Gamma Rays of 100^{254}†

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Gamma rays of 42±4 (2×10^{-2} percent) and 94±2 kev (2.8×10^{-2} percent) were found in coincidence with alpha particles of 100^{254}. The L x-rays (from the internal conversion of the 42-kev gamma ray) were measured and from the intensity the population of the first excited state was calculated to be 15±2 percent. The gamma rays are interpreted as cascading transitions resulting from the de-excitation of Bohr-Mottelson rotational states having spins of 2 and 4, even parity.

The abundances of the alpha transitions to the spin 4 states of even-even nuclides in this region (after normalizing for differences in energy separation from the ground state) exhibit a pronounced minimum for curium emitters and progressively increase for emitters of higher and lower atomic number.

INTRODUCTION

THE alpha-decay properties of even-even nuclei in the heavy-element region present a remarkably uniform picture in several aspects. The energy levels of the product nuclei as defined by analysis of the alpha spectra have spacings which correlate well with the Bohr and Mottelson[1–3] formulation for collective rotational motion in highly deformed nuclei. Other regularities observed are concerned with the degree of population of the several states and these, of course, must receive explanation through alpha-decay theory. As an example, Rasmussen[4] has obtained semiquantitative explanation for the population of the second rotational states in terms of nuclear quadrupole interaction with the emerging alpha particle wave. The interaction is such that a large intrinsic quadrupole moment can suppress the population of the second rotational state ($l=4$ transition) as compared to the $l=2$ and $l=0$ transitions to the first rotational state and the base state of the band, respectively.

In observing the trends of these properties, it is of interest to obtain data over as broad a range of mass numbers and atomic numbers as possible. The present communication is on 100^{254},[5–8] which is the heaviest nuclear species yet available for such studies.[9] The amount of this isotope which has been prepared is too small for measurement of the alpha spectrum with a high resolution spectrograph, but the gamma rays can be measured by α-γ coincidence counting and bear such close resemblance to those of other even-even nuclei which have been examined carefully that the decay scheme can be deduced with some confidence.

The isotope 100^{254} is prepared by the intensive neutron irradiation of lower elements, ultimately uranium.[5,10] Of interest here is the fact that californium isotopes, arising from β^- decay of lower elements, capture neutrons successively until the 20-day β^- emitter, Cf^{253}, is reached. This decays to 99^{253} which is an alpha emitter of 20 days half-life which captures a neutron to give the 36-hour β^- emitter 99^{254}. The 3-hour α emitter 100^{254} soon grows into equilibrium with its parent. In order to take advantage of the longer half-life of 99^{254} in working on 100^{254}, chemical isolation of the 99 fraction was effected. Such preparations therefore contained:

(1) 99^{253}: 20-day half-life, 6.64-Mev α;

(2) 99^{254}: 36-hr half-life, β^-;

(3) 100^{254}: 3.2-hr half-life, 7.22-Mev α.

EXPERIMENTAL RESULTS AND DISCUSSION

For the present measurements two preparations were employed. One of these was of low intensity and consisted of 7000 disintegrations per minute (dis/min) of 99^{253} and 300 dis/min of 36-hour 99^{254} in equilibrium with its daughter, 3.2-hour 100^{254}. The sample was mounted on an aluminum plate 0.5-mil thick. The second sample was more intense and the 100^{254} had been separated chemically from element 99 and was essentially isotopically pure. At the start of measurements it contained 30 000 dis/min.

The weak source (containing mixed radioactivities) was the first one available and was used to establish the population of alpha transitions to the first excited state (see Fig. 1). On the assumption that the decay

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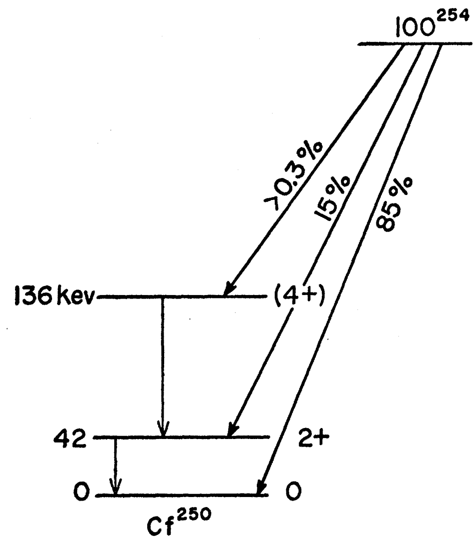

