PRECISE MEASUREMENT Of THE ENERGY OF GAMMA-RAYS FROM THE DECAY OF ^{181}Hf

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High-precision measurements of the energies of some γ-rays from the decay of ^{181}Hf have been performed with a γ-spectrometer. Using these data, the energies of all levels and γ-rays in ^{181}Ta populated in the decay of ^{181}Hf were determined with the accuracy up to a few electron-volts.

1. Introduction

Beta-decay of ^{181}Hf ($T_{1/2}$ = 42.4 days) into the levels of ^{181}Ta is well understood. The decay scheme (see Fig. 1) has been established, the intensities of γ-rays have been determined to a high accuracy [1], while the transition energies have been measured up to several tens of electron-volts at best, according to the recent compilation [2].

The data on energies of excited states of atomic nuclei known with an accuracy of several electron-volts and higher become increasingly required today. The evolution of the technique of high-precision measurements of γ-ray energies based on semiconductor spectrometers, along with the essential extension of the nuclear-spectroscopic standards mesh, have provided a great scope for comprehensive measurements of energies of excited nuclear states populated in the decay of sources with more or less noticeable life-time.

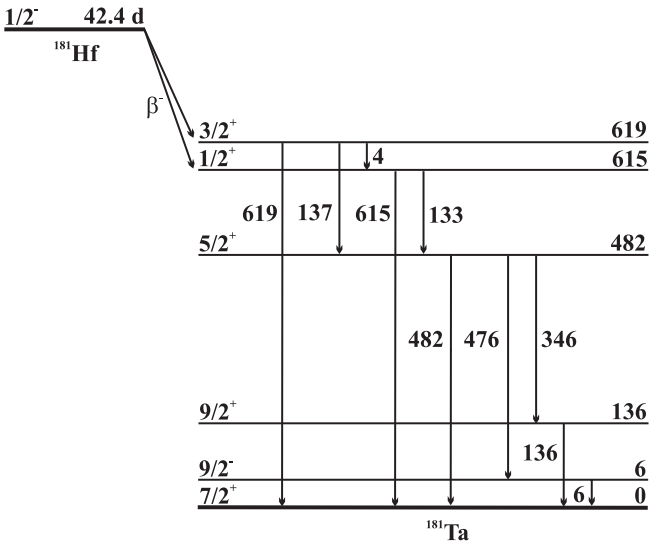

Fig. 1. The decay scheme of ^{181}Hf

Isotope ^{181}Hf appeared to be an appropriate object for such a purpose. Our long-standing researches [3–5] allowed the high-precision determination of the energies of two γ-quanta and the energy difference for three pairs of γ-transitions accompanying the decay of ^{181}Hf. In the present paper, the energies of additional three γ-rays have been measured, which enables the energies of γ-rays and all the ^{181}Ta levels populated in the ^{181}Hf decay to be measured to within several electron-volts.

2. Experimental Technique

The number of levels excited in the radioactive decay of a mother nucleus is generally less than the number of γ-rays deexciting these levels. It is not necessary for all γ-rays to be measured, in order to get information about their energies. The reference nuclear transition method can be used instead. Application of this method allows one to essentially reduce laboriousness of the experiments.

The procedure of determination of the energies of excited nuclear states and γ-rays deexciting these states by the reference nuclear transition method reduces to the following basis stages:

1) the single intense γ-lines most appropriate for measurement are chosen as reference lines;

2) the collection of references is selected from the list of recommended energy standards for nuclear spectroscopy. In order to minimize the errors arising from the ambiguity of a calibration curve, it is necessary to select such references that would be close to the measured γ-line while still being easily resolved in the spectrum;

3) the mixed radioactive source of a required composition is prepared with desired ratios of specific activities of radionuclides present in the source. For the statistical error (uncertainty in the determination of the

distance between lines) to be minimized, the reference and measured lines should have close intensities;

4) to minimize possible systematic errors, the measurements are performed by series on different detectors, at various geometries, different amplification coefficients, and different quantization levels of the input signal on amplitude-digital converters;

5) the energies of reference transitions are determined;

6) to calculate the energies of levels, a system of linear equations is derived, and then it is solved with the least-squares procedure;

7) the energies of all γ-quanta accompanying the decay of a mother nucleus are calculated on the basis of the obtained data.

