Isomerism in ^{147}Tb†

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The previous assignment of a 24-min activity to ^{147}Tb was found to be in error. Two ^{147}Tb isomers with half-lives of 1.61±0.17 h and 2.5±0.1 min were found and their γ-ray spectra determined. The 1.6-h isomer appears to be the ground state, with spin-parity $\frac{5}{2}^-$; the 2.5-min ^{147m}Tb is the high-spin state ($\geq 11/2$). The half-life and prominent γ rays of ^{147}Gd were redetermined and two new γ rays associated with the decay of 4.3-min ^{149m}Tb are reported.

INTRODUCTION

A NUCLIDE of 24-min half-life decaying with the emission of 305-keV γ rays was observed by Toth and Rasmussen[1] in the bombardment of ^{141}Pr with 100-MeV ^{12}C ions and assigned by them to ^{147}Tb. The assignment was made on the basis of (a) the growth of ^{147}Gd γ rays in the same sample with a growth rate consistent with a parent half-life of 20–30 min and (b) the absence of the 24-min activity in Pr samples bombarded with 65- and 75-MeV ^{12}C ions. No other reports on ^{147}Tb have appeared in the literature.[2]

In the course of an investigation of rare-earth nuclides produced in the interaction of uranium targets with high-energy protons,[3,4] chemically and isotopically separated samples of ^{147}Tb were examined for their radioactive properties. Since the isolation procedures took several hours, the 24-min activity was not expected to be found in appreciable intensity, and no indication of it was seen. However, the samples were found to emit x rays and γ rays which decayed with a half-life of about 1.6 h.

1.6-h ^{147}Tb

For the further investigation of the new ^{147}Tb activity, Ta targets were irradiated for 30–60 min in the 28-GeV circulating proton beam of the Alternating Gradient Synchrotron at Brookhaven. After the irradiations, the targets were dissolved in the presence of rare-earth carriers, and standard chemical separations were performed to isolate the rare-earth elements as a group from the Ta and from other radioactivities produced in the irradiations. In one experiment, the gross rare-earth fraction in the form of oxide was introduced into the ion source of the Brookhaven electromagnetic isotope separator and was mass-separated. The material collected at the mass-147 position was subsequently subjected to cation-exchange separation to obtain ^{147}Tb and ^{147}Gd fractions. In another experiment, chemical isolation of Tb and Gd fractions by ion exchange was accomplished first and these fractions were separately subjected to mass separation. Further details of the chemical and mass separations were recently given[5] in connection with a study of ^{149}Tb.

The radiations emitted by the ^{147}Tb and ^{147}Gd samples were investigated with NaI(Tl) and Ge(Li) detectors. A 2-mm-thick NaI(Tl) detector and single-channel analyzer were used to follow the x-ray decays; a calibrated 15-cm^3 Ge(Li) detector and multichannel analyzer served for the γ-ray measurements. The energies and intensities of the γ rays associated with the decay of the ^{147}Tb are shown in Table I. In addition to these γ rays which decayed with half-lives of 1.6–1.8 h (weighted average 1.61±0.17 h) the ^{147}Tb samples showed the expected growth of the principal γ rays associated[6] with the decay of the ^{147}Gd daughter and eventually those of the 24-day ^{147}Eu granddaughter.[6,7] The absolute abundances of the ^{147}Tb γ rays shown in Table I were calculated from the measured intensities and the known abundances (see below) of the ^{147}Gd γ rays. From the observed intensity of annihilation radiation it was found that about 5% of the ^{147}Tb decays proceed by β^+ emission. The observed x-ray intensity is consistent with this finding, since it indicates that most ($\gtrsim$90%) of the ^{147}Tb decays proceed by electron capture.

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[1] K. S. Toth and J. O. Rasmussen, J. Inorg. Nucl. Chem. 12, 236 (1960).

[2] A portion of the present work is reported in Proceedings of the 155th National Meeting of the American Chemical Society, San Francisco, 1968; see Y. Y. Chu, E. M. Franz, G. Friedlander, and E. Hechtl, Abstract No. 0–71.

