THE Q-VALUE OF ^{157}Tb

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ABSTRACT

Using mass-separated and chemically purified samples, the Q-value of ^{157}Tb has been remeasured as 62.9 ± 0.7 keV by determining electron capture ratios. The transition probability to the excited state was found to be $(1.1 \pm 0.1) \times 10^{-3}$, with a transition energy of 8.4 ± 0.7 keV for this branch. From the proximity to the 2s binding energy, experiments on the internal bremsstrahlung electron capture seem feasible. The half-life of ^{157}Tb was remeasured as 99 ± 10 a.

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RADIOACTIVITY ^{157}Tb [from 600 MeV p-induced spallation of Ta, mass and chemical separation]; measured X-rays, Auger electrons and internal bremsstrahlung; deduced $T_{1/2}$, EC-branching ratio to excited state, EC subshell ratios, Q-value. Natural target, Si(Li) and high-purity Ge detectors, gas proportional detectors.

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1. INTRODUCTION

The recent success of the internal bremsstrahlung electron capture (IBEC) theory [1,2] and its prospects for yielding data on the rest mass of the electron-neutrino have stimulated the interest in nuclei that undergo electron capture with the 'right' Q-value. Ideally, the Q-value should equal the binding energy of an atomic s-state of the daughter atom. For instance, species such as ^{163}Ho [3] and ^{193}Pt [2] belong to this family of nuclei. Another of its members is possibly the long-lived isotope ^{157}Tb [1].

The decay of ^{157}Tb has been characterized as proceeding by electron capture to predominantly the ground state of the ^{157}Gd daughter, with Q-values of 66 ± 6 keV [4] and ~ 60 keV [5,6] which are based on measured L-to-K capture ratios. Naumann and Hopke [4] first reported a single weak EC branch to an excited 54.5 keV level of ^{157}Gd . In fact, it is this transition that is of particular interest here, as the difference between the 2s binding energy and the respective transition energy is only 3 ± 6 keV [4].

However, in order to judge the feasibility of any future experiment related to the neutrino mass and using ^{157}Tb , it is indispensable to gain a more precise knowledge of the involved Q-value. Therefore we initiated experiments on the electron capture decay of ^{157}Tb . In the present paper we report on determinations of the Q-value, based on different, partly novel, experimental techniques.

Section 2 outlines the preparation of the source, whereas in sections 3 to 5 we describe the experiments on the Q-value of ^{157}Tb . The first approach (see section 3) is based on the relative transition probability to the excited level. In section 4 the work on electron capture ratios is discussed and, finally, in section 5 some bremsstrahlung data of the ground-state transition are outlined.

2. SOURCE PREPARATION AND HALF-LIFE

For the source preparation, the ISOLDE facility on-line to the CERN Synchrocyclotron was employed. In a hot tantalum target, radioactive atoms were formed by 600 MeV proton-induced spallation and subsequently ionized. Ions of the mass chain 157 were collected in a Faraday cup, thereby determining the total number

of atoms N_0 in the source. After some months of waiting (to allow for decay of the short-lived isobars) the sample was further purified by a twofold ion exchange chromatography [3]. A fraction of this material was finally vacuum sublimated onto a 45 μm thick aluminium backing.

The source thus produced has an area of 1 cm^2 and contains 4×10^{12} atoms of ^{157}Tb . The contamination level of ^{158}Tb atoms is as low as 20 parts per million.

By comparing the Gd K-X-ray emission rate $N_K [\text{s}^{-1}]$ with the known number of ^{157}Tb atoms N_0 for the original samples, we determine the partial K-capture half-life $T_{1/2}^K = 455 \pm 40$ a. This has been calculated from $T_{1/2}^K = N_0 \omega_K \ln 2 / N_K$, where $\omega_K = 0.934$ is the K-fluorescence yield [7]. A ^{157}Tb half-life of 99 ± 10 a is readily deduced from the K-to-total electron capture ratio 0.218 (which corresponds to 62.9 keV, see section 4).

Previously published half-life data were based on the same technique, namely the measurement of the Gd K-X-ray emission rate. Unlike above, contributions from shells with principal quantum number greater than two had not been taken into account.

