Decay of Terbium-157†

R. A. NAUMANN AND P. K. HOPKE
Frick Chemical Laboratory, Palmer Physical Laboratory
and
Princeton-Pennsylvania Accelerator, Princeton University, Princeton, New Jersey
(Received 27 February 1967)

Samples of Tb^{157} have been prepared by irradiating enriched Dy^{156} in the Materials Testing Reactor. After irradiation, the Tb^{157} samples have been prepared by ion exchange separation and the use of the Bohr Institute's electromagnetic isotope separator. The L_I-to-K electron-capture ratio has been determined as 2.65 ± 0.20 using a beryllium-windowed sodium iodide spectrometer. This value is comparable to previous determinations. It implies a Q value for electron capture to the ground state of Gd^{157} of 66 ± 6 keV which also permits electron capture to the first excited state at 54 keV. A search has been made for photons of this energy using a germanium semiconductor detector having an energy resolution of 0.75 keV. Photons having 54.35 ± 0.05 keV have been observed in Tb^{157} sources. Using the measured photon intensity and the intensity of the K x rays, a branching ratio of $(0.34\pm0.2)\%$ is calculated for electron capture to the first excited state.

THE first report of long-lived Tb^{157} was based on a mass-spectrometric examination of terbium isotopes formed after irradiation of dysprosium enriched in mass 156 with the high neutron flux available in the Materials Testing Reactor at Arco, Idaho. In the mass spectrum of the chemically isolated terbium fraction, clear evidence for long-lived Tb^{157} and Tb^{158} was obtained.[1] After the decay of the 75-day Tb^{160} simultaneously produced in this irradiation, γ radiation accompanying the electron-capture decay of long-lived Tb^{158} has been reported.[2,3]

In order to distinguish possible γ radiations of Tb^{157} from those of Tb^{158} in our sample, it has been necessary to carry out an electromagnetic isotope separation of these two isotopes. This separation has been carried out using the electromagnetic-isotope-separator facility at the Bohr Institute for Nuclear Physics. The K x rays arising from the electron capture of Tb^{157} have been observed in this sample.[2]

In the intervening period, two studies have been made of the K and L x-ray spectra accompanying the decay of Tb^{157}. Bhat and Pool[4] have reported an L_I-to-K capture ratio of 2.64 and deduced a decay energy of 60 keV, while Fujiwara *et al.*[5,6] have reported an L_I-to-K capture ratio of 2.18 and deduced a decay energy of 60 keV. No evidence for population of excited states in Gd^{157} by electron capture of Tb^{157} has previously been reported.[2,4,5,6]

The ground-state spin of Gd^{157} has been measured as $\frac{3}{2}$. Two low-lying excited states in Gd^{157} are known from the β decay of Eu^{157}: a first excited state at 54.5 keV interpreted as the spin-$\frac{5}{2}$ member of a rotational sequence based on the ground state, and a second excited state at 64.0 keV.[7] The two rotational states are assigned the asymptotic quantum numbers [521] according to the unified nuclear model.[8] Negative parity is thus inferred. Similarly, the $\frac{5}{2}^+$ second excited state at 64.0 keV has been assigned the asymptotic quantum numbers [642].

A rotational level sequence based on the ground state of Tb^{157} has been observed in the electron-capture decay of Dy^{157} which is consistent with a ground-state spin of $\frac{3}{2}$ for this state.[9] The asymptotic quantum numbers [411] and positive parity have been assigned to this state. If the Tb^{157} decay energy is between 60 and 70 keV, the above assignments suggest that branch decay to one or both of the low-lying Gd^{157} excited states is possible. We have therefore undertaken a redetermination of the L_I-to-K electron-capture ratio

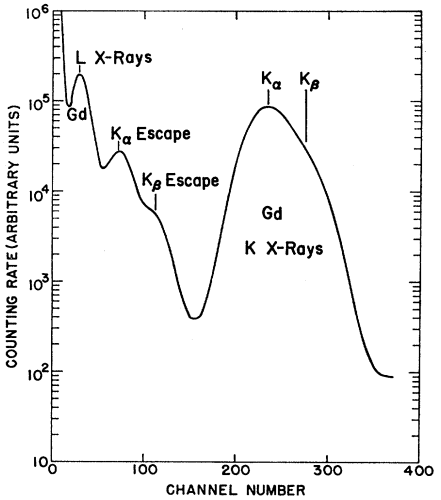

FIG. 1. The L and K x-ray spectrum of Tb^{157} observed with a beryllium-windowed NaI(Tl) detector.

