DETERMINATION OF THE ENERGY STANDARDS
BY PRECISION BETA-SPECTROSCOPY METHODS

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The technique allowing one to determine energy of transitions in atomic nuclei with high accuracy is developed. It is based on measurement of a difference of energy of internal conversion electron lines on high-resolution β-spectrometer $\pi\sqrt{2}$ with iron yoke and radius of an equilibrium orbit r = 50 cm at a constant magnetic spectrometer field. The accuracy of definition of γ-ray energies reaches from 0,2 up to 1 eV in the energy region from 100 up to 500 keV. That completely corresponds to the requirements to energy standards of 4-th order.

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1. INTRODUCTION

The relative measurements, as a rule, are carried out easier than absolute ones: various physical quantities and mistakes need not to be measured; and hence uncertainties and regular errors related to these quantities are eliminated. It is especially convenient to carry out comparisons with the objects for which the quantity to be measured is already known with high accuracy. The preliminary choice or creation of such objects is necessary. Nuclear spectroscopy standards can be used as the base objects in nuclear spectroscopy. According to B.S. Dzhelepov [1], it is possible to classify them as follows:

1. The transition in ^{86}Kr, having wave-length in vacuum λ = 6057,80211 Å, is called *standard of the first order*.

2. *Standards of the second order* are selected according to the following rules:

a) wave-length of the second-order standard should be determined by comparison with the wave-length of the first-order standard;

b) the comparison is carried out with accuracy corresponding to the best modern works.

3. *The third-order standards* are

a) the nuclear transitions, wave-lengths of which are determined by comparison with the wave-lengths of either first-order standards, or second-order ones. The comparison should be made at a modern level of accuracy by means of the methods, which are based on the well-known physical laws;

b) the transitions energies of which are determined using two standards of the third order and the Ritz rule: $E_1+E_2=E_3$.

4. Spectral lines, energies of which are determined by comparison with the standards of the second and third order, are called *the fourth-order standards*. It is possible to use the methods in which energy of the transition is determined by using a curve constructed on several standards of the second and third order.

As the second-order standard, γ411,8 ^{198}Hg is used. Its energy is 411,80205 ± 0,00017 keV [2] for today. The requirements to accuracy of definition of energy of the third and fourth-order standards constantly vary with improvement of the measurement methods. According to last Helmer review [3], the list of the recommended energy standards for nuclear spectroscopy includes only those γ-rays, for which the relative error in energy definition does not exceed 10^{-5}.

Last years, along with crystal diffraction spectrometers, X-ray HPGe-detectors which allow determination of the energy of transitions in the region up to 300 keV with accuracy about 1 eV are widely used for precision measurements of energy of γ-rays arising in the decay of radioactive nuclei. Due to them, the number of standards is increased up to 260. They cover the energy range from 24 up to 4800 keV. However, it is not enough for needs of nuclear spectroscopy.

2. EXPERIMENTAL TECHNIQUE

2.1. EQUIPMENT

We developed a technique allowing us to reach high accuracy of definition of transition energies on magnetic β-spectrometer $\pi\sqrt{2}$ with iron yoke and radius of an equilibrium orbit ρ = 50 cm. It is based on measurement of energy difference of two internal conversion electron lines at a constant magnetic field of the spectrometer. One of these lines, which energy is well-known, is using as standard.

In the experiment we measured dependence of electron rate on the value of voltage enclosed between the source and the chamber spectrometer. The distance between conversion lines is obtained directly in electron-volt. Stability of the voltage, applied to the source, is maintained through the specially constructed

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block. It ensures the voltage accuracy of $5 \cdot 10^{-5}$, and the drift of voltage for seven day of continuous work of the system does not exceed $8 \cdot 10^{-5}$. The magnetic field of spectrometer is stabilized in three points along radius by a nuclear magnetic resonance method (NMR) with accuracy not worse than $5 \cdot 10^{-6}$ [4]. The registration of conversion electrons is carried out by two Geiger-Muller counters included in the coincidence scheme.

