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Frank Asaro, Frank S. Stephens, Jr., B. G. Harvey, and I. Perlman

April 11, 1955

Complex Alpha and Gamma Spectra of  $\text{Cf}^{250,252}$

Frank Asaro, Frank S. Stephens, Jr., B. G. Harvey, and I. Perlman  
Radiation Laboratory and Department of Chemistry  
University of California, Berkeley, California

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ABSTRACT

The alpha and gamma spectra of  $\text{Cf}^{250}$  and  $\text{Cf}^{252}$  have been investigated with an alpha particle spectrograph and an alpha particle-gamma ray coincidence counter. Alpha groups of 6.112 (84.5 percent) and 6.069 Mev (15.5 percent) belonging to  $\text{Cf}^{252}$  were found, as were groups of 6.024 (83 percent) and 5.980 Mev (17 percent), belonging to  $\text{Cf}^{250}$ . L x-rays and gamma rays of 42 (0.014 percent) and 100 kev (0.01<sub>3</sub> percent) were assigned to  $\text{Cf}^{252}$ . Other gamma rays were assigned to odd-mass californium isotopes. The results are evaluated with respect to the current theory and systematics of complex alpha spectra and excited states of even-even nuclei.

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I. INTRODUCTION

In the heavy element region the alpha decay characteristics of even-even nuclei show marked regularities with respect to energy level separations and the selective alpha particle population to the various states. The level spacings correlate well with the Bohr-Mottelson<sup>1-3</sup> formulation for collective rotational motion in highly deformed nuclei, and the alpha population to the ground and first even state follow simple alpha decay predictions closely. There are small but significant departures, however, from the simple theory.<sup>4,5</sup>

On the other hand, the alpha population to the second even states varies considerably from the expectations of simple alpha decay theory.<sup>4,6</sup> The ratio of the predicted to the observed population follows a regular pattern which has been treated in terms of the spheroidal model by various authors.<sup>7</sup>

The present work was undertaken to determine to what extent the trends just mentioned would continue in the californium isotopes Cf<sup>250</sup> and Cf<sup>252</sup>. These are beta-stable isotopes with half-lives of ~10 years and ~2 years respectively.<sup>8,9</sup>

II. METHODS AND APPARATUS

A high resolution spectrograph<sup>10</sup> was used to measure the separation between the alpha groups to the ground and first even states and their relative intensities. The transmission of the instrument, however, was

$4 \times 10^{-5}$  so that in the weak sources available only the strongest alpha lines were observed.

The L x-ray intensity of the Cf<sup>250, 252</sup> mixture was determined from alpha-L x-ray coincidences. The measurements were made by coincidence counting because the gamma ray background was comparable to the intensity of L x-rays in the sample whereas the coincidence background was negligible. The alpha detector was zinc sulfide which detected not only alpha particles but also spontaneous fission particles. Compared with the alpha-L x-ray coincidences, however, the spontaneous fission coincidence contributions were very small.

The gamma ray spectrum of the californium isotopes was also measured in coincidence with alpha particles. These coincidence measurements were necessary because of the relatively large numbers of gamma rays from the spontaneous fission branching of the californium isotopes compared to those accompanying the alpha decay.

For alpha-gamma coincidence counting the alpha particle and fission fragment detector was a 1/32-inch thick thallium-activated potassium iodide crystal optically coupled with myvawax to a Dumont-6292 photomultiplier tube. The crystal counting area was masked to a rectangle 5/32 x 1/4 inch with 0.002-inch platinum foil. The output of the photomultiplier tube, after shaping and amplification, was fed into a single-channel pulse-height analyzer. The analyzer selected only the pulses caused by alpha particles and these were fed into a coincidence unit with 2-microsecond resolving time. In some cases the analyzer was adjusted to select only the pulses caused by spontaneous fission fragments. The gamma rays were detected with a 1 x 1 1/2 inch thallium-activated sodium iodide crystal optically coupled to a Dumont-6292





The isotopes listed are beta stable except those designated as  $\beta^-$  emitters. Of the alpha activity in the sample, about 85 percent was due to  $\text{Cf}^{252}$  and 15 percent,  $\text{Cf}^{250}$ . The  $\text{Cf}^{249}$  could be estimated from the irradiation history to contribute only about 0.04 percent to the alpha activity. (The mass of  $\text{Cf}^{249}$  is only about ten fold lower than that of  $\text{Cf}^{250}$  but its half-life is 400 to 500 years whereas the half-life for  $\text{Cf}^{250}$  is 10 years). Despite the low activity of  $\text{Cf}^{249}$ , it has prominent gamma rays which showed up in the gamma-ray spectrum which will be mentioned. Similarly the  $\text{Cf}^{251}$  and the alpha branching of  $\text{Cf}^{253}$ , can be estimated to be in even lower intensity.