A comparison of the results on the half-life of ^{157}Tb is made in table 1. There the original values of the quoted references are included [6,8], as well as these same data but recalculated for capture to higher shells assuming the above K-to-total capture ratio 0.218. The agreement, notably with ref. 6, is good.

3. PHOTON TO K-X-RAY RATIO

The branching ratio b for electron capture to the excited and nuclear ground state of ^{157}Gd can be expressed as

$$b(Q) = \rho \frac{f(Q-54.5)}{f(Q)},$$

where ρ is the ratio of reduced transition probabilities. The f function for electron capture of a nucleus Z_A corresponds to the integrated Fermi function of β decay and is defined [9] as

$$f(q, Z) = \frac{1}{m^2 c^4} \sum_n \frac{\pi}{2} x_n^2 (q - W_n)^2 B_n,$$

where $mc^2 = 511$ keV is the electron rest mass, and ℓ_n is the amplitude of the radial wave function, with the index n denoting the respective atomic shell, $n = K, L_I, L_{II}, M_I, M_{II}, \dots$. The bracketed quantity $q - W_n$ is the neutrino energy, with q being the transition energy to the respective nuclear level in the daughter nucleus and W_n the binding energy of electrons of the n -th atomic shell. The factor B_n finally takes account of the effects of electron exchange and overlap.

Clearly, the ratio ρ is a necessary input to derive the Q -value. For ^{157}Tb , ρ can be estimated from the known nuclear structure of the two levels involved. For a decay scheme, see insert in fig. 1.

The decay of ^{157}Tb , characterized by the Nilsson quantum numbers $I^\pi_\Omega [Nn_z \Lambda] = \frac{3}{2}^+ \frac{3}{2} [411]$, populates the $I = \frac{5}{2}$ first rotational state and the ground state of ^{157}Gd with quantum numbers $\frac{3}{2}^- \frac{3}{2} [521]$. If both (first forbidden) beta transitions were of multipolarity 1, the Alaga intensity rules would predict the ratio of reduced transition probabilities $\rho = \langle \frac{3}{2} \frac{3}{2} 1 0 | \frac{5}{2} \frac{3}{2} \rangle^2 / \langle \frac{3}{2} \frac{3}{2} 1 0 | \frac{3}{2} \frac{3}{2} \rangle^2 = \frac{2}{3}$, where the brackets denote Clebsch-Gordan coefficients.

For the ground-state transition, however, multipolarity zero may also contribute, but as the contribution turns out to be small, it is safe to infer the ratio ρ from observations of the same beta transition (or its inverse) in neighbouring species. In fact, this transition has been observed in ^{155}Tb , ^{159}Dy , ^{161}Er , and ^{161}Tb , but its feeding intensity was measured accurately only for ^{159}Dy and ^{161}Tb . From these we extract $\rho = 0.53 \pm 0.05$ [10], implying that 20% of the ground-state transition is of multipolarity zero.

The function $b(Q)$ has then been calculated using f -functions which are based on wavefunctions of Bambynek et al. [9] and Dzhelepov et al. [11]. The graph of $b(Q)$ is shown in fig. 1.

The branching ratio can be determined experimentally from the 54.5 keV photon to Gd K-X-ray intensity ratio r , as given by the relation

$$b^{\text{exp}} = r(1 + \alpha) \omega_K \frac{f_K(Q)}{f(Q)},$$

with $\alpha = 12.13$ being the total conversion coefficient [12] of the 54.5 keV transition (whose multipolarity was taken as M1 + 3% E2 [13]); $f_K(Q)/f(Q)$ is the K-to-total electron capture ratio of the ground-state transition. This latter ratio is based on refs. 9 and 11. From measured photon spectra, $r = (4.16 \pm 0.10) \times 10^{-4}$ was obtained. This differs from the value published by Naumann and Hopke [4] by a factor of about 2.

The function b^{exp} is plotted in the same fig. 1. Thus the Q-value, $61.9 \pm (0.7 + 1.0)$ keV and the branching ratio $b \approx 10^{-3}$ can be read off directly. The statistical and the systematic contributions to the uncertainty of the Q-value are given separately. The systematic part of it, 1.0 keV, arises, to about equal parts, from uncertainties in ρ as well as in the amplitudes of the higher atomic shells. It should be stressed that the results only depend on the theoretical ratio $f_{M+N+O}(Q - 54.5)/f_K(Q)$, as can readily be proven from solving $b(Q) = b^{\text{exp}}(Q)$.