To reduce possible instability in the work of the system we carried out short sets of measurements with accumulation of the received information on the computer by repeated scanning of the spectrum in both directions of the voltage change. The resolution of our spectrometer is $R_{1/2} = 0,03\%$ on $B\rho$ under $\Omega = 0,07\%$ from 4π.

2.2. THE PROGRAMS USED FOR PROCESSING OF CONVERSION SPECTRA

The rigid requirements to accuracy of transition energy definition can not be satisfied without the high-quality mathematical processing of the results of measurements. To process internal conversion electron spectra, special programs based on the method of fitting the "instrumental" peak into the spectrum region of interest [5] were constructed. This technique implies high-statistical-accuracy measuring a single conversion peak from the obtained spectrum or, if such a peak is absent, specially measured single conversion peak with the shape similar to that of the line in the studied region of spectrum. After subtraction of the background, it is described by the multiple cubic-spline interpolation, and it is used as "instrumental", i.e. defines the experimental peak shape for the subsequent analysis by the least-squares method.

Assuming that all peaks have the same shape, the region of conversion spectrum is described by

$$\overline{Y}(X) = \sum_{i=1}^{n} A_i F(B_i + X) + C + DX + EX^2 , \qquad (1)$$

where $\overline{Y}(X)$ is count rate, provided that the voltage on the source is X;

n is the number of peaks in the region;

A_i is the ratio of the intensities of the i-th conversion peak and the "instrumental" peak;

B_i is the distance along X-axis between positions of the i-th conversion peak and the "instrumental" peak; if distances between some peaks are known with high accuracy, then corresponding parameters B_i can be set initially and they need not be changed during fitting;

$F(X)$ describes "instrumental" peak in the table form;

C, D, E are the parameters describing constant, linear, and quadratic background, correspondingly. Parameters C, D, E are set initially, and they are changing during fitting. Typically, in short spectrum regions $D=E=0$. In some cases background can be described by the dependence of the more complicated form than quadratic parabola.

It is not always possible to measure the tabular line at the same field as the spectrum section of interest. Therefore, if conversion spectrum is measured at the magnetic field corresponding to the NMR frequency f_1, and the line, which is used as the tabular one, is measured at the frequency f_2; then, according to the known dependence of the line shape on the magnitude of magnetic field, the section of spectrum is described by the following expression:

$$\overline{Y}(X) = \sum_{i=1}^{n} A_i F[B_i + d_i + (X - d_i)/\alpha_{21}] + \\ + C + DX + EX^2 \qquad (2)$$

where d_i is the distance from the coordinate origin to the i-th peak position;

$\alpha_{21} = f_1/f_2 (B\rho)'_2/(B\rho)'_1$ defines transformation of the "instrumental" peak in the spectrum. Here $(B\rho)'_2/(B\rho)'_1$ is the ratio of the magnetic rigidity derivatives with respect to energy.

Often the leading tail of the "instrumental" peak is imposed by other peaks of the spectrum. In such a case, to determine the tail of the "instrumental" peak, we extrapolate it linearly or quadratically; the parameters of extrapolation are also determined during common fitting.

Initial approximate values of all the parameters, along with their increments, are entered manually from the keyboard. Final values are obtained as a result of χ^2-value minimization:

$$\chi^2 = \sum_{i=1}^{N} \left(\frac{Y_i - \overline{Y}_i}{\sigma_i} \right)^2 , \qquad (3)$$

where Y_i is the observed count rate in the i-th point;

σ_i is its uncertainty;

N is the number of points in the analyzed spectrum region.

Standard deviations for A_i parameters are determined by the formula:

$$\Delta A_i = (\widetilde{A}_{kk} \varepsilon^2)^{1/2}, k = 1, 2, ..., n, \qquad (4)$$

where $\varepsilon^2 = \chi^2/(N-m)$ is reduced χ^2-value per 1 degree of freedom;

N is the number of points in the analyzed spectrum region;

m is the number of varying parameters;

n is the number of conversion peaks in analyzed region;

$\widetilde{A}_{kk}$ are the elements of inverse matrix in the solution of the set of linear equations.