After chemical purification of the californium fraction, part of the activity was electroplated on to a 0.0005-inch platinum plate.<sup>13</sup> The plate had been masked so that the activity could deposit only on a rectangular area  $1/32 \times 1/4$  inch. The electrolytic deposit was dried and heated to a dull red color to drive off volatile matter. This sample, containing  $1.2 \times 10^5$  alpha disintegrations per minute, was used as the alpha particle spectrograph source.

The alpha-gamma coincidence source was prepared by evaporating to dryness a few drops of lactic acid solution containing  $1.4 \times 10^5$  alpha disintegrations per minute on a 0.0005-inch platinum plate. The alpha-L x-ray source was prepared by evaporating to dryness a few drops of concentrated nitric acid containing about 2000 alpha disintegrations per minute on a 0.0005-inch aluminum plate.

#### IV. EXPERIMENTAL RESULTS

##### Alpha Spectra

The electroplated sample of mixed californium isotopes described above was exposed in the alpha particle spectrograph for 141 hours. The alpha particles were detected by direct track counting of the photographic plate which was the receiver in the spectrograph. A graph of the alpha spectrum is shown in Fig. 1. The alpha particle energies were determined by comparison with  $E_{\alpha}^{220}\text{O}$  which has been measured by Briggs<sup>14</sup> as 6.282 Mev. The groups at  $6.112 \pm 0.005$  and  $6.024 \pm 0.005$  Mev were assigned to  $\text{Cf}^{252}$  and  $\text{Cf}^{250}$ , respectively, in accordance with earlier measurements with ionization chambers. Thompson et al. reported early values of 6.15 and 6.05 Mev<sup>8,11,15</sup> and later revised them to 6.13 and 6.04 Mev.<sup>16</sup> Diamond et al.<sup>9</sup> reported alpha particle energies of  $6.12 \pm 0.01$  and  $6.03 \pm 0.01$  Mev, and Magnusson et al.<sup>17</sup> reported values of  $6.117 \pm 0.010$  and  $6.033 \pm 0.010$  Mev for these isotopes.

Among even-even alpha emitters in this region there are prominent alpha groups some 40 kev lower in energy than the most abundant groups<sup>4</sup> and in an abundance of 15 to 30 percent.<sup>4,6</sup> By analogy, the groups at 6.069 and 5.980 Mev are assigned to  $\text{Cf}^{252}$  and  $\text{Cf}^{250}$ , respectively. The existence of complex structure in both  $\text{Cf}^{252}$  and  $\text{Cf}^{250}$  was determined previously from L x-ray-alpha particle coincidence measurements by Diamond et al.<sup>9</sup> Magnusson et al.<sup>17</sup> reported the energy separations to be between 40 and 45 Kev.

In order to determine the energy of the first excited state of the product nucleus one must add to the 43-kev separation between the alpha groups of  $\text{Cf}^{252}$ , a nuclear recoil energy difference of 0.7 kev. Thus the energy becomes 44 kev. Similarly, in the case of  $\text{Cf}^{250}$  decay, a 43.6-kev separation

in alpha particle energy corresponds to a 44-kev first excited state energy in the daughter nucleus. To simplify their description, the alpha groups to the ground state are designated  $\alpha_0$ , and those for the 44-kev states are designated  $\alpha_{44}$  (an alpha group to an excited state of x kev would thus be called  $\alpha_x$ ).