† Work supported by the U. S. Atomic Energy Commission.
[1] R. A. Naumann, M. C. Michael, and J. L. Power, J. Inorg. Nucl. Chem. **16**, 163 (1960).
[2] R. A. Naumann, O. B. Nielsen, and O. Skilbreid, Bull. Am. Phys. Soc. **7**, 34 (1962).
[3] H. R. Lewis, Jr., R. A. Naumann, and J. L. Power, Bull. Am. Phys. Soc. **6**, 238 (1961).
[4] M. R. Bhat and M. L. Pool, Phys. Rev. **127**, 1704 (1962).
[5] I. Fujiwara *et al.*, J. Phys. Soc. Japan **18**, 315 (1963).
[6] I. Fujiwara *et al.*, Nucl. Phys. **50**, 346 (1964).
[7] B. Harmatz, T. H. Hanley, and J. W. Mihelich, Phys. Rev. **128**, 1186 (1962).
[8] B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **1**, No. 8 (1959).
[9] J. W. Mihelich, B. Harmatz, and T. H. Handley, Phys. Rev. **108**, 989 (1957).

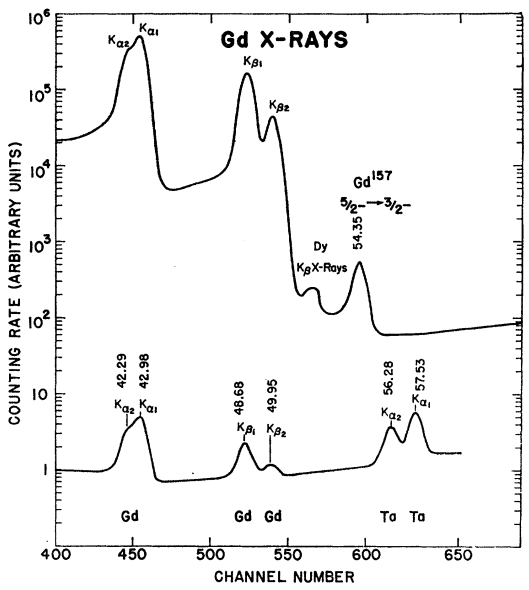

FIG. 2. Upper curve: K x-ray and γ spectrum of Tb^{157} taken with high-resolution Ge(Li) detector. Lower curve: calibration spectrum using fluorescent K x rays.

of Tb^{157} and searched for γ radiations accompanying this decay.

The isotope-separated source of Tb^{157} was deposited on 0.1-mil aluminum foil. The x-ray spectrum has been recorded using a cylindrical, cleaved sodium iodide crystal 38.1 mm in diameter by 6 mm in thickness with a beryllium window 0.127 mm thick. The spectrum recorded using a 400-channel analyzer is shown in Fig. 1. No correction is necessary for the attenuation by the window of the 7- and 45-keV L and K x rays. By integrating the areas under the L and K photopeaks and including the areas under the K_α and K_β escape peaks, an experimental L_I-to-K photon ratio, $R=0.741\pm0.030$, was obtained.

The ratio of L_I-to-K electron capture, $\lambda_{L_\mathrm{I}}/\lambda_K$, was computed as 2.65 ± 0.20 using the expression

$$\frac{\lambda_{L_\mathrm{I}}}{\lambda_K}=\left(R-\frac{n_{KL}\omega_{KL}}{\omega_K}\right)\frac{\omega_K}{\omega_L},$$

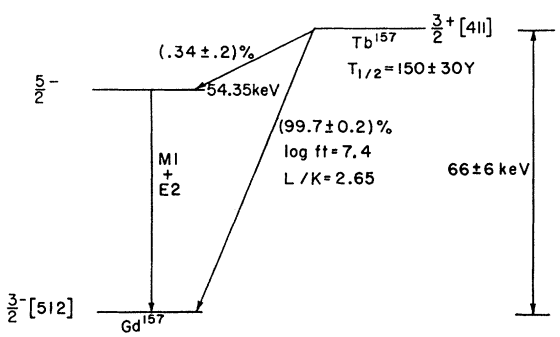

FIG. 3. Proposed decay scheme for Tb^{157}.

with the value for the L flourescence yield $\omega_L=0.198$,[10] the K fluorescence yield $\omega_K=0.925\pm0.005$,[10] and $n_{KL}=0.85$,[10] where n_{KL} is the number of L vacancies arising from an initial K vacancy, and ω_{KL} is the fluorescent yield of L x rays following a primary K vacancy and is 0.18.[10]