4. ELECTRON CAPTURE RATIOS

4.1 L-to-M ratio

If ^{157}Tb were also to decay by L-capture to the excited state in ^{157}Gd , i.e. for a transition energy $Q^* > 8.4$ keV, the Q-value could be probed very sensitively from the L-to M capture ratio ($Q^* = Q - 54.5$ keV). Consequently, an experiment was set up to search for coincidences between the L (and M) Auger electrons and the 54.5 keV γ -rays. (Throughout this paper the term 'Auger' electrons is meant to embody also 'Coster-Kronig' electrons.)

Owing to the very short range of the approximately 800 eV M-Auger electrons, a proportional counter containing the radioactive source was employed, which was in direct contact with the counting gas (flow counter). The photons were detected in a hyperpure Ge crystal. Fast coincidences were established by a time-to-amplitude converter. The timing resolution achievable was typically 100 ns FWHM. The flow of the He + 10% CH₄ counting gas was maintained at $1.0 \pm 0.1 \text{ cm}^3 \text{ s}^{-1}$, and the counting rate in the flow counter was monitored continuously. Coincidence data were taken over 20 days.

For the analysis, the Auger electron spectra gated by the coincident 54.5 keV γ -ray are compared with L- and M-Auger electron 'reference' spectra. These in turn have been created from the same set of coincidence signals by requiring, as conditions, $K\alpha_1$ - and $K\beta'_1$ -X-rays, respectively. Thus spectra of Auger electrons are obtained which originate from the filling of the X-ray-induced $2p_{3/2}$ (L_{III}) and $3p_{1/2}$, $3p_{3/2}$ (M_{II} , M_{III}) holes, respectively.

This comparison can be approximate only in so far as the primary vacancy distribution for electron capture differs from the X-ray-induced one, because electron capture leads mainly to the atomic s-states. However, it should be emphasized that the differences in the M-shell are significantly smoothed, as most 3s, 3p (M_I , ... M_{III}) vacancies undergo Coster-Kronig shifts to the 3d (M_{IV} , M_V) subshells before they are filled from the higher shells.

Besides subtraction of background and random coincidences, the spectra have been corrected for real coincidences between 54.5 keV K-conversion electrons (having an energy of 4.3 keV) and the Gd K-X-rays. The spectra are normalized in area to the γ -gated one. They are denoted by S_γ , $S_{K\beta}$, and $S_{K\alpha}$, and are shown in fig. 2. The obvious good agreement between S_γ and $S_{K\beta}$ already indicates the smallness of the L-to-M electron capture ratio P_L/P_M . Quantitatively it can be derived using the equation

$$S_\gamma = \left(S_{K\beta} + P_L/P_M S_{K\alpha} \right) / \left(1 + P_L/P_M \right).$$

A least-squares analysis leads to $P_L/P_M = (-0.010 \pm 0.003)$, with a reduced χ^2 of 1.07. The sign of P_L/P_M is attributed to the persistent, albeit small, background in the $K\beta'_1$ -gated spectrum. An upper limit on P_L/P_M of 0.006 has been estimated by assuming the $S_{K\beta}$ spectrum to vanish above 2.5 keV. This latter ratio leads to an upper limit of the Q-value of 63.4 keV.

It should be noted that the spectra S_γ and $S_{K\beta}$ could also be compared without being previously normalized to the same area. As will be shown in the next subsection, the differences in their areas can be understood.

4.2 M-to-total ratio

The same set of coincidence data can be analysed differently to estimate a Q-value.

Let $C_{\beta_1}^M$ denote the ratio of the number of coincidences between $K\beta_1'$ -X-rays and M-Auger electrons over the number of detected $K\beta_1'$ -X-rays in the corresponding photon singles spectrum. The following equation then holds (with C_γ^M defined equivalently for the 54.5 keV γ -ray):

$$C_\gamma^M = \frac{P_M}{P_{\text{tot}}} \times C_{\beta_1}^M.$$

This relation can be easily understood, as $K\beta_1'$ -X-rays produce only M-holes (see above), whereas the electron capture to the 54.5 keV level produces the whole series of M through O holes. However, in either case only the M-Auger electrons are detectable. Here the detection efficiency for each pair in the comparison (e.g. of 54.5 keV photon and 48.7 $K\beta_1'$ -X-rays) was presumed to be equal.