The intensities of the alpha groups were determined by integrating the area under the peaks. Although the alpha particles from the electroplated sample were resolved quite well considering the objective of obtaining high transmission, there appears to be an appreciable tailing on the low-energy side of the peaks. This tailing was estimated from  $\alpha_{44}$  of  $\text{Cf}^{250}$ , by assuming that all of the alpha peaks on the photographic plate had very nearly the same shape. After resolving the peaks in this manner intensities for  $\text{Cf}^{252} \alpha_{44}$  and  $\text{Cf}^{250} \alpha_{44}$  were 15.8 percent and 17.5 percent, respectively. From the spectrograph measurements the abundance of  $\text{Cf}^{250}$  alpha activity at the time of the run was 14.2 percent of the total alpha activity. The remaining 85.8 percent was due to  $\text{Cf}^{252}$ . The weighted average of the  $\text{Cf}^{250,252} \alpha_{44}$  groups was then 16.0 percent. This number will be useful for comparison with the alpha-L x-ray coincidence work discussed next.

#### V. COINCIDENCE STUDIES

Alpha-L x-ray coincidences. - The abundance of an alpha group in an even-even nucleus which populates the first excited state can be determined indirectly from the intensity of L x-rays since the corresponding gamma ray transition is by far the most prominent and is heavily converted in the L shell. The intensity is best measured by determining the alpha-L x-ray coincidence rate

per alpha disintegration. The corrections which must be applied to the raw data to obtain the total population of the first excited state are the following: (1) allowance for fluorescence yield of the L x-rays; (2) conversion in M, N--- shells; (3) geometry factor; (4) L x-ray attenuation between source and detector. The geometry factor was determined by measuring the coincidence rate of the alpha particles of  $\text{Am}^{241}$  with its 60-kev gamma ray whose abundance is known.<sup>18</sup> In order to minimize some of the uncertainties in the other correction factors,  $\text{Cm}^{242}$  was used as a standard for comparison. The alpha population to the first excited state for this isotope is known fairly accurately<sup>19</sup> and its alpha-L x-ray coincidence rate was measured under the same experimental conditions. The population of the first excited state defines the total electron vacancies and from the conversion electron spectrum,<sup>20</sup> the number of L-shell vacancies can be calculated. The measured L x-ray intensity must then be brought into agreement with this value by correcting for fluorescence yield and L x-ray absorption. These two factors were dissociated by accepting the calculated<sup>21</sup> fluorescence yield thereby ending up with an effective absorber thickness to produce the required attenuation of the particular L x-rays from  $\text{Cm}^{242}$  decay. Now the desired values for the californium isotopes could be obtained by applying these corrections in reverse fashion. Since the  $\text{Cm}^{242}$  was used as a standard, the errors are only those of second order nature in extrapolating fluorescence yields and x-ray attenuation factors over two units of atomic number.

The results of these measurements and calculations gave 15.5 percent as the weighted averaged for the abundances of the  $\alpha_{144}$  groups of  $\text{Cf}^{250}$  and  $\text{Cf}^{252}$ . This is to be compared with the 16.0 percent value from direct alpha track

counting already cited. The complete independence of the two methods lends some confidence to the alpha-L x-ray coincidence counting method which can be applied to sources of this type which are too weak to obtain the alpha spectrum.

Magnusson et al.<sup>17</sup> reported abundances of about 10 percent for both of these groups from L x-ray-alpha particle coincidence measurements using the abundance of  $\text{Pu}^{238} \alpha_{44}$ , 24 percent, as a standard. A later value of 28 percent<sup>22</sup> for the abundance of  $\text{Pu}^{238} \alpha_{44}$  would then raise the californium abundances of Magnusson<sup>17</sup> to about 12 percent.

#### Gamma rays.

Four measurements of the alpha particle-gamma ray coincidence spectrum were made. The energies and abundances of the observed radiations are shown in Table I. A graph of the results of Exp. 2 are shown in Fig. 2.

Spontaneous fission fragment-gamma ray coincidence measurements were also taken. With our detection system, a gamma ray "continuum" was observed which rose to a plateau at 125  $\rightarrow$  200 kev and then decreased continuously out to several Mev. Superimposed on the continuum was a peak at ~63 kev, which might well have been due to platinum K x-rays produced in the platinum sample mounting by the fission gamma rays. Figure 3 shows the fission fragment-photon coincidences in the region of interest. From this spectrum it seems unlikely that any of the gamma rays shown in Table I are a result of the spontaneous fission process.