Since, in fact, we use the experimental peak as the "instrumental" one, at a good quality of fit the ε^2-value is about 2.

Deviations of all the fitting parameters α_i can also be obtained using parabolic dependence $\chi^2(\alpha_i)$ around minimized value χ^2_{min}. Other parameters are fixed and they correspond to the optimized values. Standard deviations $\Delta\alpha_i$ are determined using the relation:

$$\chi^2(\alpha_i^{opt} \pm \Delta\alpha_i) = \chi^2_{min} + 1 \qquad (5)$$

where α_i^{opt} is the optimized value of α_i parameter which minimizes χ^2-value.

Precision measurements require careful analyzing and excluding all possible errors of the experiment. In our method of measurements of conversion spectra the scanning of spectra is carried out by the electrical field at a constant magnitude of the magnetic field. The scanning of the spectrum in a direct and return direction allowed us to eliminate possible errors related to relaxations of the magnetic field at a traditional way of spectra recording N(B).

The voltage divider determining the energy of electrons was collected from exact resistances that do not allow the errors to be more than 0,5% per channel. The resistances were selected in such a way that their deviations from the real value were mutually compensated.

Possible deviations due to the change of temperature, pressure, humidity and other changes of external conditions are averaged in the following way. The results are determined from several measurements of conversion spectra. Each measurement consists of the large number of short series ($\Delta t \leq 1$ hour), sometimes reaching up to 100. Each set of measurements is recorded in the computer, and then it is processed. The measurements are compared with each other to reveal whether the weight errors are compatible with the statistical straggling errors. The large mismatches were not observed. It indicates that external conditions have insignificant influence on the results of the experiment.

We did not observed calibration errors that could not be avoided by introduction of the appropriate corrections at the determination of the relative position of conversion lines.

The special attention was paid to the value of uncontrollable systematic inaccuracy of measurements at determination of the relative position of conversion lines. To estimate its value, we carried out special measurements, in which we have precisely determined the relative positions of conversion lines $L_1 52 - L_1 46$ in the decay of ^{183}Ta; and after that we compared it to the difference of energies of the corresponding γ-rays. The fragment of internal conversion spectrum is given in Fig. 1.

The absolute value of the energies of $\gamma 46$ and $\gamma 52$ have been measured earlier with high accuracy (about 0,2 eV) on the crystal-diffraction spectrometers [6]. In the measurements of conversion spectra we achieved such accuracy, that the corresponding statistical error in definition of the relative positions of conversion lines was small (about 0,25 eV). In this case the discrepancy of differences of transition energies obtained in our measurements and in the measurements with γ-rays should be explained mainly by the systematic inaccuracy of the experiment. This inaccuracy is 0,7 eV at the relative distance between lines of 6111 eV. At higher energy of conversion electrons and with decreasing the distance between lines, the systematic inaccuracy should be even less.
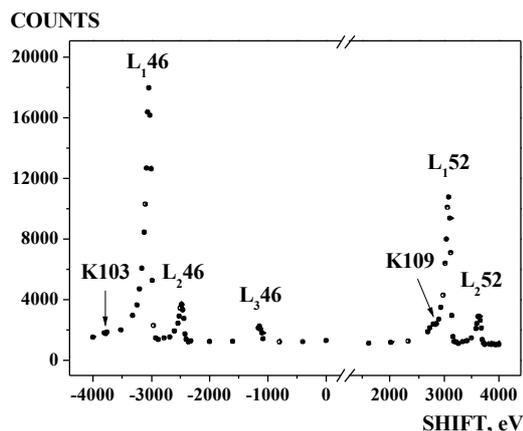

Fig. 1. *The L-subshell conversion electron spectrum of the $\gamma 46$ and $\gamma 52$ keV transitions in ^{183}W*

3. MEASUREMENT OF ENERGY OF GAMMA-TRANSITIONS IN THE DECAY OF ^{183,184,184m}RE

The possibilities of our technique completely revealed themselves in the precision energy measurements of some transitions in the decay of rhenium. The sources ^{183,184,184m}Re were received on cyclotron in reactions (d,n) and (d,2n) by means of irradiating the wolfram foil with the natural contents of isotopes by deuterons with energy 13,6 MeV. After irradiation the Re-fraction was separated by the radiochemical method, and it was electrodeposited on platinum substrates of the size 0,4×20 mm^2.