The first-forbidden electron-capture Q value for Tb^{157} has been calculated to be 66 ± 6 keV using the allowed electron-capture result

$$\frac{\lambda_{L_\mathrm{I}}}{\lambda_K}=\left(\frac{(Q-W_{L_\mathrm{I}})\,|g_{L_\mathrm{I}}|}{(Q-W_K)\,|g_K|}\right)^2,$$

where W_{L_I} and W_K are the L_I- and K-shell electron binding energies in gadolinium. g_{L_I} and g_K are the large components of the Dirac wave functions evaluated at the nuclear radius for these electrons. W_{L_I} and W_K are taken to be 8.4 and 50.4 keV, respectively, from the tables of Hill, Church, and Mihelich,[11] and g_{L_I} is equal to 0.4357, while g_K is 1.193.[12] This procedure appears justified since the uncertainties in the terms ω_L and n_{K_L} used previously appear larger than considerations involving L_II and L_III capture.

A search for γ radiations from Tb^{157} is complicated by the proximity of the anticipated 54.5-keV γ transition to the intense $K_{\beta1}$ and $K_{\beta2}$ x-ray groups lying approximately at 48.7 and 50.0 keV. The development of ultrahigh-resolution germanium detectors by the Chemistry Division of the Lawrence Radiation Laboratory, Berkeley, makes such a measurement feasible. A detector having dimensions $0.8\times0.8\times0.8$ cm with a resolution of 0.75 keV has been used to search for low-energy radiations accompanying the decay of the separated Tb^{157} sample. Figure 2 shows a portion of a spectrum accumulated over 46 h. The figure includes a calibration spectrum showing gadolinium and tantalum x rays excited by a Co^{57} source iluminating stable compounds containing these elements. Lead and cerium K x rays were also included in the calibration spectrum but are not shown in the figure. The photopeak due to 54.35-keV γ rays is clearly resolved. The small peak of lower energy is attributed to dysprosium K x rays from a small amount of Tb^{158} present in the mass-separated sample. The relative intensity r of the 54.35-keV photons to the total intensity of gadolinium K x rays is 8.42×10^{-4}. The total conversion coefficient, α_T of the 54.35-keV transition was calculated to be $\alpha_K+\alpha_L+\alpha_M+\alpha_N=15.1$, using an $M1/E2$ radiative ratio of 29[8] and the theoretical K and L conversion coefficients of Rose.[13] The empirical procedure of Chu and Perlman[14]

[10] R. W. Fink, R. C. Jopson, H. Mark, and C. D. Swift, Rev. Mod. Phys. 38, 513 (1966).
[11] R. D. Hill, E. L. Church, and J. W. Mihelich, Rev. Sci. Instr. 23, 523 (1952).
[12] K capture: I. M. Band, L. N. Zyrianova, and T. Chen-Zhui, Bull. Acad. Sci. USSR, Phys. Ser. 20, 1269 (1956); L capture: I. M. Band, L. N. Zyrianova, and Y. P. Suslov, Bull. Acad. Sci. USSR, Phys. Ser. 22, 943 (1958).
[13] M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).
[14] Y. Y. Chu and M. Perlman, Phys. Rev. 135, B319 (1964).

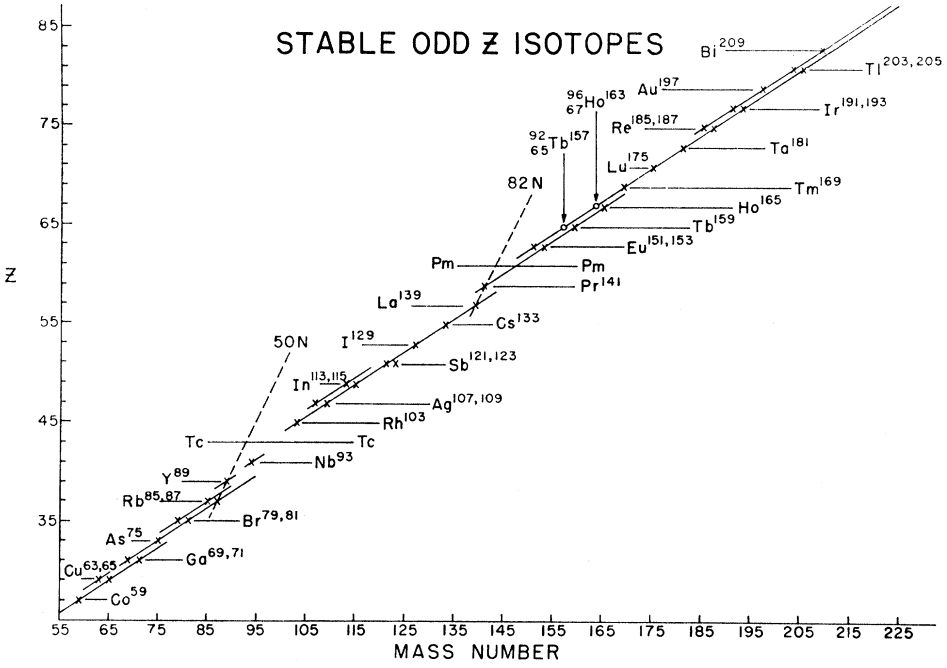

FIG. 4. The naturally occurring isotopes for elements having odd Z. Line segments join species differing by two protons and four neutrons. Dashed lines indicate neutron numbers 50 and 82.