[3] Y. Y. Chu, E. M. Franz, G. Friedlander, E. Hechtl, and P. Karol, in Proceedings of the 154th National Meeting of the American Chemical Society, Chicago, 1967, Abstract No. R-50.

[4] Y. Y. Chu, E. M. Franz, G. Friedlander, and P. J. Karol, Ref. 2, Abstract No. 0–177.

[5] Y. Y. Chu, E. M. Franz, and G. Friedlander, Phys. Rev. 175, 1523 (1968).

[6] M. P. Avotina, E. P. Grigor'ev, A. V. Zolotavin, V. O. Sergeev, V. E. Ter-Nersesyauts, J. Vrzal, N. A. Lebedev, J. Liptak, and Ya. Urbanets, Izv. Akad. Nauk SSSR, Ser. Fiz. 30, 1292 (1966) [English. transl.: Bull. Acad. Sci. USSR, Phys. Ser. 30, 1350 (1967)].

[7] C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (John Wiley & Sons Inc., New York, 1967), 6th ed.

TABLE I. γ rays of 1.6-h ^{147}Tb.

Energy (keV)	Photons per disintegration
118.5±0.3	0.051±0.010
139.4±0.5	0.24±0.03
694.4±0.5	0.32±0.04

Note added in proof. According to J. Alstad (private communication), he independently discovered 1.6-h ^{147}Tb at CERN in uranium targets bombarded with 18-GeV protons. Chemical and mass separations were used as in the present paper. Alstad observed 140- and 690-keV γ rays.

SEARCH FOR 24-MIN ^{147}Tb

Although, as mentioned above, the time required for both mass and chemical isolation of ^{147}Tb from the high-energy proton bombardments described was such that a 24-min activity could not have been directly observed, indirect evidence for the existence of a short-lived ^{147}Tb was obtained in these experiments as follows.

In a high-energy proton irradiation of a high-Z target a given spallation or fission product can be formed either directly or through α or β decay of radioactive precursors. Thus, ^{147}Gd can arise through positron and electron-capture decay of ^{147}Tb, and through α decay of ^{151}Dy, as well as by independent formation. The total production cross section for ^{147}Gd found in the Ta and U bombardments is in accord with the general yield patterns observed in this mass region. However, if one assumes that subtraction from this total of the relatively small contributions of 1.6-h ^{147}Tb decay and ^{151}Dy α decay yields the independent formation cross section of ^{147}Gd, then the resulting values are unreasonably large. Conversely, the observed yields of 1.6-h ^{147}Tb are about four or five times smaller than expected from systematics. These observations would be consistent with the existence of another isomer of ^{147}Tb sufficiently short-lived (say, $\lesssim$30 min) to have largely decayed into ^{147}Gd by the time the Gd-Tb separations were performed.

Further attempts were therefore made to observe the previously reported 24-min ^{147}Tb by shortening the procedures to include either mass or chemical identification, but not both. No activity with the reported characteristics was found either in the mass-147 fraction isolated from a Tb_4O_7 target irradiated with 30-GeV protons or in a chemically pure Tb fraction obtained from a similarly bombarded Ta target. In fact, no activity attributable to ^{147}Tb (other than the 1.6-h activity) was found in these experiments, but an activity of less than ~10-min half-life would have escaped detection. On the assumption that the 24-min species reported by Toth and Rasmussen[1] might have been a Gd rather than a Tb isotope, the Gd fraction from a Ta bombardment was investigated also. The well-known[7] 25-min ^{145}Gd which emits a 330-keV γ ray,[8] was observed.

Although it seemed unlikely that high-energy bombardments should fail to produce the 24-min ^{147}Tb if it had been made by heavy-ion reaction, a final attempt at observing it was made by duplicating as nearly as possible the experiment of Toth and Rasmussen. Targets of praseodymium oxide were bombarded with 100-MeV ^{12}C ions in the Heavy Ion Linear Accelerator at Yale University. γ spectra taken with a 25-cm^3 Ge(Li) detector showed no sign of γ radiation in the vicinity of 305 keV, nor of any γ rays decaying with a half-life of 24 min. Thus, the assignment of a 24-min activity to ^{147}Tb must definitely be considered to be in error.