On high-resolution magnetic β-spectrometer the separate sections of the internal conversion electron spectrum were measured. These sections were selected in such way that one of the lines belongs to the decay of ^{183}Re, in which the energies of γ-rays are known with high accuracy, while the other line corresponds to the decay of ^{184,184m}Re, for which such measurements have not been performed. Energies of the transitions belonging to the decay of ^{183}Re have been measured with high accuracy on the crystal-diffraction spectrometer in the decay ^{183}Ta [6]. As the binding energies of electrons in atom are known with high accuracy (0,3…0,4 eV [7, 8]), exact value of the energy difference for corresponding γ-rays, as well as absolute magnitudes of these energies, can be determined using the energy difference for conversion lines.

Gamma-ray energy is determined from the following relation:

$$E_j - E_i = \varepsilon_j - \varepsilon_i + \Delta , \qquad (6)$$

where E_j and E_i are the energies of the γ-ray under consideration and the standard one;

ε_j and ε_i are their binding energies on corresponding atomic shells;

Δ is the measured energy difference for conversion lines.

Energy of the standard γ-ray E_i, eV	Measured energy difference for conversion lines Δ	Values of energy difference Δ, eV	Energy of the studied γ-ray E_j, eV	Location of γ-transitions in the decay scheme
46485,01(20)	Δ(M₁46 – L₁56)	486,2(6)	55279,0(8)	1501, keV, 7⁻ → 446 keV, 6⁻ ¹⁸⁴W
52596,48(18)	Δ(L₂63 – N₁52)	143,7(13)	63689,0(14)	1285, keV, 5⁻ → 1221 keV, 3⁻ ¹⁸⁴W
99081,82(27)	Δ(L₂99 – M₃83)	6598,4(8)	83306,7(8)	188, keV, 8⁺ → 104 keV, 4⁻ ¹⁸⁴Re
99081,82(27)	Δ(K104 – K99)	3506,3(13)	104739,5(14)	104, keV, 4⁻ → 0 keV, 3⁻ ¹⁸⁴Re
107933,7(3)	Δ(K111 – K107)	3283,7(3)	111217,4(4)	111, keV, 2⁺ → 0 keV, 0⁺ ¹⁸⁴W
107933,7(3)	Δ(L₂111 – L₁107)	3839,3(10)	111217,2(11)	111, keV, 2⁺ → 0 keV, 0⁺ ¹⁸⁴W

Note: statistical errors of measurements are listed

Corrections related to the energy of the recoil nucleus are small for our γ-ray energy region, and hence they can be neglected.

Energy values E_i of those γ-rays which were used as the standard ones, energy difference Δ of the measured conversion lines, and obtained energies E_j of γ-rays under study are listed in the table. As the standards we used the values of transition energies that were recommended in [1] as the third-order standards.

As seen from the table, the energy of the transition 111, keV, $2^+ \to 0$ keV, $0^{+\ 184}$W was determined by measuring energy difference of two different pairs of conversion lines. Results agree well within listed errors. This is a direct confirmation of the conclusion that systematic errors are small and do not exceed 1 eV according to our estimations.

