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= ЯДЕРНА ФІЗИКА =

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ISOMER YIELD RATIOS OF ¹³³Te, ¹³⁴I, ¹³⁵Xe IN PHOTOFISSION OF ²³⁵U WITH 17 MeV BREMSSTRAHLUNG

The isomeric yield ratios of ¹³³Te, ¹³⁴I, ¹³⁵Xe in the photofission of ²³⁵U with bremsstrahlung with the end-point energy $E_e = 17$ MeV are measured. The contributions to the isomeric yield ratios produced by β -decay from nuclei of parent isobaric chain are taken into account. The distribution and average value of angular momentum of the primary fission fragments are determined by the statistical model analysis. The codes EMPIRE 3.2 and TALYS 1.4 are applied for calculations of gamma-transitions to the isomeric and ground states. Dependence of determined distributions and average values of angular momentum on shapes of the radiative strength functions and the nuclear level densities are studied.

Keywords: isomeric yield ratios, photofission, statistical model analysis, average angular momentum.

Introduction

Study of the isomeric yield ratios of the populations of the isomeric and ground states of primary fission fragments can provide information (distribution, average value) on the angular momentum in primary fission fragments [1 - 11]. It can help to consider the scission configuration and the mechanism of the spin generation in the fissioning nuclei [1, 12 - 15].

The unstable nuclei with $A \sim 90$, $Z \sim 40$ and $A \sim 132$, $Z \sim 50$ are located close to magic numbers and have rather long lifetimes. Because of this, they are often used in the isomeric ratio measurements [2 - 6].

The isomeric yield ratios of ¹³⁴I and ¹³³Te were determined in the photofission of ²³⁵U with the bremsstrahlung with the end-point energies $E_e = 12$, 15, 20 and 30 MeV in Refs. [4] and [5] respectively. The radiochemical separation techniques and direct spectrometry of irradiated uranium foils were used in these experiments.

The isomeric yield ratios of ¹³³Te, ¹³⁴I, ¹³⁵Xe in the photofission of ²³⁵U with 9.6 MeV bremsstrahlung were measured within the same experiments in Ref. [10]. Contrary to the Refs. [4, 5] the direct gamma-spectrometry of the studied isotopes is used. We present the results of the measurements by this method of the isomeric yield ratios of ¹³³Te, ¹³⁴I, ¹³⁵Xe in the photofission of ²³⁵U with 17 MeV bremsstrahlung in this contribution. The sensitive semiconductor HPGe detectors are used. It provides measurements with the high energy resolution with resulting good separation of the gamma transition peaks. It allows to perform the foolproof identification of isotopes in any time period without the radiochemical separation techniques, which requires the rather large time periods for its realization. As a result, the contributions to the isomeric yield ratios for given nuclide produced by β -decay from nuclei of parent isobaric chain are removed [9 - 11]. It is achieved by considering the implicit solutions of the differential equation of radioactive decay and growth at selected times of uranium sample irradiation, of cooling and of measurements with the use of information on decay characteristics of precursor nuclei of parent isobaric chains.

The distributions and the average values of angular momentum of the primary fragments ¹³³Te, ¹³⁴I and ¹³⁵Xe are also determined by the statistical model analysis. The codes EMPIRE 3.2 [16] and TALYS 1.4 [17] are applied to calculations of gamma-transitions to the isomer and ground states. The effect of using different expressions for the radiative strength functions and the nuclear level densities on the extracted values of the average angular momentum is studied.

Experimental method and results

The isomeric yield ratios are measured by irradiation of uranium sample with further identification of the radioactive products. Uranium sample with enrichment 90 % of 235 U isotope (238 U - 10 %) is used. The sample has a weight close to 0.5 gr. The measurement of the isomeric yield ratios is performed by using the bremsstrahlung induced by electron beam from the M-30 microtron in the Laboratory of Photonuclear Reactions of the Institute of Electron Physics, National Academy of Sciences

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of Ukraine, Uzhgorod. A cooled tantalum disk (2-mm thick) is used as the bremsstrahlung producing target after hits of the electron beam. The endpoint energy of the bremsstrahlung is equal to 17 MeV. The mean electron current is around 3μ A. The distance from the bremsstrahlung producing target to the uranium sample equals to 9 cm.

