α decay of the longest-lived Cm isotope: $^{247}_{96}$Cm

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The alpha decay of ^{247}Cm was studied fifty years ago using a 1.6 Bq source containing 99.4% ^{247}Cm, and levels in ^{243}Pu were deduced. Using the same source, γ-ray spectra were measured with a more efficient Ge detector. A γ-ray spectrum was also measured in coincidence with α particles for a period of two months and has 80 times more counts than the previously published spectrum. γ-ray energies were determined with higher precision and four new γ rays were identified. A new high-energy γ ray of 564.5 keV was observed, which is interpreted as a transition between a level at 564.5 keV and the ground state of ^{243}Pu. The 564.5-keV level has been tentatively assigned to the $7/2^-[743]$ Nilsson state.

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I. INTRODUCTION

The longest-lived Cm isotope, which decays with a half-life of 1.56×10^7 y [1] by the emission of α particles, is produced by long irradiation of ^{244}Cm in high-flux reactors. Because of its long half-life and small fraction in Cm sample, its radiations are overwhelmed by the radiations of other Cm isotopes. To observe its radiations, it is essential that the Cm sample be enriched in mass 247 fraction by passing it through an isotope separator. To study the α decay of ^{247}Cm, a 20-mg Cm sample containing 1% ^{247}Cm [1] was obtained from Oak Ridge National Laboratory in late 1960s. This sample was twice mass separated with an electromagnetic isotope separator [2] which produced approximately 1 microgram of 99.4% ^{247}Cm (by atom). To the best of our knowledge, this is the only highly enriched ^{247}Cm source which exists in the world today. Alpha and γ singles spectra of this source were measured and the energies of α groups and γ rays were determined. Alpha-gamma coincidence experiments were performed to determine the relationship between α groups and γ rays. Based on these measurements a decay scheme was constructed and the results of this investigation were published in 1971 [1].

Since more sensitive and advanced detector devices are now available, the γ-ray spectrum of ^{247}Cm was remeasured using the same source. Because Ge spectrometers are very stable, spectra were measured for long intervals of time, both in singles and in coincidence with α particles. The present investigation was undertaken for the following reasons:

1. In the previously published paper, the energy of the $9/2^+$ member of the ground-state band in ^{243}Pu was reported as 56(1) keV which agrees with the value of 55.6(1.5) keV obtained in the ^{242}Pu(d,p) reaction [3]. However, it is in disagreement with the later value of 58.13(22) keV determined in the ^{242}Pu(n,γ) reaction [4]. This energy in the isotone ^{245}Cm is 54.789(13) keV [5] and 55.00(11) keV in ^{247}Cf [6] which are closer to the value for ^{243}Pu obtained in the ^{247}Cm α-decay studies. More precise γ-ray energies are needed to resolve this difference.
2. The 346.0(8)-keV γ ray, which determines the energy of the $9/2^+$ level, was barely visible in the previously published article due to low counts. With more counts in the new spectrum, this energy could be measured with a higher precision.
3. The energy of ^{248}Cm alpha group has now been measured with high precision [7], which enables us to reduce uncertainties in the measured α-particle energies of ^{247}Cm.
4. The energy of the $7/2^-[743]$ Nilsson state is known in the isotones ^{245}Cm and ^{247}Cf. Measurable intensity to this level is expected in long counts and hence this level could be observed in ^{243}Pu.

II. SOURCE PREPARATION

As mentioned in the Introduction, the same ^{247}Cm source used in the 1971 decay studies was used for the present measurements. The procedure for the sample preparation is described in the previous publication [1]. In short, the ^{247}Cm sample was purified by column chromatography and liquid-liquid extraction following the mass separation. The purified material was dissolved in tetra-ethylene glycol, dropped onto a glass disk, evaporated to dryness, and ignited to produce the source. The ^{247}Cm activity in the source was $\approx$1.6 Bq.

III. EXPERIMENTAL METHODS AND RESULTS

A. α-particle energies

The α-particle energies measured in the previously published article were reevaluated. Because the energies of the standards were not known with high precision in 1971, the energies in Ref. [1] were reported with uncertainties of 4 keV.