2.5-min ^{147}Tb and 4.3-min ^{149m}Tb

The γ spectra of ^{12}C-irradiated Pr targets did show some rapidly decaying γ rays which, upon more careful study, were found to fall into two groups, with half-lives of 2.5±0.1 and 4.3±0.1 min. When the energy of the ^{12}C beam was varied, the relative intensities of these two groups changed: With 75-MeV ^{12}C ions, the ratio of the 4.3-min to the 2.5-min γ rays increased by an order of magnitude; with 115-MeV ^{12}C ions this ratio decreased by a similar factor relative to the 100-MeV bombardment. These results are entirely consistent with the two groups of γ rays being associated with the decays of 4.3-min ^{149m}Tb [9] (produced by ^{12}C, $4n$ reaction) and of a new 2.5-min isomer of ^{147}Tb (produced by ^{12}C, $6n$ reaction). The energies and relative intensities of the prominent γ rays of these two species are shown in Tables II and III. No γ rays have previously been reported for ^{149m}Tb.

DISCUSSION OF ^{147}Tb ISOMERISM

The 2.5-min activity observed in the ^{12}C bombardments of ^{141}Pr was too short-lived to allow chemical identification. Its assignment to ^{147}Tb thus rests on indirect evidence. As already mentioned, the dependence of its yield on ^{12}C energy indicates this assignment. Furthermore, in the ^{141}Pr+^{12}C bombardments, as in the high-energy proton experiments described earlier,

TABLE II. γ rays of 2.5-min ^{147}Tb.

Energy (keV)	Relative intensity (arbitrary units)
395.9±0.5	47
787.3±0.5	95
886.2±0.7	100

[8] K. A. Keller and H. Münzel, Radiochim. Acta 9, 176 (1968).
[9] R. D. Macfarlane, Phys. Rev. 126, 274 (1962); R. D. Macfarlane and D. W. Seegmiller, Nucl. Phys. 53, 449 (1964).

the observed large yield of ^{147}Gd demands the existence of a short-lived ^{147}Tb precursor (in addition to the 1.6-h ^{147}Tb), and the 2.5-min activity is the obvious candidate. The observations in the ^{12}C experiments were as follows.

A Pr-oxide sample bombarded with 100-MeV ^{12}C ions was dissolved and subjected to cation-exchange separation for the isolation of Tb and Gd fractions within about 1 h after irradiation. In the Tb fraction the decay of the γ rays of 1.6-h ^{147}Tb and the growth and subsequent decay of ^{147}Gd γ rays were followed. The amounts of ^{147}Gd in the Tb and Gd fractions were in the ratio 1:50. On the other hand, the ratio of cross sections of the ^{12}C, $6n$ and ^{12}C, $p5n$ reactions (leading to ^{147}Tb and ^{147}Gd, respectively) were expected to be certainly greater than unity. For the analogous pair of reactions ^{140}Ce(^{12}C, $6n$)^{146}Gd and ^{140}Ce(^{12}C, $p5n$)^{146}Eu, the cross-section ratio was found to be 3:1 in a separate experiment. The only logical explanation is that a short-lived isomer of ^{147}Tb which decays directly to ^{147}Gd was produced in much higher yield than the 1.6-h species. The 2.5-min activity is the only one observed that can be assigned to ^{147}Tb (in principle, an unobserved ^{147}Tb of $<$1-min half-life could be postulated instead). If the 2.5-min species is indeed the "missing" ^{147}Tb isomer, its yield in the ^{12}C bombardment of ^{141}Pr was about 40 times that of the 1.6-h isomer, as deduced from the cumulative ^{147}Gd yield corrected for saturation of the short-lived parent. The observed intensities of the γ rays of the 2.5-min species were also consistent with this interpretation, if little or no transition to the ^{147}Gd ground state is assumed.

The large yield ratios in favor of the 2.5-min isomer in both the high-energy proton spallation of Ta ($\sim$4–5) and in the (^{12}C, $6n$) reaction on ^{141}Pr ($\sim$40) indicate that the 2.5-min species is the high-spin isomer. There is no indication of a significant amount of isomeric transition. Rather, the two isomeric states appear to decay independently to ^{147}Gd; furthermore, the two decay schemes do not have any prominent γ rays in common (cf. Tables I and II).

