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# INDEPENDENT YIELDS OF Kr AND Xe ISOTOPES FOR THE PHOTOFISSION OF HEAVY NUCLEI

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#### **INTRODUCTION**

The distribution of fission fragments over mass numbers, A, and atomic numbers, Z, is a basic characteristic of nuclear fission. The form of those distributions is governed by the dynamics of the fission process between the saddle and scission points. This process is quite intricate; it depends on a variety of factors: the energy surface relief, scission nuclear shape, collective motion, nuclear viscosity etc. Therefore measuring isotopic (by A) and isobaric (by Z at a given A) fragment distributions is an important source of data on fission dynamics. Distributions of primary fragments (i.e. fragments produced immediately after the scission of the fissioning nucleus) are most informative.

However measuring those distributions involves certain difficulties. Each of the methods in use is of limited application and efficient only for fission fragments of a certain region. Therefore isotopic and isobaric distributions of primary fragments are adequately studied for the fission of Th, U and Pu induced by low-energy neutrons as well as for the spontaneous fission of <sup>252</sup>Cf (data on those distributions are reviewed in [1,2]).

At the same time, in order to understand the dynamics of the fission process, there is a need for a larger volume of experimental data and for new theoretical approaches to describe those data. One of the promising lines of investigation in this region of nuclear fission is measuring isotopic and isobaric distributions of fragments in reactions with y-quanta, which are characterised by a number of peculiarities:

- 1. Interaction of y-radiation with nuclei is entirely of an electromagnetic character, which is well-studied. Therefore this interaction can be calculated correctly without resorting to any models.
- 2. In the energy range  $10-16$  MeV, the interaction of y-quanta with nuclei is of a resonance character (giant dipole resonance). Its frequency corresponds to the frequency of proton oscillation relative to the neutrons in the nucleus.
- 3. There being no binding energy and Coulomb barrier allows fissioning nuclei to be produced of any, even the lowest excitation energy immediately upon y-quantum absorption.

4. Over a wide range of y-quantum energies, the angular momentum introduced into the nucleus remains practically unchanged (it is only 1*h* for dipole *y*-quantum absorption).

Those peculiarities of reactions with y-quanta enable new information about nuclear fission to be obtained. For example, experiments on nuclear photofission provided a large body of data on the fission barrier structure and the potential surface shape [3]. Those peculiarities of y-quantum-nucleus interaction may be expected to manifest themselves in fission fragment production as well. This can be illustrated by the unusual behavior of the y-quantum dependence of the yield of symmetric fission fragments  $(^{115}Cd, ^{117}Cd)$  (a maximum at 6 MeV, a bend at 5.3 MeV), which was observed in work [4].

However data on isotopic and especially isobaric fragment distributions for photofission (first of all, on independent distributions) are much sparser than those for neutron- or charged-particle-induced reactions. Mention may be only made of experiments carried out at Gent (Belgium) for some nuclei [5, 8].

This work aims to measure the independent yields of fragments (Kr and Xe isotopes) produced in the process of the  $232$ Th,  $238$ U and  $244$ Pu fission induced by y-quanta of the energy that corresponds to the giant dipole resonance and compare those yields to similar data for neutroninduced fission. The cumulative yields of those isotopes for the photofission of <sup>238</sup>U are reported in work [9]; preliminary data on the independent yields of Xe fragments for  $^{232}$ Th and  $^{238}$ U fission are given in our earlier work [10].

### **EXPERIMENTAL**

To separate Kr and Xe isotopes from a whole host of photofission fragments, the peculiarities are used of their chemical properties, which differ greatly from the properties of other elements and manifest themselves in a number of phenomena. One of them is the adsorption of atoms by a filter and the walls of a capillary through which fragments are transported from an irradiated target to radioactive radiation detectors. Kr and Xe inert gases are intensively adsorbed only at the temperature of liquid nitrogen  $(-210^{\circ}C)$ , whereas all the other fragments are absorbed at room temperature. The experimental set-up described was built around that peculiarity and included a reaction chamber, a cryostat and a teflon

capillaries for a gas flow (its schematic view is presented in Fig. 1; for a more detail description see work [11]).