FIG. 1. Decay scheme of 100^{254}.

scheme of 100^{254} is not unlike that of other even-even nuclei such as Cf^{246},[11] Cm^{242},[12] and Pu^{238},[13] this transition should lead to a 2+ state some 40 kev above the ground state and this state would be de-excited by a highly converted gamma-transition. The measurement of the L x-ray intensity would therefore form the basis for calculating the population of the 2+ state.

The alpha-particle—L x-ray coincidence rate was determined in a manner to be explained presently. The geometry of the arrangement was calibrated using the well-known 60-kev γ ray of Am^{241}. The absorption and fluorescence yield[14] corrections were made as second-order corrections by comparison in the same apparatus with the somewhat softer L x-rays from the decay of Cm^{242}. The final corrected intensity of the transition to the 2+ state was 15±2 percent. This is significantly lower than is the case for some lighter elements as will be explained below.

The coincidence counting mentioned above was done with a sodium-iodide crystal and 50-channel pulse height analyzer for the photon side and a thallium-activated potassium iodide crystal ($\frac{1}{32}$ in.$\times\frac{1}{4}$ in. $\times\frac{1}{4}$ in.) with a single-channel pulse height selector for the alpha-particle side. The α-γ coincidence counting *per se* discriminated against the gamma rays from 99^{254} decay. To eliminate coincidences with the alpha particles of 99^{253}, the pulses from the potassium-iodide crystal were fed into the single-channel pulse height selector and those within a chosen energy band were used to gate the 50-channel pulse analyzer. (The potassium-iodide crystal could give an alpha-particle energy resolution of 5 percent without difficulty at low geometry but was 7.6 percent in the manner used here.)

The second preparation which contained no 99^{253} was used for an examination of the gamma-ray spectrum. It had been mounted on a 0.5-mil platinum plate. Coincidence counting was again employed in order to decrease background effects, but since alpha-energy discrimination was not necessary, a zinc sulfide screen on the face of a photomultiplier tube could serve as a high-geometry alpha-particle detector.

The composite data from three measurements of the alpha-gamma coincidence spectrum is shown in Fig. 2.‡ In addition to the prominent L x-ray peak, two gamma rays of 42±4 kev and 94±2 kev were seen. The decay period of the L x-rays and the 94-kev gamma ray could be measured with some accuracy and both showed half lives of about 3 hours. The peak at 66 kev is seen when any emitter of gamma rays of sufficient energy is counted on platinum and is due principally to K x-rays of platinum produced by fluorescence excitation. There may be small contributions at this energy from the escape peak of the 94-kev gamma ray and also from the backscattered radiation. The line showing the "maximum contribution from spontaneous fission" was determined in the following manner. The isotope Cf^{252} which has a high ratio of spontaneous fission to alpha disintegration[15,16] had been used to determine the gamma-ray spectrum in coincidence with the fission process as it registers in the detection system employed here. The gamma-ray intensity as a function of energy in the range of interest here was flat. It was assumed that this distribution would be the same for 100^{254} and the calculated intensity is shown in Fig. 2 as a maximum value since all of the spontaneous fission fragments may not have been counted by the zinc sulfide detector. In calculating the intensities of the gamma

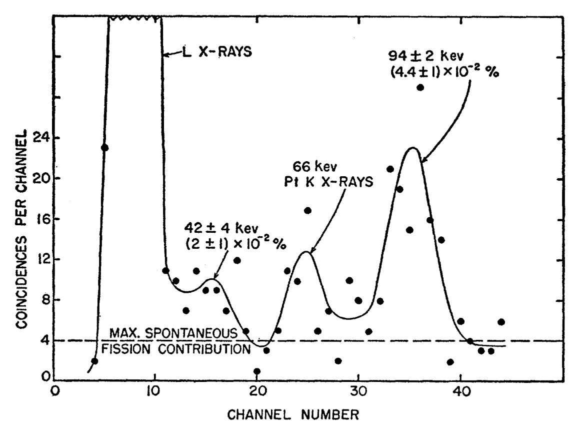

FIG. 2. Alpha-particle—gamma-ray coincidence spectrum of 100^{254}.