From the measured values we obtain the M-to-total ratio as 0.661 ± 0.006 .

This is related to the Q-value by the equation

$$\frac{P_M}{P_{\text{tot}}} = \frac{f_M(Q^*)}{f(Q^*)} = \left(1 + \frac{P_{N+O}}{P_M} \right)^{-1}$$

where Q^* is the transition energy to the 54.5 keV level, $Q^* = Q - 54.5$ keV. The nomenclature conforms to refs. 9 and 11, from whence also the numerical values have been taken. The relative amount of O-capture has been estimated based on X-ray emission rates for the filling of K-shell vacancies. From measured [14] and calculated [15] $KO/K\beta_2$ X-ray ratios we obtain $P_O/P_N \approx 0.15$.

The M-to-total ratio is displayed in fig. 3 (left-hand ordinate). We deduce a Q-value of $61.6 \pm (0.6 + 1.5)$ keV. The uncertainty quoted comprises a 0.6 keV statistical as well as a 1.5 keV systematic contribution, the latter arising from the poor knowledge of the amplitudes of the wave functions for the outer atomic shells. For example, when increasing the amplitude of the 4s (N_1) radial wave function by merely 5%, the extracted Q-value rises by 1.5 keV to 63 keV.

4.3 L-to-K capture ratio

This method relies on the electron capture to the ^{157}Gd ground state.

An experiment was set up to record X-ray spectra with both Si(Li) and high-purity Ge detectors. The efficiencies for L-X-rays and K-X-rays (~ 7 keV and ~ 45 keV, respectively) have been carefully determined using coincidences between the 53 keV γ -ray and K-X-rays of ^{133}Ba . Furthermore, the coincidence rate between L-X-rays and K-X-rays of ^{157}Tb and ^{152}Eu yields data on the efficiencies involved. These results were augmented by singles data of ^{241}Am and ^{57}Co . The ratio of efficiencies $\varepsilon(\text{L})/\varepsilon(\text{K})$ was found to be 5.24 for the Si(Li) and 0.3225 for the Ge detector. The smallness of the latter ratio is due to the attenuation of the 7 keV L-X-rays in the source backing.

The experimental L-to-K photon ratio R is then determined as 0.731 ± 0.020 by integrating the areas under the respective X-ray peaks, including the K-X-ray escape peaks. The effect of summing of an L-X-ray and its preceding $K\alpha$ -X-ray has also been corrected for.

Apart from absorption losses, the Ge detector is expected to have equal efficiencies at the L-X-ray and K-X-ray energies. With this assumption, another L-to-K photon ratio is gained, 0.753 ± 0.040 , from an independent experiment, where a 5% correction has been applied to take into account the absorption in the 13 μm thick Be window of the detector.

We insert the mean $R = 0.735 \pm 0.018$ into the following formula

$$\frac{P_L}{P_K} = \left(R - \frac{n_{KL} \omega_{KL}}{\omega_K} \right) \frac{\omega_K}{\omega_L},$$

with $n_{KL} = 0.85$ [7] the number of L-vacancies created by one primary K-vacancy; $\omega_K = 0.934 \pm 0.009$ and $\omega_L = 0.198 \pm 0.010$ [7] the K-shell and mean L-shell fluorescence yields, respectively; and $\omega_{KL} = 0.18$ the one for L-X-rays arising from an initial K-vacancy. We obtain $P_L/P_K = 2.69 \pm 0.20$, which agrees well with previously published values, as can be seen from table 2.

