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THE STUDY OF ^{175}Hf DECAY

The intensities of lines of the internal conversion electron and γ-rays emitted at the decay of ^{175}Hf nucleus ($T_{1/2}$ = 70 days) have been measured with the help of a $\pi\sqrt{2}$ magnetic β-spectrometer and two coaxial HPGe-detectors. On the basis of those data, the β-ray branching ratios and the log ft values for β-transitions are calculated. The limits on the log ft values for β-transitions to ^{175}Lu levels $9/2^+$ at 113.8 keV, $9/2^-$ at 396.3 keV, and $9/2^+$ at 546.6 keV have been found for the first time.

Keywords: radioactivity, ^{175}Hf, γ-spectrum, HPGe-detector, internal conversion, magnetic spectrometer, intensity measurements.

1. Introduction

According to last experimental data [1], ^{175}Hf nucleus ($T_{1/2} = 70$ days) decays by capturing an electron onto excited ^{175}Lu states with energies of 514.6, 432.8, 353.3, and 343.4 keV (see Fig. 1). Both the mother and daughter nuclei belong to deformed nuclei ($\beta_2 \approx 0.3$). Some of ^{175}Lu levels are identified as one-particle, and they are related to the certain values of quantum numbers $K^\pi[\mathrm{N}n_Z\Lambda]$; the others are terms of rotational bands. This practice allows β-transitions to be classified not only in accordance with the spin selection rules, but also according to the selection rules with respect to the asymptotic quantum numbers. Having calculated the parameters $\log ft$ for β-transitions, the results obtained can be compared with the corresponding systematization data.

The total energy of electron capture amounts to 686.8 ± 1.9 keV. Therefore, ^{175}Hf can also decay onto the 396.3 and 546.6-keV levels, the former being the one-particle state $\pi 9/2^- 9/2$ [514], and the latter belonging to the rotational band constructed on the state $\pi 5/2^+ 5/2$ [402]. In this case, the transitions with energies of 203 and 396 keV should be observed. In Fig. 1, they are denoted by dashed arrows. The search for those transitions was another purpose of our researches.

2. Experimental Technique

The probabilities of β-transitions differ by several orders of magnitude depending on their types. Therefore, the intensities of transitions between the levels in the daughter nucleus are different by the same order of magnitude. This circumstance considerably complicates the corresponding measurements and requires that high-resolution spectrometers should be used.

^{175}Hf radiation sources were fabricated in the course of (n, γ) reaction on a research reactor at the Institute for Nuclear Research of the National Academy of Sciences of Ukraine. Hafnium targets both with the natural content of isotopes and enriched to 13.9% of ^{174}Hf were used.

The γ-spectra were measured on a γ-spectrometer consisting of two horizontal coaxial detectors fabricated from hyperpure germanium (GEM-40195 and