Targets to be irradiated were positioned in the reaction chamber, which was a cylinder of inner diameter 40 mm and height 30 mm. The cylinder walls were 1.5 mm in thickness. To let buffer gas in and out, there were an inlet and an outlet, diametrically opposed. In experiments, one or two targets, positioned on the cylinder ends, were used. The characteristics (chemical composition, impurities, dimensions, layer and backing thickness) of the targets are tabulated in Table 1.

Inert gas was carried from a bottle to the reaction chamber through a polyethylene capillary 4 mm in diameter. The chamber gas pressure, measured with a manometer at the inlet, was adjustable with the bottle valve. The buffer gases were chemical pure He, Ar and  $N<sub>2</sub>$ , the pressure being 1-2 atm. The choice of an inert gas and a pressure depended on experimental conditions (fragment collecting efficiency, fragment transportation speed, levels of y-radiation background due to inert gas activation by bremsstrahlung from the microtron).

Fission fragments were transported from the reaction chamber to the cryostat through a 10 m teflon capillary of inner diameter 2 mm by pumped buffer gas. There was a fibrous filter installed at the capillary entrance. All the fission fragments except Kr and Xe were intensively adsorbed on that filter. A comparison of the y-radiation of fragments in the target, at the filter and in the cryostat showed that more than 90% of fragments that are not related with Kr and Xe by decay chains were adsobed by the filter and more than 50% of Kr and Xe fragments are stopped in cryostat.

The cryostat was a copper tube in the form of a spiral placed in a vacuum flask filled with liquid nitrogen. The total length of the tube was 2 m, its inner diameter and wall thickness being respectively 2.5 mm and 0.5 mm. The cryostat of such a design allowed the temperature close to - 200°C to be maintained in it, which enabled the condensation of Kr and Xe, whose condensation temperatures at the atmosphere pressure are -157°C and -112°C respectively. So, Kr and Xe isotopes were efficiently and rapidly separated from the other fission fragments, which were stopped by the filter and by the capillary walls.

It took 0.5-1.0 s for Kr and Xe fragments to be transported from the reaction chamber to the cryostat, which was measured in special

 $\overline{\mathbf{4}}$ 

experiments at varied pressure in the chamber (the valve was closed and the pump was in operation). This time, which included the time for fragments to diffuse to the outlet and the time for fragments to be transported through the capillary, depended on the kind and pressure of the gas used [11]. This time define a lower limit for the half-life of fragments can be studied  $(-0.3 \text{ s})$ . Kr and Xe predecessor fragments of a half-life greater than the time that it took them to diffuse to the outlet (0.1-0.2) also adsorbed on the filter without contributing to the yield of Kr and Xe fragments that were brought to the cryostat. Therefore fragments brought to the cryostat were produced in the process of nuclear scission, and the yields of those fragments are considered to be independent. Fragments of other elements might find themselves in the cryostat only due to the  $\beta$ -decay of Kr and Xe isotopes; the yields of them must correspond to the independent yields of the inert gases discussed. It was the yields of those nuclides that were measured in experiment since they had longer half-lives and more convenient  $\gamma$ -lines. Short-lived fragments (predecessors of Kr and Xe), which decayed in the reaction chamber, had low independent yields and didn't affect the yields of Kr and Xe. Fig. 2 presents examples of B-decay chains that contain Kr and Xe isotopes; the nuclides that were used to determine yields are marked. The radioactive decay characteristics of those nuclides (half-lives,  $T_{1/2}$ , the energy E<sub>y</sub> and intensity I, of the measured  $\gamma$ -lines) are tabulated in Table 2 [12]. Such an approach to measuring independent yields allowed the measurement sensitivity to be increased and fragments to be observed whose yields were  $10^{-3}$  of a number of fission events.

#### **EXPERIMENTAL RESULTS**

To measure independent yields of Kr and Xe fragments, bremsstrahlung from the MT-25 microtron was used (Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research). For a description of that microtron and its main parameters see [13]. The energy of accelerated electrons (to which the ultimate energy of the bremsstrahlung spectrum corresponded) was 25 MeV, the beam current being 15  $\mu$ A. The converter was a water-cooled tungsten disc of 4 mm thick. There was an aluminium screen of 20 mm thick installed downstream of the converter to keep electrons out of the reaction chamber. The electron beam was elliptical in shape, the vertical and horizontal axes being 7 and 6 mm respectively.