The 44-kev transition - The "42-kev" gamma ray is interpreted as belonging to the highly converted transitions of 44 kev leading from the first excited

states to the ground states of both  $\text{Cf}^{250}$  and  $\text{Cf}^{252}$ . The energy measurement from the alpha data is somewhat better than the gamma ray value, so the 44-kev value is preferred. The abundance of this gamma ray is  $1.4 \times 10^{-4}$  per alpha particle (Table I). With the weighted abundance of the alpha groups to the 44-kev levels of 16 percent, the conversion coefficient is  $1.1 \times 10^3$ . Since this value is nearly the same as found for the analogous transition in  $\text{Cf}^{246}$ ,<sup>23</sup> it is reasonable to assume that the corresponding conversion coefficients of  $\text{Cf}^{250}$  and  $\text{Cf}^{252}$  will not differ markedly. Therefore, as the sample activity is predominantly  $\text{Cf}^{252}$ , the measured conversion coefficient applies principally to  $\text{Cf}^{252}$ . By comparing this conversion coefficient with theoretical values<sup>24,25</sup> it is found that the transition can only be E2. The observed value is about a factor of 3 lower than the calculated value<sup>25</sup> for an M2 transition but conservation of parity requires that gamma transition to the ground state of an even-even nucleus from states populated by alpha emission be electric transitions.

Inasmuch as the 44-kev state of  $\text{Cm}^{248}$  (populated by  $\text{Cf}^{252}$  alpha decay) de-excites to the ground state by an E2 transition, the spin of the 44-kev state is 2, even parity. This is, of course, expected since nearly all even-even nuclei are found to have a 2+ first excited state.<sup>26,27,28</sup>

The 100-kev transition - Whenever a photon of ~100 kev is seen in the heavy element region it is necessary to consider the possibility that it may be a K x-ray or a gamma ray or both. For curium, the K x-ray group should produce a peak of approximately 108 kev and it is seen from Fig. 2 that most of the peak at ~100 kev lies at too low an energy. However, the

peak does tail on the high-energy side, and it seems likely that it is not produced by a single photon. In order to get a reasonable minimum value for the intensity of the gamma ray, a peak centering on the K x-ray energy of 108 kev was resolved. The shape of the subtracted peak was obtained by weighting the different components of the K x-ray group according to the relative intensities observed for uranium.<sup>29</sup>

The intensity of the 100-kev gamma ray after subtracting the maximum possible K x-ray contribution was  $1.3 \times 10^{-2}$  percent of the total californium alpha particles. The group attributed to K x-rays has a corresponding maximum intensity of  $7 \times 10^{-3}$  percent. The best energy of the gamma ray is 99 kev; it would be 101 kev if the K x-ray subtraction is not made.

This gamma ray probably represents a transition from the second even spin state\* to the first even spin state of each of the isotopes, Cf<sup>252</sup> and Cf<sup>250</sup>. This assignment is made by analogy with other even-even alpha emitters in this region.<sup>4,30</sup> For example, the gamma spectra of Cm<sup>242</sup><sup>19</sup> and Pu<sup>238</sup>,<sup>22</sup> have gamma rays of about 100 kev which are known to decay from the second to the first even states. Because of the preponderance of Cf<sup>252</sup> in the sample, the measured energy and abundance of the gamma ray apply

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\*The term even spin states refers to the sequence of 2+, 4+, 6+ excited states which is characterized as a rotational band based on the 0+ ground state of an even-even nucleus in this region.

principally to  $\text{Cf}^{252}$  decay. As these values are much the same as found in  $\text{Cf}^{246}$  decay,  $^{24}$  however,  $\text{Cf}^{250}$  would be expected to show in its decay a gamma ray of similar energy and abundance.

Other radiations - Radiations of approximately 67, approximately 180, and approximately 350  $\rightarrow$  400 keV were also observed. The 67-keV radiation is probably due in large part to platinum K x-rays arising from the fluorescence excitation of the sample backing plate by the higher energy gamma rays.

