Alpha decay of 231 U to levels in 227 Th

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F45071 Orleans, Prance (Received 26 October 1993)

The fine structure in the alpha decay of 231 U has been studied for the first time. The alpha branching ratio in ²³¹U is determined as $(4 \pm 1) \times 10^{-5}$. Levels in ²²⁷Th are deduced and tentative spins and parities suggested.

PACS number(s): 21.10.—k, 23.20.Lv, 23.60.+e, 27.90.+b

Recently it has been pointed out [1] that there is considerable controversy about the ground-state spin of 227 Th. This spin is quite important, because it is used in nuclear orientation experiments to determine the spins of a variety of levels in 2^{23} Ra following alpha decay. For this purpose $J^{\pi} = 3/2^{+}$ had been assumed [2] as suggested in earlier compilations [3,4]. However, other studies of 223Ra [5,6] following alpha decay assumed the groundstate spin-parity of 227 Th to be $1/2^+$ because of the low alpha hindrance factors to the $K^{\pi} = 1/2^{\pm}$ parity doublet bands in ²²³Ra. Furthermore, the ground-state spinparity of the isotone 2^{25} Ra is $1/2^+$. In the most recent compilation $[1]$, the assigned spin and parity of 227 Th are given as $1/2^+$ but in parentheses because of the uncertainty. If, however, the spin of 227 Th is 1/2, the nuclear orientation correlations are expected to be isotropic, in serious contradiction to the data [2].

One of the methods of studying the levels in ²²⁷Th is to look at the alpha decay of $231U$. The alpha branching was measured in 1950 by the ion chamber method to give the very small value of 0.0055% [3]. For this reason the fine structure of the alpha decay and corresponding levels in ²²⁷Th have not previously been observed. Because of the much better resolution available today, it should be possible to study the fine structure in the alpha decay of 231 U and also the associated gamma transitions.

The 231 U source was produced by bombarding a 0.7 mm thick 232 Th metal foil with 45 MeV alpha particles over a period of 8.5 h with a beam current of 3 μ A in the CERI Orleans cyclotron. The production of 231 U from the ${}^{232}\text{Th}(\alpha, 5n)$ reaction is not maximized at 45 MeV bombarding energy, but this is the maximum beam energy available. Even though the half-life of $232U$, produced by the $(\alpha, 4n)$ reaction, is 72 yr, because of its 100% alpha decay it is a serious unavoidable contaminant. Fortunately, the weak alpha groups of 231 U are situated at ≈ 150 keV higher energy.

A chemical separation of U was carried out using successive anion exchange columns and liquid-liquid chromatography. After dissolving the target in hot concentrated HCl, the bulk of the target was eliminated by fixing the U fraction on a Dowex 1×8 column (200–400 mesh) with 10M HC1. After repeated washings of the column with $10M$ HCl and $4M$ HCl, which eliminated the Th and most of the fission products, the U fraction was eluted with 0.5M HCl.

After evaporation to dryness, the U fraction was dissolved in 10M HC1 and fed onto the top of a small Tefion column (100 mm length, 3 mm diameter) filled with anion exchange resin. The last traces of protactinium were eluted with $10M$ HCl and $1M$ HF. Uranium was recovered, together with iron impurity, using a mixture of $2M$ HCl and $1M$ HF while the Mo fission product fraction remained on the top of the column.

A final purification of the U fraction utilized a column with Diethyl hexyl phosphoric acid adsorbed on Teflon as the stationary phase. After fixation in 1M HCl, iron was eliminated by washings with 6M HCl and finally the U fraction was recovered in 2 ml of 10M HCl. The separated U activity was evaporated to dryness and then heated in vacuum and deposited on 30 μ m foil.

The alpha spectra and gamma spectra in coincidence were measured in 180' close geometry. The alpha detector consisted of an ion-implanted Si detector, 2 cm^2 and 100 μ m thick. The gamma detector consisted of an intrinsic Ge crystal, 4 cm^2 and 1 cm thick with a 0.1 mm Be window.

The resulting alpha spectra are shown in Figs. $1(a)$ and $1(b)$. Figure $1(a)$ presents the alpha spectrum of 230 U (and its daughters), 231 U, 232 U, and 233 U produced in the alpha bombardment of 232Th . Figure 1(b) is a close-up of the alpha spectrum in the vicinity of 231 U. It is indeed possible to observe the fine structure in 231 U. We see alpha groups of 5471 ± 3 , 5456 ± 3 and 5404 ± 4 keV, with intensities of 34 ± 4 , 40 ± 4 and $26\pm3\%$ respectively. The previously observed alpha decay of 231 U gave 5.46 MeV with no fine structure [3]. This agrees with the strongest alpha group in the present experiments.

It should be noted that it is necessary to make the chemical separations, produce the source, and take the spectrum as quickly as possible. In spite of the 72 yr 2^{32} U half-life, alpha activity from its daughter 2^{28} Th increasingly obscures the 231 U alpha spectrum because the known 228 Th alpha group at 5423 keV [3] lies just between the 5456 and 5404 keV alpha groups of 231 U.