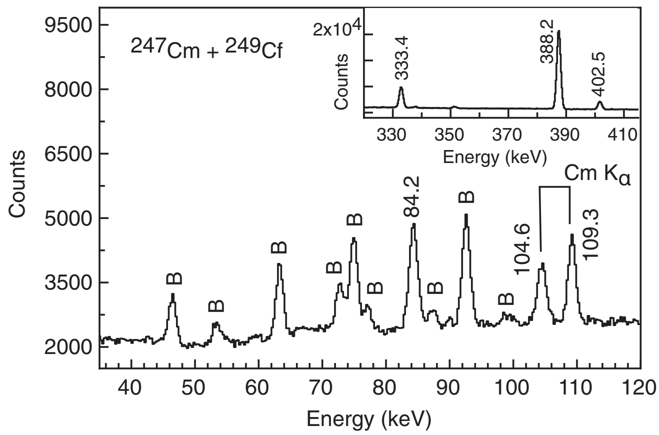

FIG. 1. Gamma-ray spectrum of a mass-separated ^{247}Cm source measured with a 25% Ge detector. A ^{249}Cf source was placed with the ^{247}Cm source for internal calibration. The spectrum was measured for 14 days. ^{210}Pb and ^{234}Th γ rays (denoted by B) are from room background.

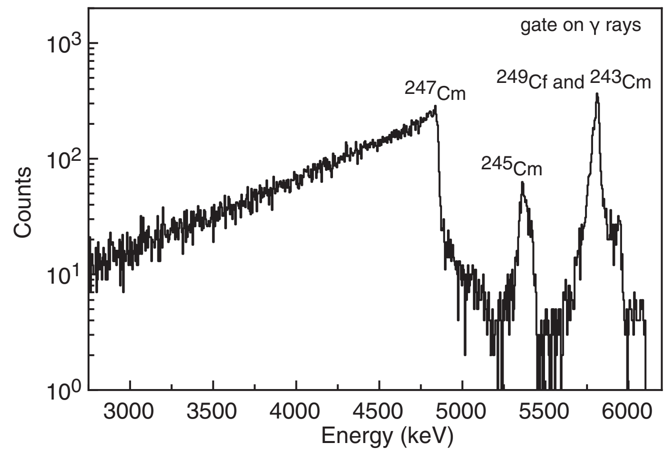

FIG. 2. α-particle spectrum of a mass-separated ^{247}Cm source measured with a 4.5cm^2 PIPS detector. The source was thick, which gave a broad peak for ^{247}Cm. ^{249}Cf, ^{245}Cm, and ^{243}Cm atoms were on the detector face which gave narrow peaks. The spectrum represents only a fraction of the total data.

With newer, more precise energies of the standards, the uncertainties in ^{247}Cm α-particle energies were reduced to 2 keV. Also, the energies of alpha groups feeding the ground-state band have been reduced by 1 keV to 5264(2), 5209(2), and 5144(2) keV. The energy of the favored transition (4868 keV) remains the same but its uncertainty has been reduced to 1 keV. This energy has also been determined with respect to the energy of the ^{248}Cm α_0 group and found unchanged.

B. γ-ray spectroscopy

The energy of the most intense 402-keV γ ray was determined wth high precision by measuring spectra of ^{247}Cm and internal standards with a high-resolution 25% Ge detector. One spectrum was measured with ^{198}Au [411.802 05(17) keV] and ^{51}Cr [320.0824(4) keV] sources for 12 days; another spectrum was measured with a ^{249}Cf source [388.17(2) keV] for 14 days. Using the precise energies of standards reported by Helmer and van der Leun [8], a value of 402.48 ± 0.05 keV was obtained for the strongest γ ray in the decay of ^{247}Cm. γ-ray energy in the decay of the daughter ^{243}Pu ($t_{1/2} = 4.956$ h) was measured as 84.2 ± 0.1 keV with respect to the ^{109}Cd γ ray of 88.0336(10) keV. Low- and high-energy portions of a spectrum of ^{247}Cm and ^{249}Cf, measured for 14 days, are shown in Fig. 1. In the spectrum shown in Fig. 1, room background γ rays are present. The ^{247}Cm γ-ray spectrum was also measured with a 2-cm^2 $\times$ 10-mm low-energy photon spectrometer (LEPS) for 8 days. In the spectrum measured with the Ge detector, the 84.2-keV peak had some contribution from the Pb $K_{\beta1'}$ (84.7 keV) peak but in the LEPS spectrum it was separated from the x-ray peak because of the spectrometer's high resolution. The energy measured with the LEPS agreed with the value determined with the Ge detector.