At present it is not possible to construct decay schemes for the two ^{147}Tb isomers. From the γ-ray abundances of the 1.6-h isomer, one can conclude that it decays in 40–70% of its disintegrations directly to the ground state of ^{147}Gd. This result, taken together with the observed capture-to-positron ratio of about 20 leads to an estimate of 2.2–2.3 MeV for the decay energy, which is consistent with an allowed ground-state transition ($\log f_0 t \approx 5.5$). As mentioned earlier, the γ-ray intensities of the 2.5-min isomer, together with the cumulative yield of ^{147}Gd, indicate little or no ground-state transition from this isomer. Its decay evidently proceeds through fairly high-lying states and, if it is to have reasonable ft values, it must therefore lie considerably above the 1.6-h level.

Since the ground state of ^{147}Gd is almost certainly $\frac{7}{2}^-$ (in analogy with the other 83-neutron nuclei ^{145}Sm, ^{143}Nd, ^{141}Ce, ^{139}Ba), the 1.6-h ^{147}Tb must be a $\frac{5}{2}^-$, $\frac{7}{2}^-$, or $\frac{9}{2}^-$ state. The only one of these spin-parity assignments which, according to Nilsson diagrams,[7] is a reasonable choice for the ^{147}Tb ground state is $\frac{5}{2}^-$. The fact that, in the sequence of odd-A Tb nuclei ^{161}Tb to ^{155}Tb, a $\frac{5}{2}^-$ state decreases in energy from 482 to 227 keV relative to the $\frac{3}{2}^+$ ground state, lends strength to this assignment for the 1.6-h ^{147}Tb. Further details of the decay schemes of the two isomers are needed to assign spin and parity to the 2.5-min state. At present, all that can be inferred is that its spin is almost certainly $\geq \frac{11}{2}$.

TABLE III. γ rays of 4.3-min ^{149m}Tb.

Energy (keV)	Relative intensity (arbitrary units)
164.2±1.0	57
798.5±0.5	100

TABLE IV. γ rays of 38-h ^{147}Gd.

Energy (keV) This work	Energy (keV) Avotina *et al.*	Photons per disintegration This work	Photons per disintegration Avotina *et al.*
217.6±0.3	217.3	0.018±0.002	...
229.9±0.2	229.2	0.573±0.051	0.570±0.080
241.0±0.2	240.6	0.011±0.001	0.012±0.001
310.5±0.2	310.0	0.029±0.003	0.037±0.002
319.0±0.2	318.65	0.015±0.001	0.020±0.002
370.5±0.2	370.0	0.132±0.013	0.159±0.016
396.5±0.1	396.0	0.262±0.033	0.318[a]
485.4±0.1	484.9	0.026±0.002	0.031±0.002
559.4±0.1	559.2	0.052±0.005	0.065±0.007
610.7±0.1	610.8	0.017±0.007	0.017±0.002
619.3±0.1	618.6	0.032±0.003	0.036±0.005
625.4±0.1	625.2	0.061±0.014	0.047[a]
765.5±0.1	765.7	0.095±0.009	0.104±0.005
776.8±0.1	776.6	0.080±0.007	0.092±0.003
861.3±0.1	861.5	0.017±0.003	0.018±0.002
893.0±1.0	893.8	0.067±0.006	0.075±0.002
928.1±0.1	928.4	0.162±0.014	0.200±0.008

[a] No error estimates are given in Ref. 6 for the intensities of these two γ rays.