5

Targets were typically irradiated for 30 min. Five minutes after irradiation the copper spiral was taken out of the reaction chamber and brought to a y-radiation spectrometer situated in a room protected against radiation from the microtron. In this time the Kr and Xe isotopes underwent B-decay.

From the y-radiation spectra measured on that spectrometer, the .areas were determined of the y-lines due to the nuclides of the daughter products of the P-decay of Kr and Xe isotopes. Those areas are related to the yields of the corresponding isotopes by the relationship:

$$
Y(A) = \frac{S(1+\alpha)Nf(t)}{tI_{\gamma}\varepsilon_1\varepsilon_2\varepsilon_3\eta},
$$
 (1)

where S is the y-line area on subtraction of the background; t the measurement time;  $\varepsilon_1$ ,  $\varepsilon_2$  and  $\varepsilon_3$  respectively the efficiencies of transportation through the capillary, adsorption in the cryostat and ydetection for Kr and Xe fragments; I, the relative  $\gamma$ -line intensity for the decay of the nuclide measured;  $\alpha$  the internal y-radiation conversion coefficient; f(t) the temporal factor, which takes account of the decay of Kr and Xe nuclei during transportation to the cryostat, the accumulation and decay of the nuclides measured; N the number of fissioning nuclei in the layer from which fission fragments escape; η the intensity of bremsstrahlung y-quanta that induce nuclear fission. Using this technique, measurement was for the first time carried out of the independent yields of 4 Kr isotopes  $(A=89-93)$  and 6 Xe isotopes  $(A=138-143)$  for the photofission of <sup>232</sup>Th, <sup>238</sup>U and<sup>244</sup>Pu

Since the Kr and Xe isotopes under study were produced in the same session of irradiation, and the measuring technique used was totally the same, the relative yields of those isotopes are obtainable from the measured  $\gamma$ -line areas taking account of the above parameters  $I_{\gamma}$ ,  $\alpha$ ,  $\varepsilon$ <sub>3</sub> and  $f(t)$  and without using the parameters  $\varepsilon_1$  and  $\varepsilon_2$  known with an inadequate accuracy. The yields thus obtained and normalised relative to <sup>91</sup>Kr and  $139$ Xe isotopes are tabulated in Table 3.

The absolute independent yields for those isotopes are obtainable by using reported data on their yields in reactions with  $\gamma$ -quanta [6-8, 14]

and neutrons of the fission spectrum and of the energy 14.7 MeV [2] (both reactions have similar isotopic distributions, and the yields are close in magnitude [15]). From those data the independent yields of <sup>91</sup>Kr and <sup>139</sup>Xe referred to the sum of the all the isobars with  $A=91$  and 139 (fractional yields) were adopted (they presented in the Table 3). The fractional independent yields of other measured Kr and Xe fragments are deduced from those magnitudes and are included also in Table 3. The mass-number dependence of the independent yields of Ke and Xe fragments referred to the total number of fission events is given in Fig. 3.

Due to bremsstrahlung having a continuous spectrum, those yields correspond to a certain range of excitation energy. That range is obtainable from a measured excitation function for the <sup>238</sup>U( $\gamma$ , f) [16] (it was supposed that the other fissioning nuclei have similar excitation functions) and a bremsstrahlung spectrum calculated for those conditions [17]. The average excitation energy and the distribution half-width proved to be 12.5 MeV and ~5 MeV respectively.

#### **DISCUSSION**

The mass-number dependencies of the independent photofission yields of Kr and Xe, presented at Fig. 3, are characterised by a maximum at a certain magnitude of A in whose vicinity a smooth drop is observed. Dependencies of this form are describable by a Gaussian distribution with the parameters:  $\overline{A}$  (average mass number),  $\sigma$  (dispersion)

$$
Y(A) = K \exp\left[-\frac{(A-A)^2}{2\sigma^2}\right],
$$
 (2)

where K is a normalising factor. The experimental mass number dependences are approximated by these distributions. They are presented on Fig.4 and, for comparison, the similar dependencies for the fission of  $^{238}$ U induced by neutrons of an energy of 14.7 MeV and for the fission of  $^{233}$ U and  $^{235}$ U induced by thermal neutrons are showed.