C. α-γ coincidence spectroscopy

Alpha-gamma coincidence measurement was performed to determine energies and intensities of weaker γ rays. Gamma rays were measured with a 25% Ge detector and α particles were detected with a 4.5-cm^2 passivated, implanted, planar silicon (PIPS) detector. The solid angle for alpha detector was ≈30% of 4π and the source was placed at the closest distance to the Ge detector (≈4 mm). The coincidence events were recorded in event-by-event mode and later sorted with appropriate gates. Data were collected for a 2-month period which gave 80 times more counts in the 402-keV peak than in the previously published work. The alpha detector was previously used with ^{243}Cm and ^{249}Cf sources which contaminated the detector. Hence, the spectrum also contained γ rays in the decays of ^{243}Cm, ^{249}Cf, and ^{245}Cm which provided internal standards. Since the ^{249}Cf source was prepared ≈45 years ago, it contained the daughter ^{245}Cm. The detector has this isotope also and hence ^{245}Cm γ rays are present in the coincidence spectrum. The presence of ^{249}Cf, ^{243}Cm, and ^{245}Cm on the detector face was confirmed by measuring coincidence spectra without the ^{247}Cm source and observing their γ rays.

The coincidence α-particle spectrum is shown in Fig. 2. As the source was thick, the ^{247}Cm spectrum is very broad and shows an exponential slope but no peaks are visible. The ^{243}Cm, ^{245}Cm, and ^{249}Cf atoms were on the detector face and hence gave narrow peaks. The α-particle spectrum in Fig. 2 has three distinct groups. The lowest energy broad peak contains ^{247}Cm α particles, the middle peak belongs to ^{245}Cm α particles, and the highest energy peak contains α particles from ^{243}Cm and ^{249}Cf decays. The γ-ray spectrum gated by the ^{247}Cm α particles is shown in Fig. 3(a), the spectrum gated on ^{243}Cm and ^{249}Cf α particles is shown in Fig. 3(b), and Fig. 3(c) contains γ rays from ^{245}Cm decay. In the spectra, only prominent peaks are labeled; very weak γ rays are known in the spectrum. In Fig. 3(a), the region above 402 keV is extremely clean and hence the very weak γ ray of 564.5 keV was identified. ^{243}Cm and ^{249}Cf γ-ray energies and intensities were used to determine the γ-ray energies and intensities in the ^{247}Cm decay.

The ^{247}Cm γ-ray spectrum in Fig. 3(a) contains the 388.17-keV peak of ^{249}Cf. This is due to the presence of the tail of the