A sample of highly enriched  $\text{Cf}^{249}$  which had grown into previously purified  $\text{Bk}^{249}$  containing small amounts of  $\text{Cf}^{250}$  and  $\text{Cf}^{252}$  was separated and subjected to alpha-gamma coincidence counting. Two gamma rays of 340 and 395 keV with abundances of 16 and 60 percent respectively, relative to the  $\text{Cf}^{249}$  alpha activity were observed. These gamma rays correspond well with some of the radiations seen in the mixed californium isotopes (see Table I). Since the alpha activity due to  $\text{Cf}^{249}$  is estimated to be  $\sim 0.04$  percent, the observed intensities of these gamma rays, 0.03 percent, is not out of line. The gamma ray at 180 keV is not accounted for by the  $\text{Cf}^{249}$  but could possibly come from the  $\text{Cf}^{251}$  or  $\text{Cf}^{253}$ . Considerations of alpha systematics and alpha decay theory, however, indicate it is barely possible for  $\text{Cf}^{253}$  to have an alpha group in as large an abundance as the 180-keV gamma ray.

## VI. DISCUSSION AND SUMMARY

$\text{Cf}^{250}$  and  $\text{Cf}^{252}$  have alpha and gamma spectra which are typical of heavy element even-even alpha emitters. Their decay scheme are shown in



Fig. 4. In both decays the most abundant alpha group goes to the ground state of the daughter nucleus. The decay of  $\text{Cf}^{249}$  and possible  $\text{Cf}^{251}$ , however, indicate a different type of spectra in which the most abundant alpha group populates an excited state in the daughter nucleus which then decays to the ground state by one or more gamma transitions. A corollary to this effect is that the ground state alpha groups in the even-even decay have hindrance factors of about 1, whereas the ground state transitions for odd mass emitters may be hindered\* by many orders of magnitude.

$\text{Cf}^{250}$  and  $\text{Cf}^{252}$  each populate first excited states in their respective daughters of about 44 keV in an abundance corresponding roughly to the predictions of alpha decay theory. More precisely these alpha groups are hindered by about a factor of 3, similar to  $\text{Cf}^{246}$ , somewhat lower than in  $100^{254}$  decay and somewhat higher than in the decay of elements of lower atomic number. The spin and parity of the first excited state populated by  $\text{Cf}^{252}$  decay was deduced from the conversion coefficient to be  $2+$  in common with nearly all other even-even nuclei.

The 100-keV gamma ray in  $\text{Cf}^{252}$  decay is interpreted as the transition from the second even state to the first even state. The energies of the first and second states are such that they can be interpreted as a Bohr-Mottelson rotational band with a consequent spin of  $4+$  for the second even state. The alpha decay to the second even state as deduced from the abundance of the 100-keV gamma ray is lower by over two orders of magnitude from the predictions of spin independent alpha decay theory. This hindrance factor is very similar to that of  $\text{Cf}^{246}$  and appears to follow the general trends observed for corresponding transitions in other even-even nuclides.

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\*"Hindrance" - a "hindrance factor" as used here is defined as the ratio of the expected abundance from alpha decay theory to the experimental abundance.

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Table I

## Alpha Particle-Gamma Ray Coincidence Results

Exp. No.	42-keV gamma ray*		67-keV radiation		100-keV gamma ray		K-ray maximum abundance (x10 <sup>-5</sup> )	~180-keV gamma ray		Other radiations	
	Energy (keV)	Abundance (x10 <sup>-4</sup> )	Energy (keV)	Abundance	Energy (keV)	Abundance (x10 <sup>-4</sup> )		Energy (keV)	Abundance	Energy (keV)	Abundance
1	43	1.5x10 <sup>-4</sup>	66-73	2.6x10 <sup>-5</sup>	99	1.3x10 <sup>-4</sup>	5x10 <sup>-5</sup>	~190	>8x10 <sup>-5**</sup>	†	--
2	41	1.3x10 <sup>-4</sup>	67	4.3x10 <sup>-5</sup>	96	1.0x10 <sup>-4</sup>	7x10 <sup>-5</sup>	†		†	--
3	~40	1.5x10 <sup>-4</sup>	70	1.2x10 <sup>-4</sup>	99	1.4x10 <sup>-4</sup>	9x10 <sup>-5</sup>	~180	2.2x10 <sup>-4</sup>	†	--
4	~40	1.4x10 <sup>-4</sup>	63-67	2.5x10 <sup>-5</sup>	100	1.4x10 <sup>-4</sup>	7x10 <sup>-5</sup>	~180	8x10 <sup>-5</sup>	350-400	3x10 <sup>-4</sup>

† Best Value

*42	1.4x10 <sup>-4</sup>	--	--	100	1.3x10 <sup>-4</sup>	7x10 <sup>-5</sup>	--	--	350-400	3x10 <sup>-4</sup>
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\*This energy value is not as accurate as that derived from the separation of the peaks in the alpha spectrum.