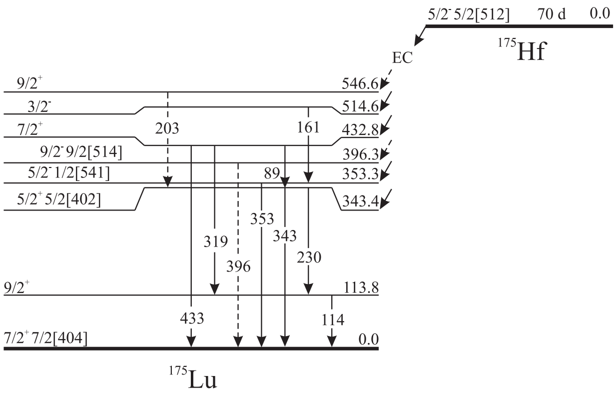

Fig. 1. Fragment of the ^{175}Hf decay scheme

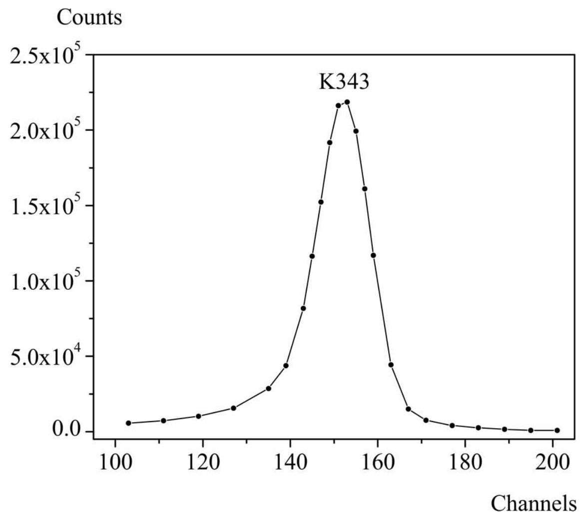

Fig. 2. ICE spectrum of γ343 keV transition at K-shell of ^{175}Lu. The ordinate axis demonstrates the number of counts during 1520 s for each point

GMX-30190 with a resolution of 1.73 or 1.89 keV, respectively, at the γ1332 line of ^{60}Co) and a 919 SPECTRUM MASTER multichannel buffer (ORTEC). The detectors were carefully graduated by the registration efficiency with the use of standard spectrometric sources of γ-radiation, namely ^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{228}Th, and ^{241}Am in the energy interval from 26 to 1620 keV. The shape of the registration efficiency curve was well described by the Campbell function [2]. The graduation error did not exceed 2% in the whole energy interval for both detectors.

Main attention was focused on the correct determination of intensities of weak γ-lines. In order to reduce systematic errors to a minimum, measurements were carried out in series on detectors of various types, at various amplification coefficients, and at various channel widths of an analog-to-digital converter (8192 and 16384 quantization levels for the input signal). In whole, 38 measurement series were performed.

The spectra of internal conversion electrons (ICEs) on the K- and L-shells of ^{175}Lu were studied with the help of a $\pi\sqrt{2}$ magnetic β-spectrometer with an iron yoke and an equilibrium orbit radius of 50 cm. The dependence of the electron counting rate on the voltage applied between the radiation source and the spectrometer chamber was registered. The magnetic field was kept constant and stabilized at three points along the radius using the nuclear magnetic resonance method. The stabilization scheme provided the spectrometer field stability at a level of 10^{-5} within a 24-h time interval. The high voltage applied to the radiation source also was stabilized with a relative accuracy of 5×10^{-5}.

The spectrometer registration system consisted of two Geiger–Müller counters arranged along the electron trajectories at a distance of 170 mm from each other. The counters were moved away from each other in order to reduce the background signal of a spectrometer at its operation in the coincidence mode. Counter 1 had an internal diameter of 15 mm and was oriented vertically, whereas counter 2 had a diameter of 46 mm and was oriented horizontally. The own background value of counter 1 was equal to 10 count/min. In the coincidence mode, the average counting rate amounted to 4 count/h.

The spectrometer resolution was 0.03% of the pulse at a solid angle of 0.07% of 4π. The spectrometer characteristics allowed the relative intensities of conversion lines to be measured with an accuracy of 1% [3].

The better resolution of a β-spectrometer and a considerably lower background intensity near the probable location of ICE lines of γ-transitions with energies of 203 and 396 keV made it possible to weaken the restriction on the intensities of those transitions by more than two orders of magnitude in comparison with the results of γ-spectroscopy. The sections of ICE spectra of γ343 and γ203 keV transitions on the K-shell of ^{175}Lu are depicted in Figs. 2 and 3, respectively.

In general, two series of measurements similar to those exhibited in Fig. 3 were performed. The total exposition time amounted to 16000 s at every spectral point.