We have also been able to measure the β^{-}/α branching ratio in 231 U. This was done in the following way. The $232U/231U$ alpha intensity ratio was determined using Fig. 1(b). Then, after the complete beta decay of U into 2^{31} Pa, the 2^{32} U/ 2^{31} Pa alpha intensity ratio is obtained. One then takes the ratio of 232 U/231Pa to 232 U/ 231 U and corrects for the relative half-lives to get the branching ratio. The value obtained is $(4 \pm 1) \times 10^{-5}$ in relatively good agreement with 5.5×10^{-5} [3] previously observed. It must, however, be noted that if there is (are) high intensity alpha group(s) hidden in the 232 U alpha group [Fig. 1(b)], the branching ratio would increase. The value determined then is a lower limit. Using this branching ratio one calculates the hindrance factors shown in Fig. 1(b).

Two gammas were observed at 68.4 ± 0.1 and 53.3 ± 0.1 keV following the alpha decay of 231 U as indicated in Fig. 2. Their energies fit quite well between the 5404 keV alpha and the 5471 and 5456 keV alphas as suggested in

FIG. 1. (a) The alpha spectra of 230 U (and daughters), 231 U, 232 U, and 233 U observed after a U separation following the bombardment of 232 Th with 45 MeV alphas. (b) Close-up of the alpha spectra of ²³¹U and ²³²U. Energies of the ²³¹U spectrum are given in keV together with the intensities and the hindrance factors. The alpha branching ratio of ²³¹U and the gamma decay following alpha emission are also indicated.

Fig. 1(b). Two other gammas, 64.3 and 67.0 keV, with weaker intensities were also observed, but we are unable to assign them in the level scheme of 227 Th. The L-x-ray intensities also coincident with the alphas are consistent with the assignment of the two gamma transitions as $M1$. (The close lying 57.8 keV $E2$ in ²²⁸Th was used for internal calibration in this case. See Table I.)

We use as a basis of our interpretation of the levels in 227 Th the suggestions of Leander and Chen [7] who have interpreted the much earlier experimental work of Novikova et al. [8] who studied the β ⁻ decay of ²²⁷Ac. In this scheme [7] the three observed levels are interpreted as a $K^{\pi} = 1/2^{+}$ band with a $1/2^{+}$ level as the ground state and a $5/2^+$ level at 9.3 keV and a $K^{\pi} = 3/2^+$ bandhead at 24.5 keV. We note that within experimental error the energy difference between the 9.3 and 24.5 keV states is the same as the experimental energy difference in the 5471 and 5456 keV alpha groups and the 68.4 and 53.3 keV gamma transitions. This in turn suggests that the alpha decay of 231 U does not populate the 227 Th ground state, but does populate excited states at 9.3, 24.4, and 77.7 keV. The 77.7 keV state is then depopulated by two gamma rays of 68.4 and 53.3 keV to the 9.3 and 24.4 keV states, respectively. This level scheme is shown as an inset in Fig. 2 where the energy levels have been slightly adjusted to fit the recent data.

The observation of $M1$ transitions from the 77.7 keV state feeding two positive parity states with tentative spins $5/2^+$ and $3/2^+$ suggests spins $5/2$ or $3/2$ with positive parity. In view of the approximate rotational spacing we very tentatively suggest that the 77.7 keV state may be the $5/2^+$ rotational state built on the $K = 3/2$ bandhead at 24.4 keV.

It is interesting to note that none of the alpha decay hindrance factors (HF's) is low enough to suggest that the configurations populated in 227 Th have the same configuration as the 231 U ground state. The ground state of

FIG. 2. The gamma spectrum obtained in coincidence with the alphas in the energy range 5260—5500 keV. The level scheme deduced for 227 Th from these experiments is indicated in the inset. The transitions observed in bold print are those observed in these experiments. The other transitions were observed in the β^- decay of ²²⁷Ac [8].

TABLE I. Relative L -x-ray and gamma ray intensities.

$E\gamma$ (keV)	Multipolarity	$(L-x-ray / \gamma-ray)$ expt.	$(L-x-ray / \gamma-ray)$ calc.
57.8		$38 + 4$	$E2 = 38$
$53.3 + 68.4$	$M1 (+E2)$	$7+2$	$E1=0.2$, $M1=4$, $E2=36$

²³¹U would be expected to have $J^{\pi} = 3/2^{+}$ or $5/2^{+}$ and the same K quantum number since in both the isotones 227 Ra and 229 Th these states are within 2 keV of the ground state, being reversed in these two cases [9,10]. The state with the lowest HF (36) is the 77.7 keV state with tentative $J^{\pi} = 5/2^{+}$. It is possible that this state Coriolis couples with the state with the same configuration as the 231 U ground state to achieve this relatively low HF. It then in turn could Coriolis couple with the $5/2^+$ state at 9.3 keV, allowing a reasonable alpha population and HF = 62. However, the $1/2^+$ ground state being unable to Coriolis couple with any state related to the 231 U ground state might, as observed, not be populated.

In summary, we have observed the fine structure of the alpha decay of 231 U for the first time and deduced the low-lying levels in 227 Th. Tentative spins and parities have been assigned that are consistent with a ground state of $1/2^+$ for ²²⁷Th.

One of us (R.K.S.) wishes to thank the National Science Foundation for support under Contract No. PHY92- 07336 with Florida State University, and for the hospitality of the CSNSM and the IPN at Université de Paris-Sud, Campus Orsay. All of us appreciate the help received from the cyclotron team in Orleans.

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