[b] The alpha decay hindrance factors were calculated with the spin-independent equations of Preston[6] 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	Theoretical[b]				
						E1	E2	M1	M2	
36.8±0.5	31±6[a]	$L_I(L_{II})$	26.0(25.1)	62.8(61.9)	>19	0.37	152	26	746	(M1,E2, or M2)
44.7±0.9	31±6[a]	K	134.8	179.8	4.1±0.9	.11	0.15	5.7	18.5	M1(E2?)[d]
54.8±1.7	12±3	M_1	6.8	61.6						
(59?)[c]	~6	N_1	1.8	~61						
(71?)[c]	~3	M_1	6.7	~78						
(~76?)[c]	~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	.057	0.11	2.5	7.1	M1 (E2?)[d]
153.8±0.9	5.0±1.2[a]	L_I	26.0	179.8	0.7±0.2	.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	.013	0.35	0.55	2.9	
235±1.6	2.6±1	M_I	6.7	242						

[a] In determining the conversion coefficients these intensities were increased 10% to compensate for losses due to coincidences between the 63 and 108 keV transitions.

[b] The theoretical conversion coefficients were taken from the Tables of Sliv and Band (Ref. 7). The value listed after the L_I subshells are actually the sum of conversion in the L_I, L_{II}, and L_{III} subshells.

[c] The values are dubious and represent one of a number of possible resolutions of the electron energy groups.

[d] These are considered the most likely assignments, although E1-M2 admixtures would also be consistent with the electron and gamma ray coincidence data.

Table III. Theoretical intensities of gamma rays deexciting 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	M1	E2
				$(g_R=0.30)$	$(g_R=0.20)$	(coll.)
				$(g_n=0.6g_f)$	$(g_n=0.6g_f)$	
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

Figure Captions

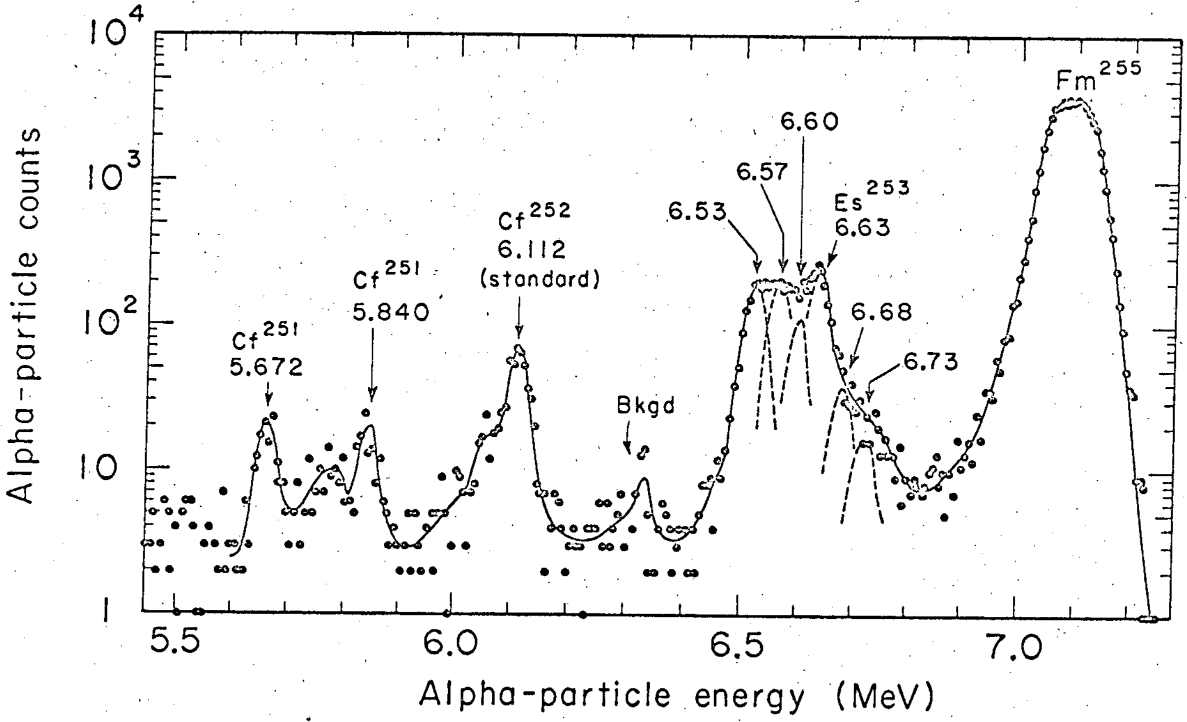
Fig. 1. Fm^{257} α spectrum taken with a Frisch grid chamber (Ω = 40%).