The parameters of these distributions for  $^{232}Th$ ,  $^{238}U$  and  $^{242}Pv$ photofission nuclear reactions are tabulated in Table 4 (the magnitude of the A for  $^{238}U$  is in good agreement with that obtained in work [7]). For comparison the similar parameters are presented for the fission of  $^{235}$ U,  $^{233}$ U and  $^{238}$ U by the neutrons [2]. A comparison of those parameters permits a number of conclusions to be drawn:

- 1. The average mass number for Kr and Xe fragments shows a slight increase as the Z and A of the fissioning nucleus increases. It is close to the magnitude of the A for the fission of  $^{238}$ U induced by 14.7 MeV neutrons, but substantially larger than that for the fission of  $^{235}$ U and  $^{233}$ U induced by thermal neutrons. This points to the fact that an increase in the neutron excess of the fissioning nucleus results in an increase in the A (this excess can be characterised by the ratio (N-Z)/N, which is tabulate in Table 4).
- 2. At the same time the distribution dispersion shows a noticeable increase as the atomic number of the fissioning nucleus increases (for example, it increases by a factor of 1.5 from  $232 \text{Th}$  to  $244 \text{Pu}$ ). This means that the yields of the most neutron-rich fragments differ significantly. For example, the yield of <sup>143</sup>Xe for the photofission of  $244$ Pu is one order of magnitude larger than for the photofission of  $232$ Th, this difference increasing rapidly as the number of neutrons in the fragment increases.
- 3. The deviation of the measured yields from the Gaussian distribution as well as their even-odd distinctions is not great and lies within the limits of experimental error.

The Kr and Xe fragments produced in the fission of  $^{232}$ Th are complementary, i.e. they are produced in the same fission event. A comparison of the sum of their average mass numbers ( $\overline{A}(Xe) = 138.9$ ) and  $\overline{A}(Kr) = 91.3$ ) with the mass number of the fissioning nucleus (A = 232) allows the number of neutrons emitted from those fragments to be determined. This number proves to be equal to  $v = 1.8(2)$ . With the fractional yields of <sup>91</sup>Kr and <sup>139</sup>Xe equal to 0.8 and 0.85 respectively, it is in good agreement with reported numbers of fission neutrons from fragments of a specified mass number and their excitation energy dependence [15, 18]. From those data it follows that in this region of fragment mass numbers the ratio of the numbers of neutrons from the light and heavy fragments is 1.3 and has only a weak dependence on excitation energy. This corresponds to  $v = 1.0$  for Kr isotopes and  $v = 0.8$ for Xe isotopes.

The magnitudes of v obtained allow one to determine the fragment charge shift relative to the unchanged charge distribution  $Z_0/A_0$ , which corresponds to the ratio of the atomic to mass number of the fissioning nucleus. For a fragment of atomic number Z, the charge shift is expressed as

$$
\Delta Z = \frac{Z_0}{A_0} (A + v) - Z. \tag{3}
$$

The thus obtained magnitudes of the  $\Delta Z$  for the average mass numbers of Kr and Xe fragments produced in the photofission of  $^{232}$ Th,  $^{238}$ U and  $^{244}$ Pu are tabulated in Table 5. They prove to be close to the corresponding magnitudes of the  $\Delta Z$  for the neutron-induced fission of heavy nuclei [1, 19].

To describe isotopic and isobaric fission fragment distributions, a number of models are used. In the last years, the diffusion model has gained wide acceptance [20, 21]. This model assumes the break-up of the nucleus to be a quasi-equilibrium process from the charge mode standpoint, which allows a statistical approach to be used to describe the charge distribution. The main mechanism that governs fragment charge isobaric distributions is associated with the collective motion of nucleons, which manifests itself as dipole isovector oscillations. However the oneparticle motion of nucleons is likely to make a certain contribution. The characteristic time for both kinds of motion  $(-10^{-22} s)$  is far less than the time it takes for the nucleus to descent from the saddle point to the scission point ( $\sim$ 3·10<sup>-20</sup> s). To find the charge of a fragment, the minimum potential energy criterion is used. The deformation dependence of potential energy was calculated by the liquid drop model, taking account of shell corrections. Calculations of isobaric charge distributions for nuclear fission in the region of Th - Pu carried out by that model showed that the charge shift relative to the unchanged charge distribution is 0.2 - 0.4 [22]. Those magnitudes of the  $\Delta Z$  correspond to experimental data obtained in our work on photofission as well as in other works on the fission induced by various particles.