The technique of such measurements and the problems concerning the preparation of a mixed radioactive source of optimal composition were reported in [6, 7] in detail.

As the energy standards for determining the energies of reference transitions, the γ-lines from the ^{192}Ir decay were used, because they are best suited for such a purpose. These lines are the recommended standards for the γ-ray energy calibration, and their energies are determined with a high accuracy [8, 9]. The reference γ-lines are located close enough to the γ-lines under consideration, while still being easily resolved in the spectrum, which allows their energies to be measured with a high accuracy.

The energy differences for three pairs of γ-lines (γ346-γ316, γ615-γ612, and γ619-γ612) were measured with a gamma-spectrometer, which comprises two horizontal detectors made from high-purity germanium (coaxial GEM-40195 with the 1.73-keV resolution for γ1332-line of ^{60}Co and planar GLP-36360/13 with the 580-eV resolution for γ122-line of ^{57}Co) and a multichannel ORTEC buffer 919 SPECTRUM MASTER. The first γ-transition from each pair is excited in the ^{181}Hf decay, while another transition, whose energy is known with a high precision, accompanies the ^{192}Ir decay.

To prepare a mixed radioactive source of the required composition, ^{181}Hf was obtained in the (n, γ) reaction under irradiation of enriched hafnium (^{180}Hf isotope content is 96.2%) with a research reactor WWR-M. ^{192}Ir ($T_{1/2}$ = 74 days) was also produced in the (n, γ) reaction under irradiation of enriched iridium (^{191}Ir isotope content is 94%) by slow neutrons.

The functional dependence of the energy calibration of a γ-spectrometer was investigated in detail. It was established that the deviation from linearity does not exceed 3×10^{-5} for the energy range from 84 to 604 keV. To minimize possible systematic errors, we performed a series of measurements using sources with different ratios of specific activities of ^{181}Hf and ^{192}Ir, at different gains and channel widths of an amplitude-digital converter (4096 and 8192 quantization levels of the input signal). In all, we performed 27 series of measurements of the γ346-γ316 energy difference, 17 series of measurements of the γ615-γ612 energy difference, and 12 series of measurements of the γ619-γ612 energy difference.

3. Results and Discussion

The γ-spectra were treated using the programs developed by us [10] based on the method of fitting the "instrumental" peak into the spectrum region of interest. This method allows a high-precision measurement of energies and intensities of the components in the case of lines of asymmetric shape and overlapping lines. This technique implies measuring a single gamma peak from the obtained spectrum (or, if such a peak is absent, a specially measured single gamma peak with shape similar to that of the line in the studied region of the spectrum) with high statistical accuracy. After the subtraction of the background, it is described by the multiple cubic-spline interpolation, and it is used as an "instrumental" peak, i.e. defines the experimental peak shape for the subsequent analysis by the least-squares method.

First, the differences between the γ-line energies were determined as a weighted mean from the results of all measurements, and then the transition energies were found. The measurement results are in good agreement with one another. All measured differences in γ-line energies, energies of reference γ-ray photons, and the obtained energies of the γ-transitions under study are listed in Table 1. The measurement errors and their components are also given. In addition, Table 1 contains the results of our previous studies.

As can be seen in Table 1, the energies of γ346 and γ482 transitions were determined from measurements of the energy differences for two different pairs of γ-lines. The results correlate within two root-mean-square deviations. For γ346, the difference is (5.5 $\pm$ 3.5) eV, while it amounts to (7.9 $\pm$ 3.4) eV for γ482. In our opinion, the reason for this discrepancy is the following.

It is the common knowledge that the distance between the reference line and a γ-line in the spectrum plays an important part in such measurements. On the one hand, the closer these lines, the less is the

nonlinearity of the spectrometer calibration curve. On the other hand, the closer these lines, the larger is an error of the determination of a line location in the decomposition of the doublet into components.