Fig. 2. Fm^{257} α spectrum taken with a Au-Si surface barrier detector (Ω = 3%).

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

Fig. 4. Fm^{257} electron spectrum in coincidence with alpha particles.

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



MUB-14059

Fig. 1.

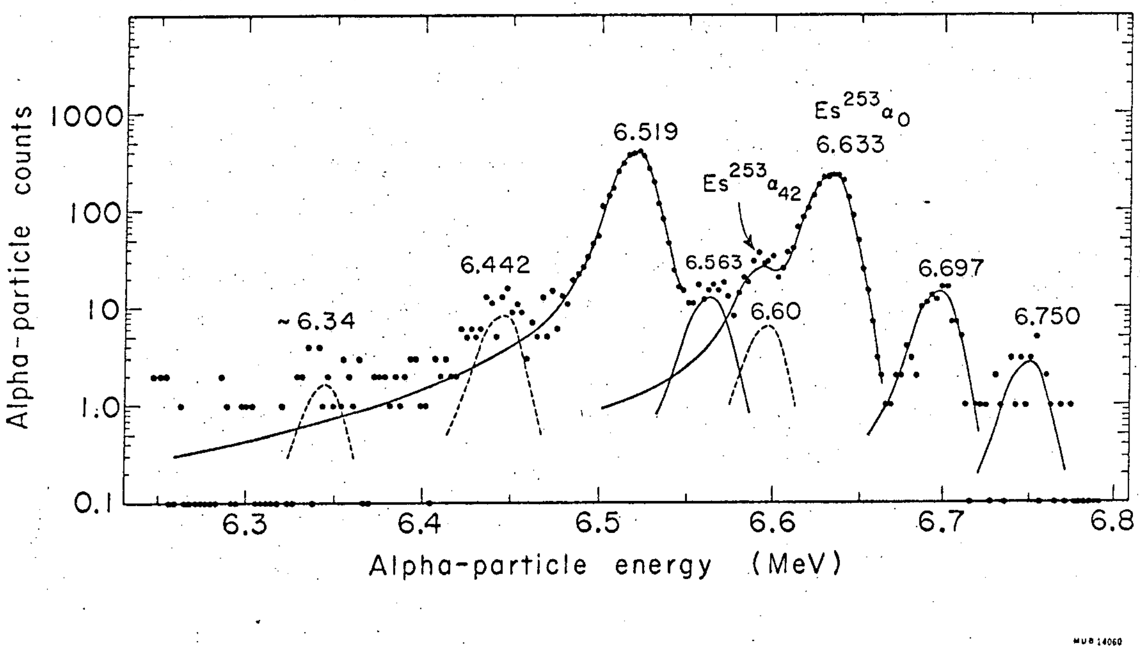

Alpha-particle counts
1000
100
10
1.0
0.1
6.3
6.4
6.5
6.6
6.7
6.8
Alpha-particle energy (MeV)
~6.34
6.442
6.519
6.563
$Es^{253}\alpha_{42}$
6.60
$Es^{253}\alpha_0$
6.633
6.697
6.750
MUB 14060

Fig. 2.