Experiments showed that the isotopic photofission fragment distributions for heavy nuclei are close to the similar distributions for neutron-induced fission. Obviously the dipole oscillations of the electric charge of nuclei that are induced during the absorption of y-quanta are damped out by the moment when fragments are formed. The other peculiarities of the interaction of y-radiation with nuclei that have been

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mentioned above are likely to affect fragment production at lower excitation energies (in the vicinity of and below the fission barrier).

The results of this work allow the conclusion to be drawn that the nuclear fission induced by y-radiation is an efficient way for producing the most neutron-rich nuclides. That the fraction of those nuclides is large is due to the following: the fragments produced have low excitation energy (therefore the number of neutrons emitted from them is small);  $\gamma$ radiation has high penetrability, which enable large amounts of fissionable matter to be used; the isotopic fragment distribution dispersion is quite wide. So photofission is a promising way both for research on the nuclear structure of fragments and for producing intense beams of the accelerated neutron-rich radioactive nuclei. The study of the reactions induced by these nuclei is very interesting, since it allows to get the principle new information about nuclear structure. For example, the unusual wide space distributions of weakly bound neutrons in some nuclei (neutron halo) were observed in such kind reactions. Lot of project for the acceleration of neutron-rich nuclei are developed now. One of these is the project DRIBs in Laboratory of Nuclear Reactions JINR [23]. The fission fragments are produced at the irradiation of thick uranium target by the bremsstrahlung of microrron and after the mass-separation are accelerated on the 4-meter isochronous cyclotron U400.

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## **TABLE 1 CHARACTERISTICS OF THE TARGETS**



TABLE 2 **CHARACTERISTICS OF THE STUDIED NUCLIDES RADIOACTIVE DECAY** 

Primary	$T_{1/2}$	Daughter	$T_{1/2}$	$E_{\gamma}$ , keV	$I_{\nu}$ , %
fragment		Isotope			
89Kr	$3,2 \text{ m}$	89Rb	15 <sub>m</sub>	1032	58
				1248	43
$91$ Kr	9 <sub>s</sub>	$91$ Sr	9,5h	750	23,6
				1024	33,4
$^{92}$ Kr	2s	$^{92}Sr$	2,7h	1384	90,1
		92Y		934	13,9
$^{93}\mathrm{Kr}$	1 <sub>s</sub>	93Y	10,1 h	267	7,32
$^{138}\mathrm{Xe}$	14 <sub>m</sub>	$138$ Cs	32 <sub>m</sub>	1436	76,3
				2218	15,2
$^{139}\mathrm{Xe}$	40 <sub>s</sub>	$^{139}\mbox{Ba}$	83 m	166	23,6
$\rm ^{140}Xe$	14s	$^{140}\mathrm{Ba}$	12,8d	537	24,4
		$140$ La	40 h	1596	95,4
$141$ Xe	1,8s	$^{141}Ba$	18 m	190	46
		$141$ La	4 h	1355	1,64
$142$ Xe	1,2s	$142$ La	93 m	641	47,4
				2398	13,3
$143$ Xe	0,8s	143Ce	33h	293	42,8

	$\sqrt{232}$ Th( $\gamma$ ,f)		$\frac{238}{2}U(\gamma,f)$		$\sqrt[244]{P}u(\gamma,f)$				
Fragmen									
	$\tilde{\text{Y}}_{\underline{\text{rel}}}$	$Y_{\rm fr}$	$\bar{\rm Y}_{\rm rel}$	${\rm Y}_{\rm fr}$	$\tilde{\text{Y}}_{\underline{\text{rel}}}$	${\rm Y}_{\rm fr}$			
$\overline{{}^{89}\text{Kr}}$	0,15(2)	0,10(2)	0,29(1)	0,18(2)					
$91$ Kr	1,00	0,62(5)	1,00	0,60(5)					
$^{92}$ Kr	0,95(1)	0,59(5)	0,80(2)	0,46(4)					
$^{93}$ Kr	0,25(1)	0,15(2)	0,25(2)	0,15(2)					
137Xe			0,27(3)	$0,16(2)$ *					
138Xe	0,85(1)	0,53(4)	0,65(3)	0,38(4)	0,84(4)	0,47(4)			
139Xe	1,00	0,63(5)	1,00	0,59(5)	1,00	0,56(5)			
140Xe	0,85(7)	0,53(5)	0,94(7)	0,56(5)	1,03(8)	0,57(5)			
$^{141}Xe$	0,19(1)	0,12(1)	0,53(3)	0,31(3)	0,73(4)	0,41(4)			
142Xe	0,09(1)	0,06(1)	0,26(2)	0,16(2)	0,46(4)	0,26(3)			
$143$ Xe			0,08(2)	0,05(1)	0,24(4)	0,14(2)			
$*$ nonar $[6]$									