After irradiation, the uranium sample is transferred to a separate laboratory where spectroscopic system is located. The γ -spectra of the photoreaction products are measured by the system of HPGe detectors of CANBERRA and ORTEC with the amplifier 2024 and the multichannel analyzer 8192, which are connected to computer for the data-processing. The energy resolution of the detectors is 2 keV for the 1332 keV γ -line of ⁶⁰Co, and detection efficiency is 30 % in comparison to the efficiency for (3 in.×3 in.) NaI(Tl)-detector.

Typical experimental spectra of ²³⁵U photofission products are shown in Fig. 1.



Fig. 1. Typical total gamma-spectra of ²³⁵U photofission products after different time periods: a - t = 30 min; b - t = 40 min. Gamma-lines of identification of isotopes ¹³³Te ($T_{1/2}(^{133m}Te) = 55.4$ min) and ¹³⁵Xe ($T_{1/2}(^{135m}Xe) = 15.29$ min) are indicated.

The experimental spectra are analyzed by the Winspectrum code [18]. This code permits to analyze the total spectra in different time periods. It allows the identification of the isotopes by the γ -ray energies and the lifetimes. The experimental values of the isomeric yield ratios are determined with removing the contributions to populations of ¹³³Te, ¹³⁴I and ¹³⁵Xe states from β -decay of nuclei of parent isobaric chains (see,

for example, Ref. [9] for details).

The measured isomeric yield ratios $R_{exp} = Y_m / Y_g$ are presented in Table 1 with $Y_m (Y_g)$ for the fission product yields for isomer (ground) state. The numbers in parentheses are the uncertainties (one-sigma standard deviation) of the isomeric ratio values.

Table 1. Isomeric yield ratios R_{exp} of photofission fragments of ²³⁵U with the bremsstrahlung with different E_e . In first column, spin and parities of the isomer and ground states of nuclide are indicated

	R _{exp}					
Isomeric pair	12.0 MeV	15.0 MeV	20.0 MeV	30.0 MeV	9.6 MeV	17.0 MeV
	[4] - I; [5] - Te	[10]	[this work]			
$^{133\mathrm{m,g}}\mathrm{Te}\left[\frac{11}{2}^{-};\frac{3}{2}^{-}\right]$	1.8(0.5)	1.7(0.4)	2.3(0.9)	2.0(0.8)	2.3(0.3)	2.8(0.7)
$^{134m,g}I\left[8^{-};4^{-}\right]$	1.04(0.38)	1.17(0.19)	1.13(0.14)	0.96(0.11)	0.49(0.05)	2.8(0.6)
$135m,gXe\left[\frac{11}{2};\frac{3}{2}\right]$	_	_	_	_	0.14(0.01)	0.15(0.03)

The experimental values of the isomeric yield ratios for the irradiation of 235 U by bremsstrahlung with different the end-point energies [4, 5, 10] are also presented in Table 1. For clear comparison, the data for ^{133}Te and ^{134}I are shown in Fig. 2 too.



Fig. 2. Isomeric yield ratios R_{exp} of the primary fragments ¹³³Te and ¹³⁴I in photofission of ²³⁵U with the bremsstrahlung with different the end-point energy E_e .

As it can be seen in Table 1 and Fig. 2, our data for ¹³³Te are similar to the one at 9.6 MeV and those of Ref. [5]. For ¹³⁴I, there is a rather big difference between the data for bremsstrahlung with the endpoint energies 17 and 9.6 MeV and also with data of Ref. [4]. The difference of isomeric yield ratios at $E_e = 17$ and 9.6 MeV is probably resulted from dependence of R_{exp} on the excitation energy of fissioning nucleus, because the additional fission channel (γ , *nf*) opens for the bremsstrahlung at 17 MeV the end-point energy. It should be also noted that we have not used radiochemical procedures for initial separation of isotopes of ¹³⁴I.