Finally, the expression

$$\frac{P_L}{P_K} = \frac{f_L(Q)}{f_K(Q)} \approx \frac{g_{L_I}^2(Q - W_{L_I})^2}{g_K^2(Q - W_K)^2} \left[1 + \frac{f_{L_{II}}^2}{g_{L_I}^2} \right] X^{L_I/K}$$

links the Q-value and the experimental L-to-K capture ratio (the right-hand side of the equation neglects the difference between L_I and L_{II} binding energies); g_K , g_{L_I} , and $f_{L_{II}}$ denote the amplitudes of the respective radial wave functions (for $Z = 65$), whereas W_{L_I} and W_K are the ($Z = 64$) gadolinium binding energies [16]. The exchange and overlap correction $X^{L_I/K} = B_{L_I}/B_K$ is a few percent larger than unity. The L-to-K ratio is displayed in fig. 3 (right-hand ordinate) for parameters taken from refs. 9 and 11.

We obtain the Q-value of $62.9 \pm (0.6 + 0.1)$ keV, where 0.1 is the systematic uncertainty due to differences between the theoretical calculations.

The derived Q-values are also given in table 2, together with the value of the 1977 mass evaluation for comparison [17].

Although all data on the Q-value of ^{157}Tb based on the L-to-K capture ratio seem to mutually agree within their uncertainties, the result of ref. 4 is notably high. This is especially surprising in the light of the good agreement of the input data, P_L/P_K , on which the Q-value is based. Probably it is simply due to a miscalculation in ref. 4.

5. BREMSSTRAHLUNG

Ideally, the Q-value could be obtained directly from the end point of the continuous bremsstrahlung spectrum.

A spectrum of photons in the X-ray domain coincident with L-Augur electrons has been created from the same data described in section 4. However, its limited statistics does not allow the end point to be extracted directly. Rather, the *whole* spectrum has to be examined. Consequently, theoretical spectra have been calculated based on the IBEC theory [1]. These spectra are to be compared with the experimental one, folding in a Gaussian resolution function.

The spectra are shown in fig. 4. Clearly, the Q-value can be constrained to the interval 60 to 64 keV. As the theoretical curve with the parameter $Q = 62$ keV describes the experimental one satisfactorily, we adopt as the Q-value 62 ± 2 keV.

No elaborate fitting of the spectra has been attempted, for two reasons. First, the response of the detector to monoenergetic photons is non-Gaussian, giving rise to the observed pedestal at the low-energy side of the $K\alpha$ -X-ray peak. Secondly, the $K\beta$ lines appear in the coincidence spectra. They, in turn, originate from two sources. When requiring a coincidence with Auger electrons of the interval 2.5 to 12 keV, there is a 2% M-Auger electron contribution connected with the $K\beta'_1$ -X-rays. Furthermore, in this interval the 4.3 keV K-conversion electrons of the 54.5 keV transition are also present, and are accompanied by K-X-rays as well. Thus the $K\beta'_2$ -X-ray peak, which can arise only from this effect, is understood quantitatively.

The effect of distortion of the spectra owing to pile-up was found experimentally to be small (see the insert in fig. 4).

6. CONCLUSION

The Q-value of ^{157}Tb has been measured by five different methods.

In a first group of experiments, electron capture ratios involving the excited level have been measured. In a second one, the L-to-K capture ratio for the ground state has been remeasured. The results obtained are summarized in table 3.

Consequently it can be stated that the transition energy $Q^* = Q - 54.5$ keV to the excited nuclear level is smaller than the 2s binding energy of 8.376 keV. This in turn implies that for this branch the calculations are based solely on the wave functions of the outer atomic shells, which are comparably uncertain. Explicitly, the Q-value based on the M-to-total ratio changes by as much as 1 keV by using the $g_{N_I}^2/g_{M_I}^2$ wave-function ratio of ref. 9 rather than that of ref. 11.

Experimental L-to-K capture ratios, on the other hand, are known to be well reproduced by theory. These arguments are reflected in the quoted systematic uncertainties. Therefore we do not average the results obtained but rather adopt 62.9 ± 0.7 keV as the Q-value of ^{157}Tb .

Unlike the above-discussed methods, the end point of the bremsstrahlung spectrum could yield a Q-value directly. However, because of the limited statistics, only the consistency with the other results can be stated.

From the adopted Q-value we re-evaluate the branching ratio $b = (1.1 \pm 0.1) \times 10^{-3}$, and the absolute transition probabilities, the ft-values. For the ground-state transition, a $\log ft = 7.28 \pm 0.05$ is obtained, and for the transition to the 54.5 keV level, a $\log ft = 7.66 \pm 0.08$.