The full width at half maximum (FWHM) is about 0.7 keV in the region of γ346 and 0.9 keV in the vicinity of γ482. When employing γ344 ^{152}Eu as the reference line, we get 1656 eV for the distance between lines amounting up to 2.4 FWHM. Using γ484 ^{192}Ir as the reference line results in 2394 eV for the distance, which comprises 2.7 FWHM. In both cases, the lines were close enough and were not resolved completely, which had eventually an effect on the determination of a location of the lines in the spectrum.

Only comparison with the data of independent measurements can provide the answer to the question how well the energies of the γ346 and γ482 lines agree with each other. Such data do really exist. In [11], the differences in energies between lines of internal conversion electrons $K136 - K133 = (3192.6 \pm 0.30)$ eV have been measured very precisely with a magnetic β-spectrometer. According to our measurements on a γ-spectrometer, this difference in transitions is found to be $\gamma482 - \gamma346 - \gamma133 = (3191.0 \pm 3.9)$ eV. One can see an excellent agreement. This fact directly confirms that the systematic errors are insignificant, and we estimated them correctly.

Unfortunately, we failed to determine the energies of the γ615 and γ619 keV transitions with a reasonable accuracy. Low intensities of these γ-lines in the spectrum give no way of attaining a sufficient statistical accuracy of measurements.

Using the data on the transition energies and the Ritz rule for cascade transitions ($E_1 + E_2 = E_3$, where E_3 is the energy of the closing direct transition between the boundary levels), we formulated the system of linear approximate equations of different weights to calculate the level energies:

$$a_1x + b_1y + \cdots + m_1\nu = t_1 \pm \Delta t_1, \tag{1}$$

$$a_2x + b_2y + \cdots + m_2\nu = t_2 \pm \Delta t_2, \tag{2}$$

$$\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots, \tag{3}$$

$$a_Nx + b_Ny + ... + m_N\nu = t_N + \Delta t_N, \tag{4}$$

where a, b, ..., m are the specified numbers (generally they are equal to $\pm$ 1 or zero), t and Δt are the transition energies and their errors; x, y, ..., ν are the unknown level energies.

Since N is larger than the number of unknowns, the system was solved by the least-squares method [12]; i.e., we determined such values of unknowns, at which the sum

$$\sum_{i=1}^{N} p_i(t_i - a_ix - b_iy - \cdots - m_i\nu)^2 \tag{5}$$

$(p_i = (\Delta t_i)^{-2};\ i = 1, \ldots, N)$ was minimum.

The errors of all parameters can be obtained using the parabolic dependence $\chi^2 = \chi^2(\alpha_i)$, where $\alpha_i(x, y, \ldots, \nu)$ is the parameter under study. In this case, all other parameters are fixed and correspond to optimal values. The standard errors $\Delta\alpha_i$ are determined, by using the relation

$$\chi^2(\alpha_i^{\rm opt} \pm \Delta\alpha_i) = \chi^2_{\rm min} + 1, \tag{6}$$

T a b l e 1. Measured differences in the energies of γ-lines, energies of reference γ-quanta, and obtained energies of γ-transitions under study

Measured difference in the energies of γ-lines	Differences in energies, eV	Energy of the reference γ-quantum, eV	Energy of the desired γ-quantum, eV	Errors in determining the γ-transition energy, eV					Note
				statistical	calibration	reference energy	systematical	total	
$\gamma133(^{181}Hf)$-$\gamma130(^{169}Yb)$	2534.0	130522.93	133056.9	0.8	1.0	0.06	–	1.3	[5]
$\gamma346(^{181}Hf)$-$\gamma344(^{152}Eu)$	1656.0	344278.5	345934.5	2.1	0.6	1.2	2.2	3.3	[5]
$\gamma346(^{181}Hf)$-$\gamma316(^{192}Ir)$	29433.8	316506.18	345940.0	0.7	0.9	0.17	–	1.2	
weighted mean			345939.4					1.8	
$\gamma484(^{192}Ir)$-$\gamma482(^{181}Hf)$	2394.1	484575.1	482181.0	1.7	1.9	0.4	1.5	3.0	[5]
$\gamma482(^{181}Hf)$-$\gamma468(^{192}Ir)$	14120.0	468068.9	482188.9	0.4	0.4	0.26	1.4	1.5	[5]
weighted mean			482187.3					3.2	
$\gamma615(^{181}Hf)$-$\gamma612(^{192}Ir)$	2737	612462.15	615199	19	0.1	0.26	–	19	
$\gamma619(^{181}Hf)$-$\gamma612(^{192}Ir)$	6530	612462.15	618992	34	0.2	0.26	–	34	
$\gamma482(^{181}Hf)$-$\gamma476(^{181}Hf)$	6222.0			1.5	0.2		–	1.5	[4]
$\gamma137(^{181}Hf)$-$\gamma133(^{181}Hf)$	3794.0			0.9	0.1		–	0.9	[3]
$\gamma137(^{181}Hf)$-$\gamma136(^{181}Hf)$	601.4			0.9	0.1		–	0.9	[3]