^{147}Gd

The radiations emitted by the mass-separated ^{147}Gd samples from proton-bombarded Ta and by the Gd sample from Pr+^{12}C were examined with the NaI(Tl) and Ge(Li) detectors. From the decay measurements on the x rays and the most prominent γ rays, a half-life of 38.06±0.12 h was deduced by means of the CLSQ least-squares program.[10] A wide range of half-life values has been reported for this nuclide.[7] Our result is in good agreement with the value of 38.5±0.5 h

[10] J. B. Cumming, National Academy of Sciences, National Research Council, Nuclear Science Series Report No. NAS, NS-3107, 1962, p. 25 (unpublished).

recently reported by Avotina *et al.*[6] and with Grover's value of 37 h.[11] The only previously reported half-life determination with a mass-separated ^{147}Gd sample[12] yielded a value of 21.7±2.5 h, grossly at variance with our data.

In Table IV the energies of the most prominent γ rays associated with the decay of 38-h ^{147}Gd are listed, together with their abundances. The absolute abundances were calculated from the intensities of the 122- and 198-keV γ rays of ^{147}Eu that grew into the ^{147}Gd samples. For the number of 122- and 198-keV quanta per ^{147}Eu disintegration, the literature values[7] 0.20 and 0.24, respectively, were used. As can be seen in Table IV, the present values for the ^{147}Gd γ-ray energies and abundances are in fairly good agreement with those given by Avotina *et al.*[6]; however, these authors reported many additional γ rays which are either too low in intensity to have been seen in the present study or outside the energy range investigated.

[11] J. R. Grover, Phys. Rev. **126**, 1540 (1962).
[12] A. M. Friedman, J. Milsted, and O. Skilbreid, Phys. Rev. **129**, 1752 (1963).

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Transition Matrix Elements in the Even-Even Osmium Isotopes: A Comparison with Pairing-Plus-Quadrupole Model Calculations*

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The energy-level structures and transition matrix elements between collective excitations in the even-even osmium isotopes ($A=186$–192) have been studied using Coulomb excitation with oxygen projectiles having incident energies between 42 and 80 MeV. The deexcitation γ radiations have been observed with NaI(Tl) and Ge(Li) detectors, singly, in coincidence with inelastically backscattered O^{16} ions, and in coincidence with γ rays from the $2^+\rightarrow 0^+$ and $4^+\rightarrow 2^+$ transitions in each isotope. Angular-correlation studies have been performed on all observed transitions following Coulomb excitation by backscattered ions. In the four isotopes studied, all levels through the 6^+ state of the ground-state band as well as the second excited $2^{+\prime}$ and $4^{+\prime}$ states have been populated, and the transition moments associated with the excitation of these states have been measured. In Os^{192}, previously unobserved radiations have been assigned to the deexcitation of a 6^+ state at 1088 keV and a $4^{+\prime}$ state at 907 keV. Except for the observation of the 0^+ state at 1086 keV in Os^{188}, a search for low-lying 0^+ states in the other osmium isotopes did not reveal any definite candidates. Electric-quadrupole transition matrix elements have been deduced from the data with a model-independent analysis using the Winther–de Boer computer code for multiple Coulomb excitation. The latter quantities, together with measured branching ratios and $B(M1)$ values, have been compared with several macroscopic and microscopic nuclear models, with particular attention to the pairing-plus-quadrupole model of Kumar and Baranger. The latter has proved to be the most complete and successful of the models tested in predicting both absolute $B(E2)$ values, and the variation of these with neutron number in the osmium isotopes. However, the model in its original form makes the transition from deformed to spherical shape more rapidly, with increasing mass, than appears to be the case experimentally, thereby suggesting the need for small variations in the relative strength of the pairing and quadrupole nucleon-nucleon residual forces assumed as input to the microscopic calculations. A description of the osmium isotopes, as typified by shallow potential minima for moderately prolate deformations (in the cases of $Os^{190,192}$ asymmetric equilibrium configurations) and by pronounced softness to both β and γ vibrations, emerges from the comparisons made.

I. INTRODUCTION

In the evaluation of the significance of various theories of nuclear collective motion, the study of the transitional nuclei at the upper and lower mass limits of the rare earths has been very useful. In particular, a description, on both the microscopic and phenomonological levels, of the evolution from the harmoniclike structure of the "vibrator" nuclei to the rotational-like structure of nearby nuclei with large static quadrupole deformations has been sought in recent investigations. It is evident from the existing data that as one moves away from those few nuclei for which the rotational and vibrational limits are good

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