Calculations of average angular momentum of primary fission fragments

The experimental values $R_{exp} = Y_m / Y_g$ are used to determine the angular momentum distribution P(J) of fixed shapes and average angular momentum \overline{J} of primary fission fragments [1, 6, 19, 20] (in units of \hbar)

$$\overline{J} = \sum_{J} JP(J) / \sum_{j} P(j).$$
⁽¹⁾

The following expressions for the angular momentum distributions are taken [1, 6, 19, 20]

$$P(J) = P(J,\lambda) = (2J+1)\exp(-J(J+1)/2B^2 - \lambda J),$$
(2)

and

$$P(J) = P(J,\mu) = (2J+1)\exp(-J(J+1)/2(B+\mu)^2),$$
(3)

where *B* is the spin cut-off parameter. Note that the spin distribution (3) has the similar functional form as that for the spin distribution of the nuclear level density. The parameters λ , μ are found from fitting values of the calculated isomeric ratios to $R_{exp} = Y_m / Y_g$. The parameter *B* will be defined below.

The theoretical values of the isomeric ratios are determined by the statistical model analysis using some statements of the Huizenga - Vandenbosh method [1, 19, 20]. Specifically, we adopt the following assumptions: 1) the gamma-cascades populate the ground- and isomer-states of a given nuclide just after neutron emission of primary fragments when the intrinsic (thermal) excitation energy of the nuclide is lower than the neutron separation energy S_n ; 2) all neutrons are evaporated with zero orbital momentum; 3) the excitation energies of states before γ -decay are shared with some distribution function ϕ , because given nuclide can be formed after neutron emission with different neutron multiplicity. As a result, the theoretical value of the isomeric yield ratio is described by the formula

$$R_{th} = \sum_{J,\pi} \int dU \cdot \varphi(U) \cdot P(J,\chi) \cdot \sigma_m(U,J,\pi) / \sum_{j,\pi} \int dU \cdot \varphi(U) \cdot P(j,\chi) \cdot \sigma_g(U,j,\pi).$$
(4)

Here, $\sigma_m(U,J,\pi)$ ($\sigma_g(U,J,\pi)$) is the probability of population by the gamma-decay of the isomeric (ground) state from the states with the excitation energy *U*, the spin *J* and the parity $\pi = \pm 1$. We use the step function for the energy distribution function $\varphi(U)$ with non-zero value within the excitation

 $\Delta U = U_{\min} \div U_{\max}$ energy interval with $U_{\min} = 0.5S_n + n\Delta_0 + E_{rot}(J)$ and $U_{\max} = S_n + n\Delta_0 + ,$ $+E_{rot}(J)$, where $n\Delta_0$ is the pairing energy correction with $\Delta_0 = 12 / \sqrt{A}$ and n = 2,1,0 for even-even, odd, odd-odd nuclei. The rotational energy $E_{rot}(J)$ is taken in the approximation of spherical nuclei $E_{rot}(J) = J(J+1)/(2F)$ with the moment of inertia (in units of $\hbar^2 \text{MeV}^{-1}$) $F = 0.0194 A^{5/3}$ ([25], p. 96). Note that the value of spin cut-off parameter B in Eqs. (2), (3) is calculated by the expression of the Fermi gas model in spherical atomic nuclei $B^2 = F \cdot T$ (in units of \hbar^2) with the temperature $T = \sqrt{U} / a$ at the average excitation energy $\overline{U} = (U_{\min} + U_{\max})/2$ and the nuclear level density parameter is $a = A/10 \text{ MeV}^{-1}$.