Table 1. **Intensities of γ-rays emitted at ^{175}Hf decay**

E_γ, keV	Intensity, rel. units						
	This work	[6]	[7]	[8]	[9]	[10]	[11]
89.362	34.5 ± 1.5	28.86 ± 0.22	–	26.1 ± 3.4	29 ± 5	–	40
113.801	3.46 ± 0.20	3.52 ± 0.32	–	3.6 ± 0.5	3.5 ± 0.6	–	3.6
161.20	0.45 ± 0.14	–	–	0.27 ± 0.10	–	–	0.3
229.609	8.29 ± 0.19	8.13 ± 0.20	–	8.9 ± 1.8	8.8 ± 1.8	–	7.3
318.971	1.57 ± 0.23	–	–	2.1 ± 0.5	–	–	–
343.410	1000	1000	1000	1000	1000	1000	1000
353.3	2.66 ± 0.18	2.72 ± 0.20	2.1 ± 0.2	2.7 ± 0.5	–	2	–
432.771	17.5 ± 0.4	17.09 ± 0.31	–	17.1 ± 2.3	19 ± 4	–	16.0

The ICE spectrum near the probable location of line $L_1$396 keV was studied with the same carefulness. A higher energy of internal conversion electrons at this transition made it possible to carry out the measurements in the coincidence mode, which considerably reduced the background signal. The total exposition time at those measurements amounted to 8960 s at every spectral point.

3. Results and Their Discussion

The γ-spectra were analyzed using the software package WinSpectrum [4]. The analysis also included the half-life period in order to exclude possible influences of radionuclide impurities. The final values of γ-line intensities were determined as weighed averages over 38 measurement series. Either the weight error or the spread one, depending on which of them was larger, was used as the uncertainty of experimental values. The results of our measurements are quoted in Table 1 together with the data obtained in the best works. The energies of γ-transitions were taken from work [5]. One can see that our data agree well with the results obtained in other experimental works. The application of detectors of different types allowed the determination accuracy for the relative intensities of γ-rays to be improved.

The ICE spectra were analyzed taking advantage of self-developed software programs [3]. Similarly to the case of γ-spectrum for ^{175}Hf, no ICE lines were observed, which could belong to the transitions with energies of 203 and 396 keV. Only the limiting intensity ratio values for lines K203 and $L_1$396 keV were obtained: $I(\mathrm{K}203)/I(\mathrm{K}343) \le 4.6 \times 10^{-5}$ and $I(\mathrm{L}_1 396)/I(\mathrm{K}343) \le 3.7 \times 10^{-6}$.

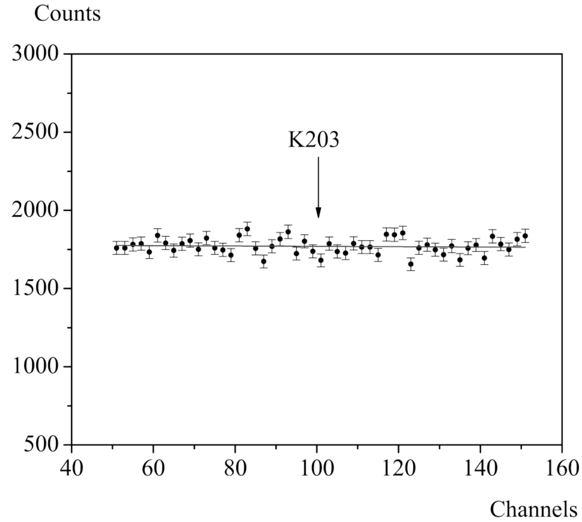

Fig. 3. ICE spectrum near a possible location of line K203 keV. The ordinate axis demonstrates the number of counts during 3200 s for each point. Solid curve demonstrates the background signal for this spectral section

The intensities of branches for the electron capture by ^{175}Hf, I_ε^k, were determined as solutions of a system of equations, each of which described the balance between the transition intensities for a definite k-th excited state of ^{175}Lu:

$$I_\varepsilon^k + \sum_i I_\gamma^i (1 + \alpha_i) = \sum_j I_\gamma^j (1 + \alpha_j), \qquad (1)$$

where I_γ^i and α_i are the intensities and the total conversion coefficients, respectively, for γ-transitions that populate the k-th level; and I_γ^j and α_j are their analogs for γ-transitions that depopulate this level.