\*\*The gamma ray peak would have been just partially observed in this experiment.

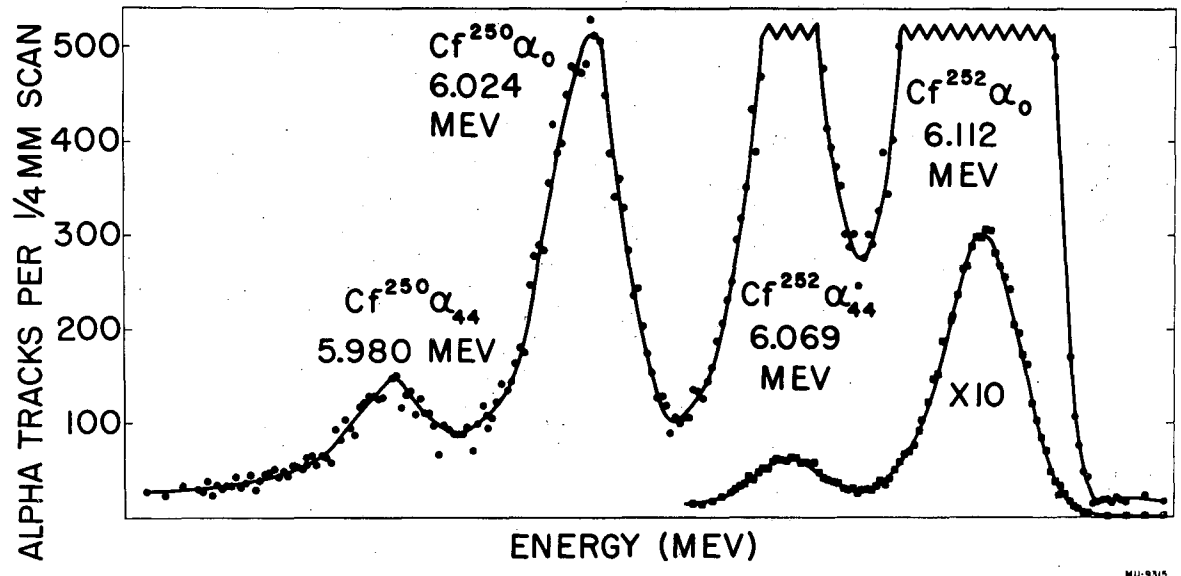
† These gamma rays would not have been observed in this experiment.

† The contribution of gamma rays of 99<sup>253</sup> produced by Cf<sup>253</sup> decay would be less than 10 percent of any of californium gamma rays.

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Fig. 1 Alpha Spectra of  $Cf^{250}$  and  $Cf^{252}$ .