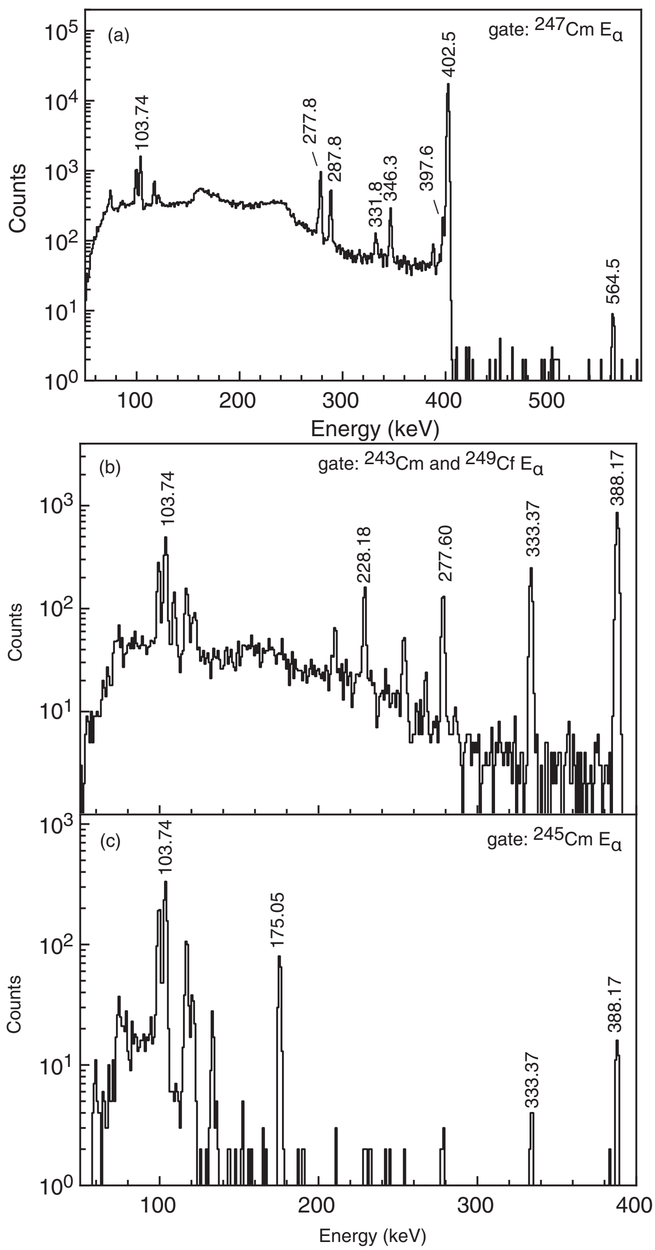

FIG. 3. Gamma-ray spectra measured with a 25% Ge detector in coincidence with α particles in the decay of ^{247}Cm detected with a 4.5 cm^2 PIPS detector. The coincidence events were collected for a period of 2 months. The spectra were generated by placing gates on ^{247}Cm, ^{249}Cf, ^{243}Cm, and ^{245}Cm α groups. The 564.5-keV peak has 30 counts.

^{249}Cf and ^{243}Cm alpha peaks under the ^{247}Cm broad peak. The presence of the 388.17-keV peak indicates that all the peaks of Fig. 3(b) will be present in Fig. 3(a). Thus tiny amounts of the 333.37-keV peak of ^{249}Cf and the 277.60-keV peak of ^{243}Cm will distort the 331.8- and 277.8-keV peaks of ^{247}Cm. For this reason a γ-ray spectrum gated by all α particles above the ^{247}Cm alpha peak and normalized to the 388.17-keV peak area in the spectrum Fig. 3(a) was subtracted from the spectrum in Fig. 3(a). This subtracted spectrum was used to determine energies and intensities of ^{247}Cm γ rays. In Fig. 3(a), there is a

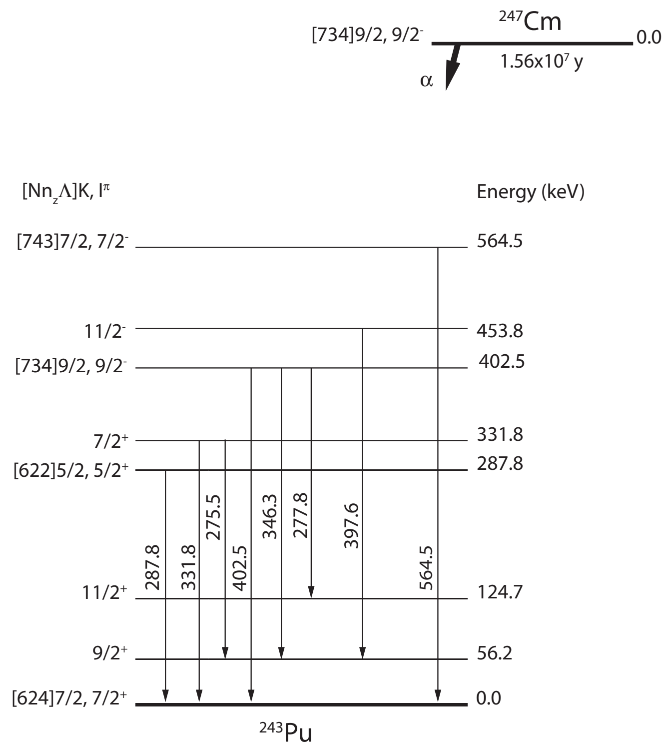

FIG. 4. Partial α-decay scheme of ^{247}Cm. It is same as shown in Ref. [1], but it has one new level at 564.5 keV and has more precise γ-ray energies and intensities.

shoulder on the left side of the 277.8-keV peak whose energy was determined as 275.5 ± 0.4 keV. For absolute intensities, the relative values were normalized to the absolute intensity of the 402.48-keV γ ray to 72% as determined by Fields *et al.* [1] and these are given in Table I. It should be noted that the standard and ^{247}Cm γ-ray energies are so close that there is no contribution to the uncertainties in energies due to the energy calibration. The uncertainties in energies are due to the peak locations only.

IV. DISCUSSION

A. ^{247}Cm α-decay scheme

The decay scheme of ^{247}Cm was deduced by Fields *et al.* in 1971. The arguments for the single-particle assignments are presented there. However, more experiments have been performed since that publication which provide further support for those assignments. The level structure of ^{243}Pu is expected to be similar to the structure of the well-studied isotone, ^{245}Cm [5]. The ground-state spin of ^{247}Cm has been measured [9] as 9/2 and has been assigned to the $9/2^-[734]$ single-particle state. Thus the 402.48-keV level which receives favored alpha transition should have $9/2^-[734]$ assignment. Its $11/2$ member is identified at 453.8 keV.