For the electron capture to the excited state we thus obtain 8.4 ± 0.7 keV as the transition energy, which is remarkably close to the resonance energy of 8.376 keV. Therefore, future efforts should be aimed at observing the internal bremsstrahlung for this branch. Experiments with this objective have to be carried out in coincidence with photons or conversion electrons de-exciting the 54.5 keV level of the gadolinium daughter.

The minute branching ratio of 10^{-3} could, of course, impede any measurement attempting this. Nevertheless it seems feasible, taking advantage of the significant enhancement of the bremsstrahlung spectrum. In fig. 5 a calculated spectrum is shown which is based on the calorimetric detection of the total energy release [18]. Here the transition energy Q^* is the parameter. For those spectra as close as 500 eV to the 2s binding energy, the shape changes and a pronounced peak becomes visible at the high-energy end. No experimental resolution has been folded in.

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Table 1

Half-life of ^{157}Tb

$T_{1/2}[\text{a}]$	P_L/P_K	$T_{1/2}^{\text{corr}}[\text{a}]$	References
280 ± 120	2.64	220 ± 95	8
150 ± 30	2.18	100 ± 20	6
		99 ± 10	This work

Table 2

Q-values derived from L-to-K electron capture ratios

L-to-K photon ratio	P_L/P_K	Q (keV)	References
	2.18	~ 60	6
	2.64	~ 60	5
0.741 ± 0.030	2.65 ± 0.20	66 ± 6	4
0.735 ± 0.018	2.69 ± 0.20	62.9 ± 0.7	This work
Mass systematics		57.6 ± 2.3	15

Table 3

Results on the Q-value of ^{157}Tb of the different measurements

Experimental quantity	Level involved	Q $\pm \Delta Q$ (keV)
$I(54.5)/I(\text{Gd K-X})$ $\equiv r = (4.16 \pm 0.10) \times 10^{-4}$	Excited	$61.9 \pm (0.7 + 1.0)$
$P_M/P_{\text{tot}} = 0.661 \pm 0.006$	Excited	$61.6 \pm (0.6 + 1.5)$
$P_L/P_M < 0.006$	Excited	< 63.4
$P_L/P_K = 2.69 \pm 0.20$	Ground state	$62.9 \pm (0.6 + 0.1)$
Bremsstrahlung	Ground state	62 ± 2

Figure captions

- Fig. 1 : The branching ratio b as a function of the ground-state transition Q -value. The theoretical curve has been calculated from a ratio of reduced transition probabilities $\rho = 0.53$. The experimental one (with its uncertainty band) was obtained from the measured γ to K -X-ray ratio. The insert shows the decay scheme of ^{157}Tb .
- Fig. 2 : Experimental Auger-electron spectra, all normalized to the same area. The histogram is coincident with 54.5 keV γ -rays, whereas the smooth curves are coincident with $K\beta_1$ - and $K\alpha_1$ -X-rays, respectively. From the good agreement between the $K\beta_1$ and γ -gated spectra, a P_L/P_M ratio of less than 0.006 is obtained.
- Fig. 3 : Calculated electron capture ratios as a function of the ^{157}Tb Q -value. The left-hand ordinate refers to the M -to-total capture ratio, the right-hand one to the L -to- K capture ratio. The measured quantities are indicated together with the uncertainties.
- In the case of P_L/P_K , the solid curve has been computed on the basis of ref. 9, the dashed one using ref. 11.
- Fig. 4 : Photons that are coincident with electrons having energies between 2.5 and 12 keV. The $K\beta'_2$ -X-ray peak originates from coincidences with the 54.5 keV K -conversion electrons (see text). The insert displays the pulser peak in order to demonstrate spectral distortions due to pile-up.
- Fig. 5 : Calculated spectra of the total energy released are shown as a function of energy for the electron capture decay of ^{157}Tb to the 54.5 keV excited level in ^{157}Gd . As the parameter, the transition energy $Q^* = Q - 54.5$ is indicated in keV. Throughout the calculation a vanishing neutrino rest mass has been assumed. The pronounced peaks at 7.9 and 8.4 keV are of particular interest.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5