Table 2. **Branching intensities and log ft values for β-transitions of ^{175}Hf**

E_ε, keV	Level E, keV	I_ε, %	Log ft	β-transition type	Additional classification
140.2	546.6	$\leq$0.014	$\geq$8.6	First forbidden unique	Unhindered
172.2	514.6	0.076 ± 0.024	9.24 ± 0.14	Allowed	Hindered
254.0	432.8	20.1 ± 0.8	7.27 ± 0.02	First forbidden	Unhindered
290.5	396.3	$\leq$ 0.010	$\geq$ 10.7	Second forbidden	"
333.5	353.3	0.155 ± 0.028	9.66 ± 0.09	Allowed	Hindered
343.4	343.4	79.6 ± 1.2	6.99 ± 0.02	First forbidden	Unhindered
573.0	113.8	$\leq$0.017	$\geq$11.2	First forbidden unique	"
686.8	0.0	$\leq$7*	$\geq$8.7*	First forbidden	"

* Data of work [1].

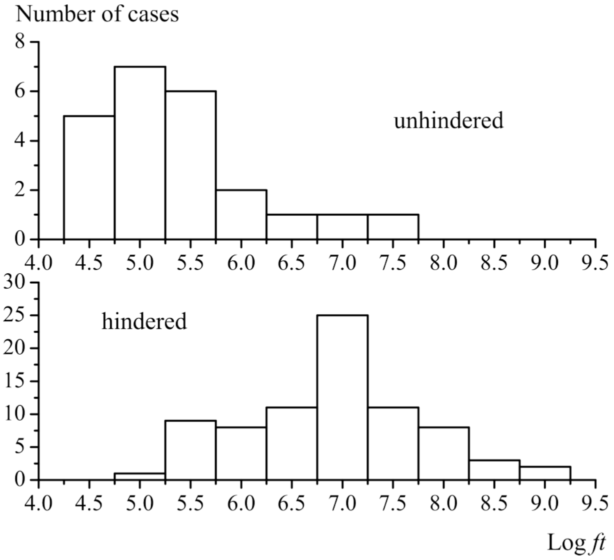

Fig. 4. Distribution of experimental log ft values for allowed β-transitions

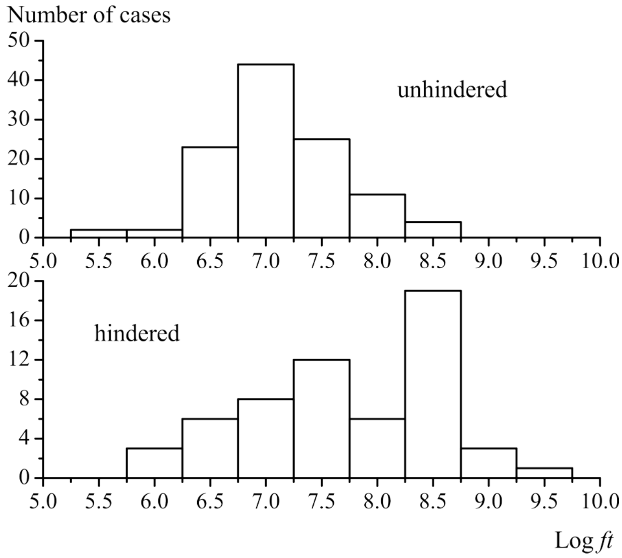

Fig. 5. The same as in Fig. 4, but for first forbidden β-transitions

Conclusions concerning the multipole composition of transitions were drawn after a detailed analysis of experimental data on the internal conversion [8, 9, 11–15] and angular correlations [16]. Then those conversion coefficients were calculated with the help of the software program BrIcc [17, 18].