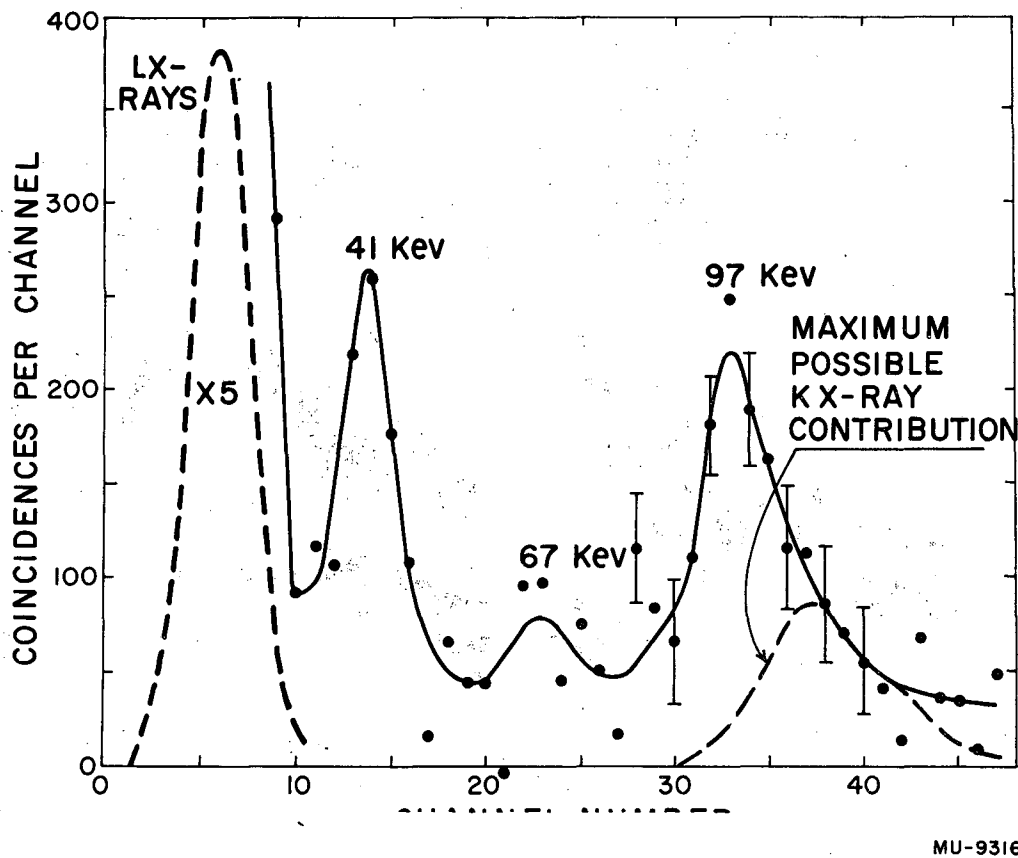
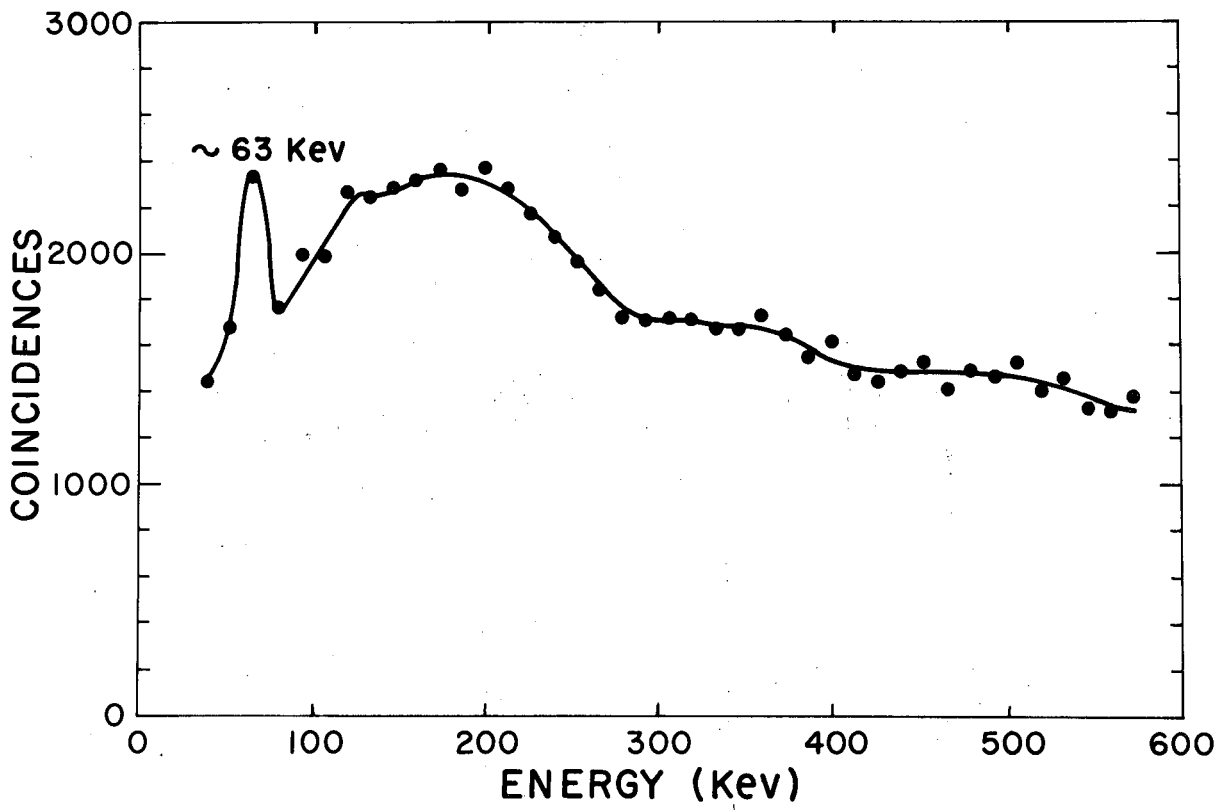