The ground state of ^{243}Pu was given an assignment of $7/2^+[624]$ Nilsson state from its β^- decay properties [10,11]. The γ-ray energies from the decay of the $9/2^-$ level at 402.48 keV give the energies the $9/2$ and $11/2$ members of the

TABLE I. Energies and intensities of γ rays produced in ^{247}Cm α decay. The 84.2-keV γ-ray intensity is from Ref. [1].

Energy (keV)	Intensity (photons per 100 ^{247}Cm α decays)	Transition (initial level → final level)
84.2 ± 0.1	23 ± 2	^{243}Pu γ ray
99.5 ± 0.1	1.20 ± 0.10	Pu $K_{\alpha 2}$
103.7 ± 0.1	1.90 ± 0.16	Pu $K_{\alpha 1}$
117.0 ± 0.2	0.72 ± 0.07	Pu $K_{\beta 1'}$
120.7 ± 0.3	≈0.25	Pu $K_{\beta 2'}$
275.5 ± 0.4	≈0.3	331.8 → 56.2
277.8 ± 0.2	2.20 ± 0.20	402.48 → 124.7
287.8 ± 0.2	1.30 ± 0.10	287.8 → 0.0
331.8 ± 0.2	0.30 ± 0.06	331.8 → 0.0
346.3 ± 0.2	0.9 ± 0.1	402.48 → 56.2
397.6 ± 0.3	0.7 ± 0.1	453.8 → 56.2
402.48 ± 0.05	72 ± 6 (norm)	402.48 → 0.0
564.5 ± 0.4	0.071 ± 0.015	564.5 → 0.0

ground-state band. Using these energies and the rotational energies formula, we determine the K value of this band as $7/2$, in agreement with the assignment of $7/2^+$[624]. The higher-spin members of the ground-state band have been identified in ^{244}Pu(^{208}Pb, ^{209}Pb γ) reaction studies [12].

The 287.8-keV level decays by an $M1$ transition to the ground state [1]. Hence it must have positive parity. Since no transition is observed to the $9/2^+$ member of the ground-state band, the most likely spin is 5/2. It is given $5/2^+$[622] assignment. Its 7/2 member is identified at 331.8 keV. These assignments are further supported by the ^{242}Pu(d,p) and ^{244}Pu(d,t) transfer reaction studies [4]. The 287.8-keV γ ray was also observed in the β^- decay studies of ^{243}Np [13] and ^{242}Pu$(d,p\gamma)$ reaction [14].

The decay scheme of ^{247}Cm with the energies measured in the present study is shown in Fig. 4. A very weak γ ray of 564.5 keV has been observed in the coincidence spectrum. We interpret this γ ray as the decay of the $7/2^-$[743] state to the ground state. This Nilsson state is identified in the isotones ^{245}Cm and ^{247}Cf at 643.5 and 678.0 keV, respectively, and the strongest γ ray in its decay is to the ground state. Because of the expected low intensity of the α group populating the 564.5-keV state, this α group has not been observed. The intensity of this alpha group is estimated to be 0.18% using the average of hindrance factors in ^{249}Cf and ^{251}Fm alpha decays. Although we do not have sufficient data to make a definite assignment, the similarities in γ-ray energies and intensities to those in ^{245}Cm and ^{247}Cf make it a fairly reasonable assignment.

B. Summary

Gamma-ray energies and intensities in the α decay of ^{247}Cm were measured with high precision with a high-resolution germanium spectrometer. From precise energies of γ rays we have determined the energy of the 9/2 member of the ground-state band as 56.2 ± 0.2 keV which is 2 keV lower than the value of 58.13 ± 0.22 keV determined from (n,γ) studies. The energy of the 11/2 member is measured as 124.7 ± 0.2 keV, which is in good agreement with the value of 124.9 keV calculated with the rotational formula. The present measurements confirm the single-particle assignments made in the previous publication. It should be noted that the relative intensities of γ rays deexciting the $9/2^-$[734] level are dramatically different from respective intensities in ^{245}Cm and ^{247}Cf. A new γ ray of 564.5 keV was observed and it is interpreted as the 564.5 → 0.0 transition. The 564.5-keV level is tentatively assigned to the $7/2^-$[743] Nilsson state.

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