TABLE 3 INDEPENDET YIELDS OF Kr and **Xe** FISSION FRAGMENTS

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### TABLE 4

## ISOTOPIC DISTRIBUTION PARAMETERS OF Kr AND Xe FISSION FRAGMENTS



\*this work

## **TABLE 5 CHARGE SHIFTS OF Kr AND Xe FISSION FRAGMENTS** RELATIVE UNCHANGED CHARGE DISTRIBUTION





Fig. 1. Block diagram of the experimental set-up.



Fig. 2. Examples of  $\beta$ -decay chains of fission fragments that contain Kr and Xe isotopes. The isotopes whose  $\gamma$ -spectra were measured are marked.



Fig. 3. Mass number distributions of the fractional yields of Kr and Xe fragments for the photofission of  $^{232}$ Th,  $^{238}$ U and  $^{244}$ Pu nuclei. The dots are experimental data; the solid curves Gaussian distributions.



Fig. 4. Mass number distributions of the independent yields (referred to the number of fission events) of uranium isotopes for the reactions  $^{238}U(\gamma,f)$  - 1,  $^{238}U(n_{14.7\text{ MeV}},f)$  - 2,  $^{235}U(n_{\text{therm}},f)$  - 3,  $^{233}U(n_{\text{therm}},f)$  - 4.

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Гангрский Ю. П. и др. Е15-2002-147 Независимые выходы изотопов Кг и Хе при фотоделении тяжелых ядер

Представлены результаты измерений выходов первичных осколков (образующихся непосредственно при разрыве ядра) Кг (А=87-93) и Хе (А=148-143) при фотоделении ядер <sup>232</sup>Th, <sup>238</sup>U и <sup>244</sup>Pu. Эксперименты проведены на тормозном излучении микротро на при энергии ускоренных электронов 25 МэВ. Использовалась методика переноса вылетевших из мишени осколков газовым потоком по капилляру и конденсации инертных газов Кг и Хе в криостате при температуре жидкого азота. Осколки всех остальных элементов задерживались фильтром на входе в капилляр. Идентификация изотопов Кг и Хе проводилась по у-спектрам их дочерних продуктов. Получено распределение независимых выходов изотопов Кг и Хе по их массовым числам, проведено сравнение этих распределений с аналогичными данными при делении тепловыми и быстрыми нейтронами, определены сдвиги зарядов осколков относительно неискаженного зарядового распределения. Обсуждаются перспективы использования осколков фотоделения в исследованиях структуры ядер с большим избытком нейтронов.

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Gangrsky Yu. P. et al. E15-2002-147 Independent Yields of Кг and Xe Isotopes for the Photofission of Heavy Nuclei

Presented are the yields of primary fission fragments (produced in the process of the rupture of the nucleus) of  $Kr(A=87-93)$  and  $Xe(A=148-143)$  for the photofission of  $232$ Th,  $238$ U and  $244$ Pu, which were measured using bremsstrahlung from a microtron the energy of accelerated electrons being 25 MeV. A technique was used that includes transportation of fragments escaped from the target with a gas flow through a capillary and the condensation of Kr and Xe inert gases in a cryostat at the temperature of liquid nitrogen. Fragments of all the other elements were retained with a filter at the entrance of capillary. Kr and Xe isotopes were identified by the  $\gamma$ -spectra of their daughter products. The mass number distributions are obtained of the independent yields of Kr and Xe isotopes, which are compared with the similar characteristics for the fission induced by thermal and fast neutrons; the charge shifts for the fragments under study relative to the unchanged charge distribution are determined. The perspectives of the photofission fragments using in the study of neutron-rich nuclei structure are discussed.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, J1NR.

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