As an example, some other results of our precision measurements can be given. Fragments of γ-spectrum and spectrum of internal conversion electrons for the same transitions from the decay of ¹⁸¹Hf that were measured on the semiconductor and magnetic spectrometer, correspondingly, are shown in Fig. 2 and Fig. 3.
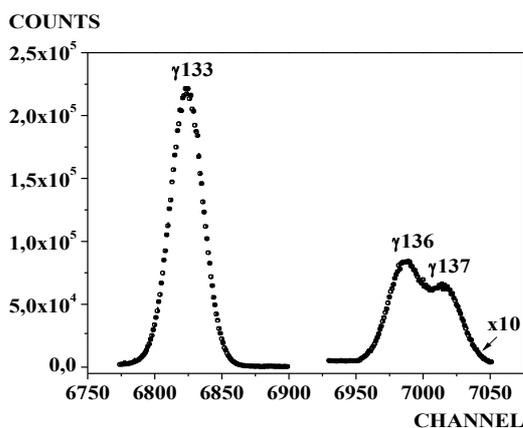

Fig. 2. The γ-ray spectrum of ¹⁸¹Hf measured by using a 5 см³ HPGe-detector with resolution of 490 eV FWHM at γ122 keV ⁵⁷Co

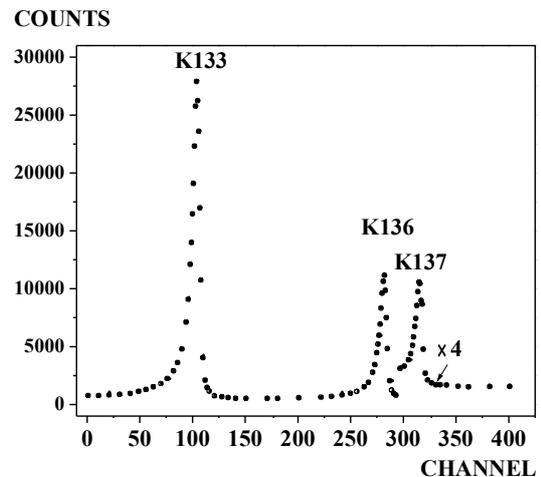*Fig. 3. The conversion electron spectrum for ¹⁸¹Hf measured by using an iron yoke double-focusing magnetic β-spectrometer with a momentum resolution of $R_{1/2} = 0,04$ %*

Higher resolution of β-spectrometer allows us not only to resolve studied transitions in the spectrum, but also to determine their energy difference with the accuracy about 0,3 eV [9].

4. CONCLUSIONS

The proposed technique allows one to reach accuracy of determination of energy transitions from 0,2 up to 1 eV for the energy region from 100 keV up to 500 keV. That completely corresponds to the requirements to the 4-th-order standards.

If we investigate transitions of weak intensity or in the case when it is not possible to produce radioactive sources of high specific activity, the proposed method of transition energy determination is more preferable that one which makes use of crystal-diffraction spectrometers.

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ОПРЕДЕЛЕНИЕ ЭНЕРГЕТИЧЕСКИХ НОРМАЛЕЙ МЕТОДАМИ ПРЕЦИЗИОННОЙ БЕТА-СПЕКТРОСКОПИИ

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Разработана методика, позволяющая с высокой точностью определять энергии переходов в атомных ядрах. Методика основана на измерении разности энергии линий электронов внутренней конверсии на β-спектрометре высокого разрешения типа $\pi\sqrt{2}$ с железным ярмом и радиусом равновесной орбиты ρ = 50 см при постоянном магнитном поле спектрометра. Точность определения энергии переходов достигает от 0,2 до 1 эВ для области энергии от 100 до 500 кэВ, что полностью соответствует требованиям, предъявляемым к энергетическим нормалям 4-го порядка.

ВИЗНАЧЕННЯ ЕНЕРГЕТИЧНИХ НОРМАЛЕЙ МЕТОДАМИ ПРЕЦИЗІЙНОЇ БЕТА-СПЕКТРОСКОПІЇ

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Розроблено методику, яка дозволяє з високою точністю визначати енергії переходів в атомних ядрах. Методика основана на вимірах енергії електронів внутрішньої конверсії на β-спектрометрі високої роздільної здатності типу $\pi\sqrt{2}$ з залізним ярмом і радіусом рівноважної орбіти ρ = 50 см при постійному магнітному полі спектрометра. Точність визначення енергії переходів сягає від 0,2 до 1 еВ для області енергії від 100 до 500 кеВ, що повністю відповідає вимогам до енергетичних нормалей 4-го порядку.