was used to estimate α_M, and α_N was assumed to be 0.3 α_M. The ratio of the decay constants for electron capture to the excited and ground states, respectively, was calculated using the relation

$$\frac{\lambda^*}{\lambda}=\frac{r(1+\alpha_T)\omega_K}{(1+\lambda_L/\lambda_K)}=(3.4\pm0.2)\times10^{-3},$$

where ground-state capture from M and higher electron shells has been neglected. No evidence has been found for population of the second excited state in gadolinium at 64.0 keV. The decay scheme for Tb^{157} is shown in Fig. 3.

The observation of the 54.4-keV γ ray indicates that the Q value for Tb^{157} electron capture has at least this value. Some uncertainty remains as to whether L capture is necessarily responsible for population of the first excited state in Gd^{157}. To examine this question one may consider an estimate made on the assumption that L_{I} electron capture to the first excited state results in neutrinos of zero energy. In this case M capture proceeds at the fastest rate possible consistent with nonoccurrence of L capture. The energy of the neutrino emitted in L_{I} capture to the ground state is $E_{L_{\mathrm{I}}}=54.4$ keV, the difference in energy between the ground state and the first excited state. The energy of the neutrinos emitted in M_{I} capture populating the excited state is then $E_{M_{\mathrm{I}}}{}^*=6.5$ keV, the difference between the L_{I} and M_{I} binding energy. The corresponding ratio of the decay constants for excited-state M_{I} capture to ground-state L_{I} capture is

$$\frac{\lambda_{M_{\mathrm{I}}}{}^*}{\lambda_{L_{\mathrm{I}}}}=\left(\frac{E_{M^{\mathrm{I}}}{}^*|g_{M_{\mathrm{I}}}|}{E_{L_{\mathrm{I}}}|g_{L_{\mathrm{I}}}|}\right)^2=4.2\times10^{-3},$$

where the nonrelativistic hydrogenic estimate $|g_{M_{\mathrm{I}}}|^2/|g_{L_{\mathrm{I}}}|^2=8/27$ has been assumed for the M_{I} and L_{I} relative electron density. In addition, the same matrix element has been used for electron capture to these two members of the same rotational band. This estimate shows that the observed branching ratio could be accounted for by capture to the excited state from only the M_{I} and higher electron shells. A search for coincidences between 54.4-keV γ rays and L x rays, although difficult, would resolve this question.

A useful correlation between the neutron and proton numbers of stable isotopes was reported by Duckworth[15] in 1949 in his survey of possible undiscovered stable or near-stable isotopes. This correlation related the then known lightest stable isotopes of elements having an even Z. Linear stability curves defined by the simultaneous addition of two protons and four neutrons connect sequences of six or seven such members and thus serve as a representation of the light-β-stability limit for even-Z elements.

Similar curves may be constructed for the class of stable isotopes having odd proton number and even neutron number. Such a graph containing all known members is shown in Fig. 4. Stability lines with the two-proton, four-neutron slope again successively connect these stable isotopes. The perturbation due to the neutron closed shells at 50 and 82 is evident. Of particular interest to us has been the nonoccurrence of Tb^{157} and Ho^{163} in spite of their location on a stability segment commencing with Eu^{151} and ending with Tl^{205}. Accordingly, it is not surprising that both these species have very low decay energies.

[15] H. E. Duckworth, Phys. Rev. **75**, 1438 (1949).

We would like to express our thanks to Dr. Earl K. Hyde and the Lawrence Radiation Laboratory, Berkeley, for allowing us to use their ultrahigh-resolution germanium detector and to Richard C. Jared, who kindly took the spectra for us. We also wish to thank Jeffrey Dundon for his assistance in the analysis of these data.

We are grateful to Ottar Skilbreid of the Bohr Institute, Copenhagen, for his kind assistance in carrying out the isotopic separation.