Using the angular momentum conservation law, the summations over spin J in Eqs. (1), (4) (and below) are performed on integer values of $J \ge 0$ for nuclei with integer values of the ground state spins $\binom{134}{53}I$ and on the half-integer values of $J \ge 1/2$ for nuclei with the half-integer values of the ground state spins $\binom{133}{52}$ Te, $\binom{135}{54}$ Xe).

The population probabilities $\sigma_g(U,J,\pi)$, $\sigma_m(U,J,\pi)$ are calculated by the codes of the EMPIRE 3.2 [16] and TALYS 1.4 [17]. Different models for the radiative strength functions (RSF) and for the nuclear level densities (NLD) are used in the calculations. Namely, we take the models of Standard Lorentzian (SLO), Enhanced Generalized Lorentzian (EGLO), Modified Lorentzian (MLO1) for the RSF (see [16, 17, 25, 26] for details). For the NLD, microscopic combinatorial level densities of the Hartree -Fock - Bogoliubov method (HFB), the Gilbert - Cameron model (GC) and the Empire Global Specific Model (EGSM) are used [16, 17, 25].

The parameters λ , μ of the angular momentum distributions (2), (3) of primary fission fragments $^{133}_{52}$ Te, $^{134}_{53}$ I, $^{135}_{54}$ Xe are presented in Table 2. The values of the parameters are found from χ^2 - fitting the theoretical isomeric ratios (4) to their experimental data $R_{exp} = Y_m / Y_g$. The population probabilities are calculated by EMPIRE 3.2 code with the RSF within MLO1 approach. All other input parameters of the code are taken by default.

The values of the parameters λ , μ determined with the RSF of other shapes vary up to 10 %.

The normalized angular momentum distributions $p(J) = P(J) / \sum_{j} P(j)$ of primary fission fragments ¹³³Te, ¹³⁴I and ¹³⁵Xe calculated by Eqs. (2) and (3)

with parameters from Table 2 are shown in Fig. 3. Note that the distributions p(J) have discrete values – integer for ¹³⁴I and half-integer for ¹³³Te, ¹³⁵Xe, and the lines in the Fig. 3 are drawn for better representability. The curves of p(J) with $\lambda = \mu = 0$ correspond to the spin distributions of the NLD within of the Fermi gas model in spherical compound nuclei.

Table 2. Values of parameters of the angular momentum distributions of primary fission fragments ¹³³₅₇Te , ¹³⁴₅₄I , ¹³⁵₅₄Xe

I	Parameter values				
Isomeric pair	В	λ	μ		
^{133m,g} Te	6.21	0.11(0.04)	-1.61(0.48)		
^{134m,g} I	6.32	-0.09 (0.04)	3.4(1.9)		
^{135m,g} Xe	6.41	0.77(0.08)	-4.63(0.14)		

As it can be seen in Table 2 and Fig. 3, the values of the parameters of λ and μ for ¹³³Te are not so big in comparison with *B* and the shapes of angular momentum distributions (2), (3) agree rather closely with that of the spin distribution of the nuclear level density in a compound nucleus. For fission fragments ¹³⁴I and ¹³⁵Xe, the parameters λ , μ have larger values. This indicates on an importance of the non-equilibrium processes in formation of the fission products ¹³⁴I, ¹³⁵Xe.

The mean values and uncertainties of angular momentum of primary fission fragments $^{133}_{52}$ Te , $^{134}_{53}$ I, $^{135}_{54}$ Xe for the spin distributions given by Eqs. (2) and (3) with the population probabilities calculated by EMPIRE 3.2 are shown in Figs. 4 and 5. The results presented in Fig. 4 are derived with the population probabilities calculated with the RSF within SLO, EGLO and MLO1 approaches. The results presented in Fig. 5 are obtained with the population probabilities calculated using different models of the NLD (EGSM, GC, HFB). All other input parameters of the code are taken by default.