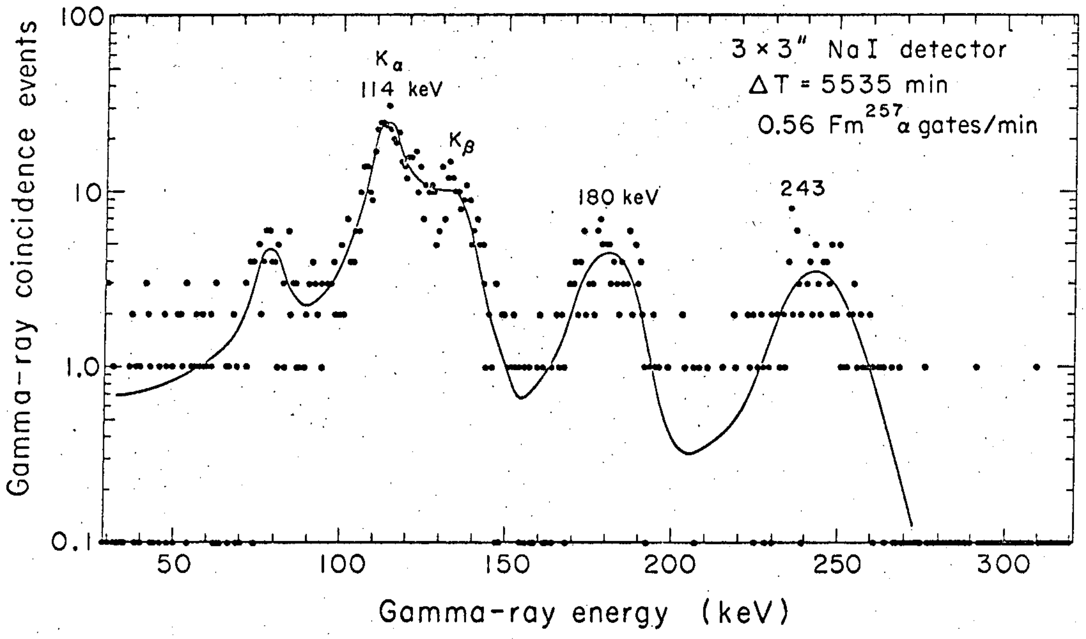
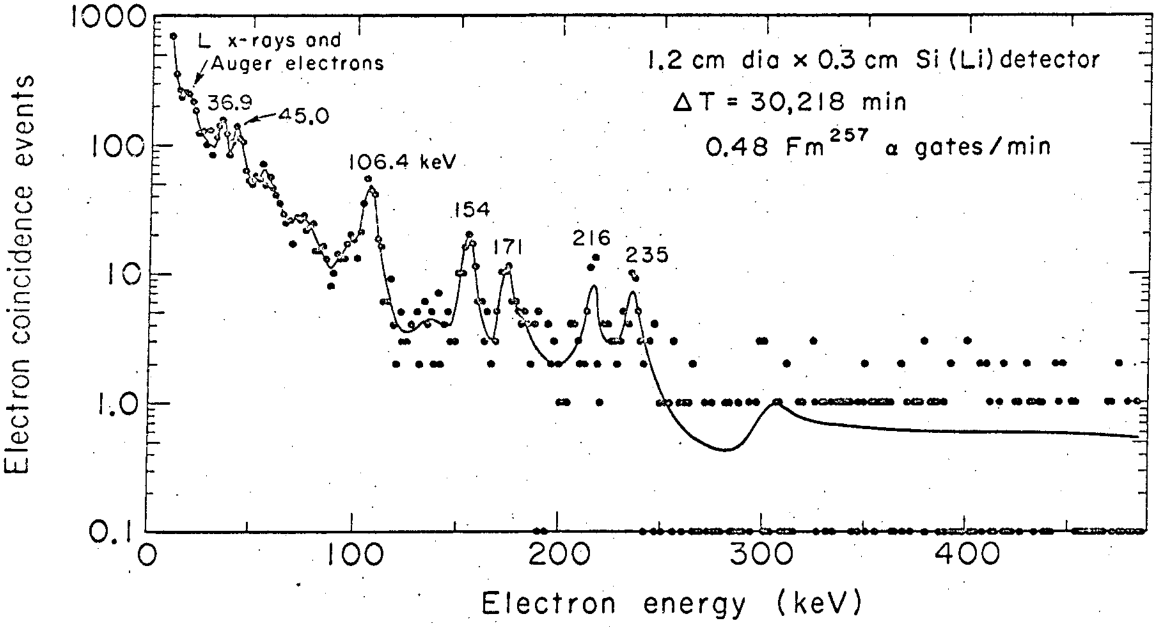

3 × 3" NaI detector
ΔT = 5535 min
0.56 Fm^{257} α gates/min
K$_\alpha$
114 keV
K$_\beta$
180 keV
243
Gamma-ray coincidence events
100
10
1.0
0.1
50
100
150
200
250
300
Gamma-ray energy (keV)
MUB 14061

Fig. 3.



MUB 14062

Fig. 4.