where $\alpha_i^{\rm opt}$ is the optimal value of the parameter α_i which minimizes χ^2.

After the determination of the energies of nuclear excited states, it was quite easy to calculate the γ-transition energies between these states. The results of calculations, along with the weighted mean values from the compilation [2], are shown in Table 2.

It should be noted that the Ritz rule refers to the energies of transitions, not to γ-quanta: the energy of a recoil nucleus at emitting a γ-quantum should be taken into account. The transition energy is defined by the expression [13]

$$E_n = E_\gamma + \frac{E_\gamma^2}{2Mc^2}, \qquad (7)$$

where M is the mass of the recoil nucleus. Corrections for transition energies following from this formula have been taken into account in the calculation of the energies of ^{181}Ta levels and the energies of γ-rays deexciting these levels.

We determined the energies of 5 levels of ^{181}Ta and the energies of 10 γ-quanta accompanying the decay of ^{181}Hf with an accuracy exceeding the known values by an order of magnitude. Most of them completely correspond to the requirements imposed on the energy standards.

4. Conclusions

At present, the list of the recommended energy standards for nuclear spectroscopy includes about 240 γ-lines covering the energy range from 24 up to 4806 keV [8, 9]. For all of them, the relative error in energy definition does not exceed 10^{-5}.

The reference line should be close to the measured γ-line for the precision determination of transition energies. This allows the minimization of the errors arising due to an ambiguity of the calibration curve of a spectrometer. Therefore, in the choice of reference γ-lines, the presence of convenient nuclear-spectroscopic standards in a given part of the spectrum is of no small importance.

^{181}Hf isotope is sufficiently produced in the slow-neutron reaction (n, γ) (activation cross-section is about 14 barns [14]. There is no need for using hafnium enriched by ^{180}Hf isotope, because the content of this isotope in a natural isotopic mixture exceeds 35%. ^{175}Hf ($T_{1/2}$ = 70 days) which is produced in such a case does not prevent ^{181}Hf from being used as a calibration source.

T a b l e 2. Energies of the ^{181}Ta levels and γ-quanta excited in the ^{181}Hf decay

Present work		[2]	
level energies, eV	γ-quantum energies, eV	level energies, eV	γ-quantum energies, eV
6222.0(15)	6222.0(15)	6237(20)	6240(20)
136248.7(14)	136248.6(14)	136262(13)	136269(13)
482188.0(11)	345938.9(18)	482168(23)	345970(40)
	475965.3(19)		475990(90)
	482187.3(11)		482170(30)
615244.5(13)	133056.4(17)	615190(30)	133027(18)
	615243.4(13)		615170(110)
619038.3(11)	3793.8(17)	618990(50)	3900(100)
	136850.1(16)		136970(60)
	619037.2(11)		618660(80)

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ПРЕЦИЗІЙНІ ВИМІРИ ЕНЕРГІЇ ГАММА-ПРОМЕНІВ ІЗ РОЗПАДУ ^{181}Hf

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Р е з ю м е

На γ-спектрометрі з високою точністю поміряно енергії опорних γ-переходів із розпаду ^{181}Hf. Спираючись на ці дані з похибкою в декілька електрон-вольт визначено енергії γ-променів та всіх рівнів ^{181}Ta, які збуджуються в розпаді ^{181}Hf.