Results presented on panels *a* and *b* correspond to the angular momentum distribution given by Eqs. (2) and (3) respectively. For clear representation, the values \overline{J} are scattered for given nuclide and different model. Isotope ²³⁵U has spin-parity of $7/2^-$, therefore the absorption of dipole electric gamma-rays leads to compound nucleus ²³⁵U with spins and parities of $5/2^+$, $7/2^+$ and $9/2^+$. In Figs. 4 and 5 these spin values are marked by dashed lines.

We can see in Figs. 4 and 5 that there is no large difference of derived values of \overline{J} for various models of the radiative strength function and the nuclear level density. The mean values of \overline{J} agree within the uncertainties and their difference does not exceed ~ 8 %.



Fig. 4. Average angular momentum \overline{J} of primary fission fragments ¹³³Te, ¹³⁴I, ¹³⁵Xe calculated with population probabilities within the EMPIRE 3.2 code with different shapes of the RSF (SLO, EGLO, MLO1): a – results with the angular momentum distribution given by (2); b – results with (3). For clear representation, the values \overline{J} are scattered for given nuclide and RSF model. Dashed lines mark spins (5/2, 7/2, 9/2) of compound nucleus states of ²³⁵U formed after dipole gamma-ray absorption.

There is difference between average angular momentum \overline{J} of primary fission fragments $^{133}_{52}$ Te, $^{134}_{53}\mathrm{I}$, $^{135}_{54}\mathrm{Xe}$ and the spins of compound nucleus states of ²³⁵U formed just after dipole gamma-ray

absorption, see Figs. 4 and 5. It indicates on the presence of some mechanism during the fission process which generates angular momentum in the primary fragments (specifically in $^{135}_{\ 54} Xe$).

A



Fig. 5. Average angular momentum \overline{J} of primary fission fragments ¹³³Te, ¹³⁴I, ¹³⁵Xe calculated with population probabilities within the EMPIRE 3.2 code with different models of the NLD (EGSM, GC, HFB): *a* – results with the angular momentum distribution given by (2); *b* – results with (3). For clear representation, the values \overline{J} are scattered for given nuclide and NLD model. Dashed lines mark spins (5/2, 7/2, 9/2) of compound nucleus states of ²³⁵U formed after dipole gamma-ray absorption.



Fig. 6. Comparison of calculated root mean square angular momentum $J_{rms} = (\overline{J^2})^{1/2}$ of primary fission fragments of ¹³³Te (*a*) and ¹³⁴I (*b*) with results of Refs. [4, 5, 10] at different end-point energies E_e . For clear representation, the values J_{rms} are scattered for given E_e . Angular momentum distribution is taken by Eq. (3) and input models and parameters by default are used in the EMPIRE 3.2 and TALYS 1.4 codes.

The values of root mean square angular momentum $J_{rms} = (\overline{J^2})^{1/2}$ of primary fission fragments of ¹³³Te and ¹³⁴I at different end-point energies E_e are presented in Fig. 6. The root mean square angular momentums are calculated by equation (4) using data for R_{exp} at $E_e = 17$ MeV and data from Refs. [4, 5, 10]. The population probabilities are calculated within the EMPIRE 3.2 and TALYS 1.4 codes with the default input models and parameters for the RSF and the NLD. Eq. (3) is used for the angular momentum distribution. The results of calculations J_{rms} from Refs. [4, 5, 10] are also indi-

cated on the Fig. 6. The value J_{rms} from Ref. [10] is recalculated from the \overline{J} using the relationship $J_{rms} = 2\overline{J} / \sqrt{\pi}$.

The results of calculations with the population probabilities within the codes of EMPIRE 3.2 and TALYS 1.4 correlate well. The values of J_{rms} recalculated for data [4, 5] by Eq. (4) are in agreement with the ones from Refs. [4, 5]. This indicates on the reliability of the simplified statistical method (4) for calculation of the isomeric yield ratios in the primary fission fragments and thereby for determination of the angular momentum distribution and values \overline{J} , J_{rms} .