Fig. 2 Alpha particle - gamma ray coincidence spectrum of  $Cf^{250,252}$  from experiment 2 (See Table 1).

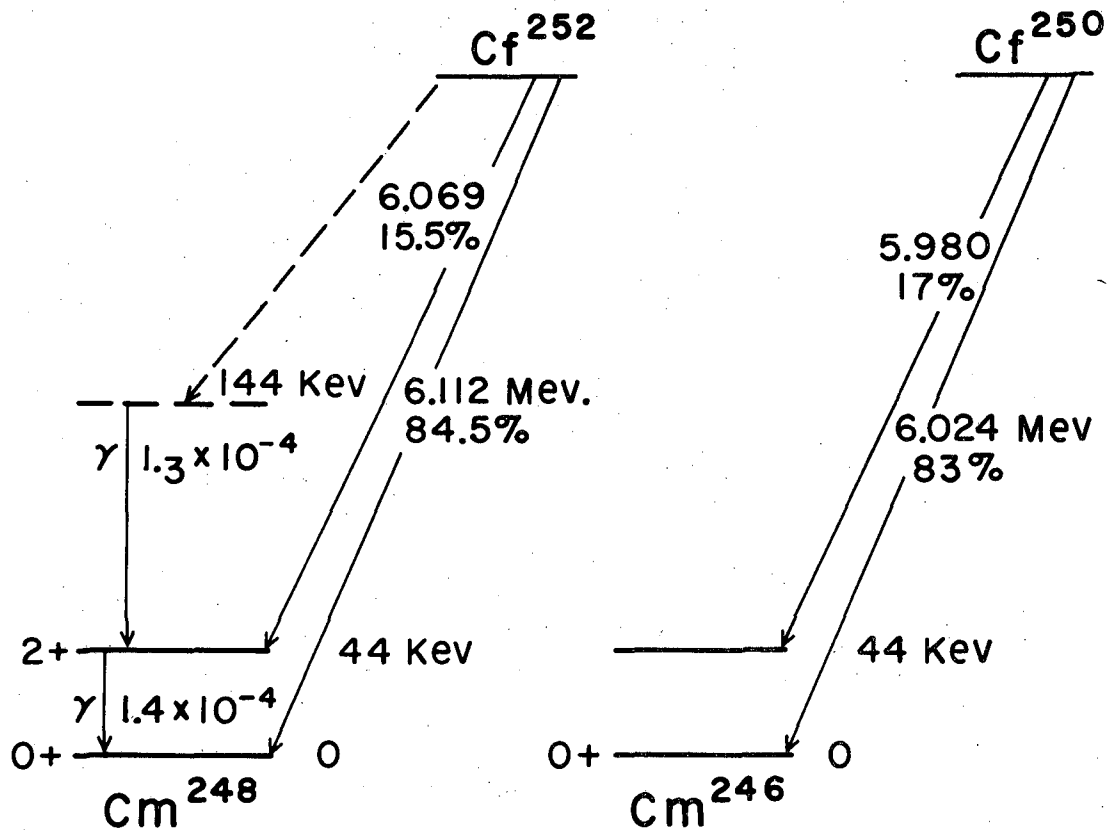
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Fig. 3. Partial spontaneous fission fragment-gamma ray coincidence spectrum of Cf<sup>250,252</sup>.





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Fig. 4 Alpha decay schemes of  $\text{Cf}^{250}$  and  $\text{Cf}^{252}$ .