PHYSICAL REVIEW VOLUME 160, NUMBER 4 20 AUGUST 1967

Resonance Neutron Capture in the Even-A Isotopes of Tungsten*

W. V. PRESTWICH AND R. E. COTÉ
Argonne National Laboratory, Argonne, Illinois
(Received 24 March 1967)

The γ-ray spectra following neutron capture in the 4.14- and 21.2-eV resonances of W^{182} and the 18.8-eV resonance of W^{186} have been observed with a 6-cc Ge(Li) detector. Transitions corresponding to the $W^{182}(n,\gamma)W^{183}$ reaction have also been observed in the 115- and 250-eV resonances. In a special experiment, using a NaI detector, the absolute width of the ground-state transition for the 4.14-eV in resonance W^{182} was determined relative to the ground-state transition for the 7.62-eV resonance in W^{183}.

I. INTRODUCTION

THE low-lying level structure of odd-A nuclei in the deformed region is generally well described by rotational spectra built upon intrinsic Nilsson orbits.[1] The capture of an s-wave neutron by an even-even target leads to compound states in the odd-neutron nucleus with spin $\frac{1}{2}$ and positive parity. Since for initial states at excitations near the neutron separation energy the radiative strength to low-lying levels is primarily $E1$, the capture process will selectively populate $\frac{1}{2}^-$ and $\frac{3}{2}^-$ final states. These can be described in the unified model as the two lowest-spin members of a rotational band built upon a $K=\frac{1}{2}^-$ Nilsson orbit, and the lowest spin (or quasi-ground-state) member of a $K=\frac{3}{2}^-$ band. Other states that are members of the $(\frac{1}{2}^-,\frac{3}{2}^-)$ subset of levels may have their dynamical origin in vibration or core-excitation modes, and these normally will occur at energies greater than 1 MeV.

The eigenstates of the odd-neutron isotopes of tungsten, W^{183}, W^{185} and W^{187} have been studied in detail by several workers[2-4] through analysis of (d,p) angular distributions and reduced strengths. In particular, they have identified the lowest-lying states of $\frac{1}{2}^-$ and $\frac{3}{2}^-$ members of the $K=\frac{1}{2}^-$,[510] orbital and the $K=\frac{3}{2}^-$,[512] orbital. Erskine concludes that the $J=\frac{3}{2}^-$ states are, in fact, mixtures of the shell-model states.

Recently, work on the capture γ-ray spectra for both tungsten and hafnium isotopes has been reported. Martin, Harvey, and Slaughter[5] have measured the thermal-capture spectra of W^{183}, W^{185}, and W^{187}; and the spectra associated with capture in the 4.14-eV resonance in W^{182} and the 18.8-eV resonance in W^{186} have been measured by Faler, Spencer, and Dixon.[6] The thermal-capture spectra of hafnium have been measured by Namenson *et al.*[7,8] The results obtained show systematic effects, which would not be expected for a statistical compound state. The first regularity is observed among the primary transitions to the [510] band; the ratio of the intensity of the transition to the $\frac{3}{2}^-$ state to that to the $\frac{1}{2}^-$ state has similar values in the thermal spectra of W^{183} and Hf^{179} and in the 4.14-eV spectrum of W^{183}. Second, for these same spectra, the radiative strength to the [512] band appears to be consistently less than that to the [510] band.

In the investigation reported upon here, the relative radiative widths for the above-mentioned final states have been determined for capture in higher-energy resonances of W^{182}. In addition, the absolute radiative width for the ground-state transition following capture in the W^{182} 4.14-eV resonance has been measured.

II. EXPERIMENTAL ARRANGEMENT AND RESULTS

A. The Ge(Li) Spectra

A 1-kg sample of natural tungsten sheet was placed in the pulsed beam of the Argonne fast chopper[9] at a flight path of 6.43 m. Pulse-height spectra corresponding

* Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **29**, No. 16 (1955).
[2] A. Iosaya, Phys. Rev. **130**, 234 (1963).
[3] J. R. Erskine, Phys. Rev. **138**, B66 (1965).
[4] R. H. Siemssen and J. R. Erskine, Phys. Rev. Letters (to be published).
[5] M. J. Martin, J. A. Harvey, and G. G. Slaughter, Bull. Am. Phys. Soc. **11**, 336 (1966).
[6] K. T. Faler, R. R. Spencer, and D. R. Dixon, Bull. Am. Phys. Soc. **11**, 336 (1966).
[7] A. Namenson, H. E. Jackson, and R. K. Smither, Phys. Rev. **146**, 844 (1966).
[8] A. I. Namenson and H. H. Bolotin, Phys. Rev. **158**, 1206 (1967).
[9] L. M. Bollinger, R. E. Coté, and G. E. Thomas, in *Proceedings of the Second International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 14, p. 239.