Conclusions

The isomeric yield ratios of fission fragments 133 Te, 134 I, 135 Xe are measured in the photofission reactions (γ , f), (γ , nf) of 235 U with the bremsstrahlung with the end-point energy 17 MeV. The experimental values of isomeric yield ratios of the primary fission fragments are determined with removing the contributions produced by β -decay from nuclei of parent isobaric chain.

The average values and the distributions of angular momentum of the primary fragments ¹³³Te, ¹³⁴I and ¹³⁵Xe are determined by the statistical model analysis. New simplified statistical method for calculation of the isomeric yield ratios and thereby for determination of the angular momentum distribution and the average angular momentum in the primary fission fragments are proposed and tested. In this method, the probabilities of populations by the

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gamma-decay of the ground and isomeric state are calculated by the codes of EMPIRE 3.2 [16] and TALYS 1.4 [17]. Note that all currently available experimental data related to the properties of the low-lying states (energies, spins, parities, probabilities of gamma-transitions and their multipolarities) from library of the RIPL [25] are used in these codes.

It is shown that the values of average angular momentum dependent weakly on models of the radiative strength function and the nuclear level density. The mean values of angular momentum agree within the uncertainties and their difference does not exceed ~ 8 %. The difference between average angular momentum of primary fission fragments and the spins of compound nucleus states of 235 U formed just after dipole gamma-ray absorption is demonstrated. It confirms the presence of a mechanism during the fission process, which generates the angular momentum in the final fragments.

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ІЗОМЕРНІ ВІДНОШЕННЯ ВИХОДІВ ¹³³Те, ¹³⁴І, ¹³⁵Хе ПРИ ФОТОПОДІЛІ ²³⁵U ГАЛЬМІВНИМ ВИПРОМІНЮВАННЯМ З МАКСИМАЛЬНОЮ ЕНЕРГІЄЮ 17 Мев

Виміряно ізомерні відношення виходів ¹³³Te, ¹³⁴I, ¹³⁵Xe при фотоподілі ²³⁵U гальмівним випромінюванням з максимальною енергією $E_e = 17$ MeB. Враховано внески до ізомерних відношень виходів ядер, утворених з β-розпаду ядер материнського ізобарного ланцюга. Використовуючи статистично-модельний аналіз, отримано розподіли та значення середніх кутових моментів первинних фрагментів поділу. При розрахунках у-переходів на ізомерний та основний стани використовувались коди EMPIRE 3.2 та TALYS 1.4. Досліджено вплив різних видів радіаційно-силових функцій та густин ядерних рівнів на значення середніх кутових моментів первинних фрагментів поділу.

Ключові слова: ізомерні відношення, фотоподіл, статистично-модельний аналіз, середні кутові моменти.

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ИЗОМЕРНЫЕ ОТНОШЕНИЯ ВЫХОДОВ ¹³³Те, ¹³⁴I, ¹³⁵Хе ПРИ ФОТОДЕЛЕНИИ ²³⁵U ТОРМОЗНЫМ ИЗЛУЧЕНИЕМ С МАКСИМАЛЬНОЙ ЭНЕРГИЕЙ 17 МэВ

Измерены изомерные отношения выходов ядер ¹³³Te, ¹³⁴I, ¹³⁵Xe при фотоделении ²³⁵U тормозным излучением с максимальной энергией $E_a = 17$ МэВ. Учтен вклад в изомерные отношения выходов ядер от β -распада ядер материнской изобарной цепочки. Используя статистично-модельный анализ, получены распределения и значения средних угловых моментов первичных фрагментов деления. В расчетах у-переходов на изомерное и основное состояния использовались коды EMPIRE 3.2 и TALYS 1.4. Исследовано влияние различных видов радиационно-силовых функций и плотностей ядерных уровней на значения средних угловых моментов первичных фрагментов деления.

Ключевые слова: изомерные отношения, фотоделение, статистично-модельный анализ, средние угловые моменты.

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