The results obtained for the branching intensities of electron capture by ^{175}Hf are quoted in Table 2. These data were used, while calculating log ft for β-transitions. For this purpose, we used the software program LOGFT [19]. The results of calculations are also listed in Table 2. It should be noted that the restrictions on the parameter log ft for β-transitions on levels $9/2^+$ 113.8 keV, $9/2^-$ 396.3 keV, and $9/2^+$ 546.6 keV of ^{175}Lu were obtained for the first time.

The last column in Table 2 gives the classification of β-transitions not only according to the spin and parity selection rules [20], but also the selection rules for the asymptotic quantum numbers [21]. Decays, in which those selection rules are obeyed, are called unhindered; otherwise, hindered.

The violation of the selection rules for the asymptotic quantum numbers at the decay of deformed nuclei is assumed to result in a reduction of the probability of β-transitions by 1 to 3 orders of magnitude. Let us try to determine whether this is true by comparing our results with the magnitude of log ft for β-transitions of this kind in the concerned range of nuclei.

Among plenty of works dealing with the systematization of probabilities of β-transitions depending on their type, we may distinguish those by L.M. Zyryanova [22], K.Ya. Gromov [23], and B. Singh *et al.* [24]. In [22], the experimental values of log ft for unique β-transitions, which were known at that time, were reported. However, the body of those data turned out insufficient for making also the classification according to the selection rules for the asymptotic quantum numbers. The corresponding analysis for allowed β-transitions was made, in particular, in work [23]. It was found that all 28 experimental values of log ft known at that time for the transitions concerned fell within the following limits: for the allowed unhindered transitions, $4.6 < \log ft < 4.8$, and for the allowed hindered transitions, $5.5 < \log ft < 8.0$. However, for the classification of transitions of other types, the available body of experimental data turned out too small.

About 3900 experimental values of log ft for β-transitions, grouped according to their types, were reported in work [24], and the parameters of the corresponding distributions were calculated. However, the classification according to the selection rules with respect to the asymptotic quantum numbers was not made. We have analyzed all the data available at present [25] on the allowed, first forbidden, and first forbidden unique β-transitions in odd deformed nuclei of the rare-earth element group ($151 < A < 193$), which were taken from work [24], and confronted them, where it was possible, with the corresponding quantum numbers $K^\pi[Nn_Z\Lambda]$. In general, 287 transitions were managed to be identified. In Figs. 4 to 6, the distributions of experimental log ft values for transitions of various types are exhibited separately for unhindered (upper histograms) and hindered (lower histograms) β-decays.

The experimental log ft values for β-transitions fall within the following limits:

• allowed unhindered transitions: $4.5 \leq \log ft \leq$ ≤ 7.7, the average value equals 5.3;

• allowed hindered transitions: $5.2 \leq \log ft \leq 9.1$, the average value equals 6.9;

• first forbidden unhindered transitions: $5.6 \leq$ $\leq \log ft \leq 8.5$, the average value equals 7.1;

• first forbidden hindered transitions: $5.9 \leq$ $\leq \log ft \leq \leq 9.5$, the average value equals 7.7;

• first forbidden unique unhindered transitions: $8.5 \leq \log ft \leq 10.3$, the average value equals 9.1;

• first forbidden unique hindered transitions: $8.7 \leq$ $\leq \log ft \leq 11.3$, the average value equals 9.5.