‡ *Note added in proof.*—Improved data has shown the abundance of the 94-kev gamma ray to be 2.8×10^{-2} percent. The alpha group to the 136-kev state then has an abundance >0.2 percent.

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rays from the alpha decay process, we assumed a spontaneous fission gamma-ray background of one-half the maximum value shown in Fig. 2. This assumption could introduce a 10 percent error in the intensity of the 94-kev gamma ray and a correspondingly larger error for the 42-kev peak.

The 42±4 kev gamma ray did not give a sufficient number of events to determine its energy with accuracy, hence the large limits of error. Its intensity was 2×10^{-2} percent (of the alpha disintegrations), and under the very likely assumption that the gamma ray is from the transition giving rise to the L x-rays, the conversion coefficient can be calculated. It will be recalled that L x-ray measurements indicated 15 percent population of the first excited state; therefore the conversion coefficient is 750. Within the accuracy of our information this is in good agreement with an $E2$ transition similar to that found for all other even-even nuclei in this region.

The 94-kev transition also has its analogy in the alpha decay of other heavy even-even nuclei and has been placed accordingly in the decay scheme (Fig. 1‡) defining a state at 136 kev. The energy spacing agrees with the assignment to a 4+ state as part of a rotational band according to the Bohr-Mottelson theory.[2,3]

The intensities of alpha transitions to the second even-spin states of even-even nuclei have been discussed in previous publications[4,17] and are of interest because they are not explained by previous alpha-decay theory. It is found that the intensities of alpha transitions to these states are much lower than would be calculated from simple alpha-decay theory and follow a trend illustrated by Fig. 3. Here the ratio of expected alpha intensity to measured intensity is plotted as the "hindrance factor" on the ordinate scale.

The point for 100^{254} was determined according to the following reasoning: it is assumed that the 94-kev transition is analogous to the 103-kev transition[12] of Cm^{242} and has the same conversion coefficient. From the measured gamma-ray intensity, 4.4×10^{-2} percent,

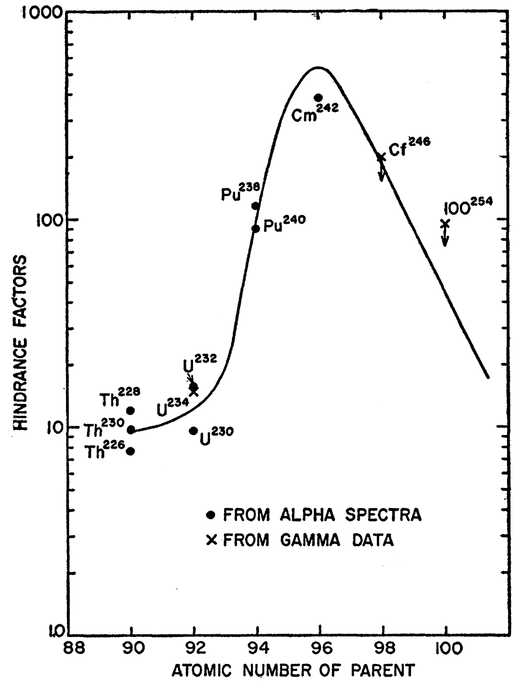

FIG. 3. Hindrance factors for alpha groups to the second even-spin state of even-even nuclei.

the calculated intensity of the alpha group populating this state turns out to be at least 0.3 percent. From this the hindrance factor can be calculated as indicated in Fig. 3.

Rasmussen[4] has obtained semiquantitative explanation of this phenomenon by taking into account the interaction between the outgoing alpha-particle wave and the intrinsic nuclear quadrupole moment. He finds that the population of the 4+ state should be depressed depending upon the magnitude of the quadrupole distortion. It is also of interest that the coupling should cause alpha-particle waves of each angular momentum value to go through a node which may explain the maximum effect in the region of curium alpha emitters.

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