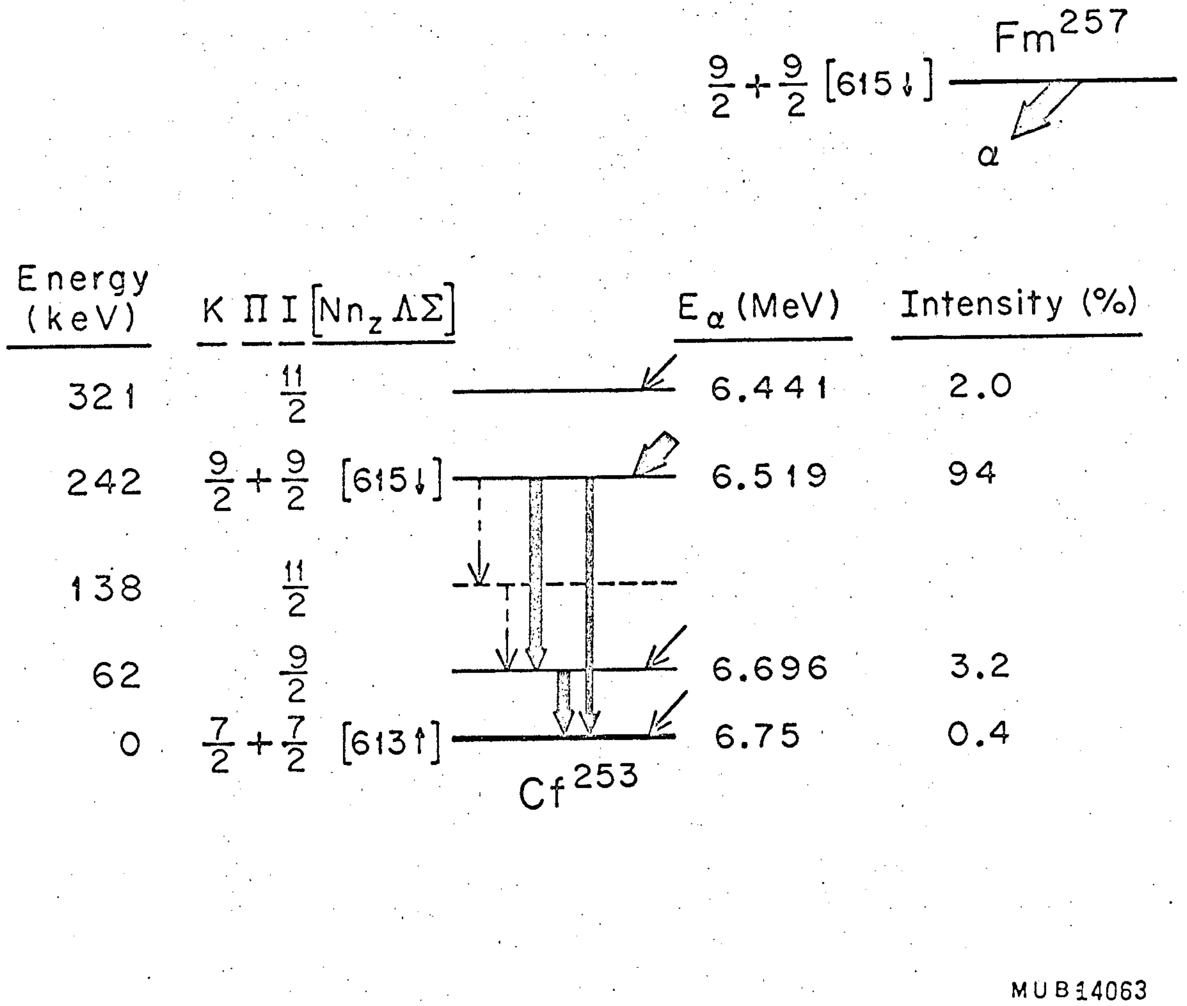

$\frac{9}{2}+\frac{9}{2}$ [615↓]
Fm^{257}
α
Energy (keV)
K Π I [$Nn_z\Lambda\Sigma$]
E_α (MeV)
Intensity (%)
321
$\frac{11}{2}$
6.441
2.0
242
$\frac{9}{2}+\frac{9}{2}$ [615↓]
6.519
94
138
$\frac{11}{2}$
62
$\frac{9}{2}$
6.696
3.2
0
$\frac{7}{2}+\frac{7}{2}$ [613↑]
6.75
0.4
Cf^{253}
MUB 14063

Fig. 5.