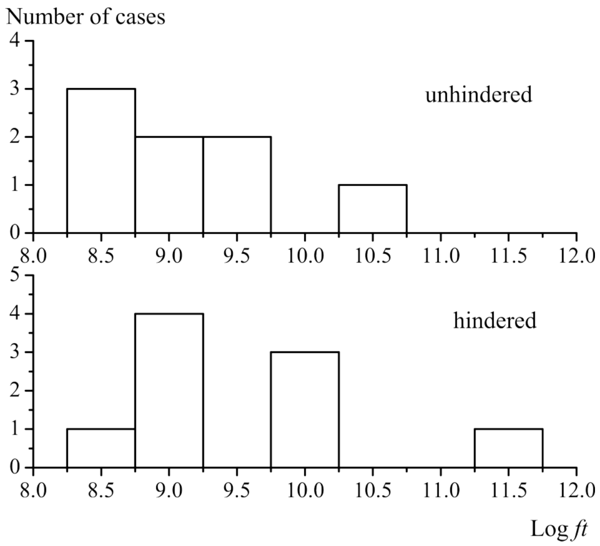

Fig. 6. The same as in Fig. 4, but for first forbidden unique β-transitions

From our research, it follows that the difference between the probabilities of unhindered and hindered β-decays is more substantial for allowed transitions. To our knowledge, the analysis for the first forbidden and first forbidden unique β-transitions was carried out for the first time.

The experimental values of log ft for first forbidden unhindered β-transitions in Table 2 agree very well with our systematization. The log ft values for allowed hindered β-transitions are at the upper limit of values for the decays of such a type. This circumstance can be explained by the fact that those transitions are accompanied by the variation in the quantum number K by two, i.e. such transitions are K-forbidden.

Only the limiting log ft values were obtained for the first forbidden unique unhindered transitions onto levels $9/2^+$ 113.8 keV and $9/2^+$ 546.6 keV of ^{175}Lu. Provided that the experimental accuracy could be increased by an order of magnitude, we may expect for their reliable observation. The estimated log ft value for the double forbidden β-transition onto level $9/2^-$ 396.3 keV of ^{175}Lu does not contradict experimental data for this range of nuclei (see, e.g., work [26]).

4. Conclusions

Owing to the application of a combined approach to the measurement, processing, and analysis of data, as well as to the application of various types of semiconducting detectors, we managed to improve the determination accuracy for the relative intensities of γ-rays emitted at the ^{175}Hf decay. Using the measurement data for the spectra of internal conversion electrons, the limiting values for the intensities of transitions with energies of 203 and 396 keV were obtained for the first time.

The intensities and log ft values were calculated for seven branches of electron capture by ^{175}Hf. The restrictions on log ft for β-transitions onto ^{175}Lu levels $9/2^+$ 113.8 keV, $9/2^-$ 396.3 keV, and $9/2^+$ 546.6 keV were also obtained for the first time.

The experimental log ft values for allowed, first forbidden, and first forbidden unique β-transitions in odd deformed nuclei of the rare-earth element group ($151 < A < 193$) were systematized in accordance with the selection rules for the asymptotic quantum numbers. The statistical distributions were plotted and the average log ft values were calculated for the transitions of various types. It was found that the difference between the probabilities of unhindered and hindered β-transitions is observed for all types of transitions. It is the largest for the allowed decays (by a factor of 40) and the smallest for the first forbidden (by a factor of 4) and first forbidden unique β-transitions (by a factor of 2.5). Such an analysis for the first forbidden and first forbidden unique β-transitions was performed for the first time. The systematization will be useful both at the analysis of newly obtained log ft values and while estimating the probabilities of still non-registered decay branches of deformed nuclei.

All log ft values obtained in this work for β-transitions at the ^{175}Hf decay agree with the presented systematization and do not contradict other experimental data for this range of nuclei. This fact allows a conclusion to be drawn that the identification of ^{175}Lu levels and the values of asymptotic quantum numbers that were assigned to them is adequate.

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ДОСЛІДЖЕННЯ РОЗПАДУ ^{175}Hf

Р е з ю м е

За допомогою магнітного β-спектрометра типу $\pi\sqrt{2}$ та двох коаксіальних HPGe-детекторів поміряні інтенсивності ліній електронів внутрішньої конверсії та γ-променів із розпаду ^{175}Hf ($T_{1/2} = 70$ днів). Ґрунтуючись на цих результатах, розраховані інтенсивності гілок електронного захвату та log ft β-переходів. Вперше отримані обмеження для log ft β-переходів на рівні $9/2^+$ 113,8 кеВ, $9/2^-$ 396,3 кеВ та $9/2^+$ 546,6 кеВ ^{175}Lu.