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PRODUCTION OF PHOTOFISSION FRAGMENTS AND STUDY OF THEIR NUCLEAR STRUCTURE BY LASER SPECTROSCOPY

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The prospective nuclear structure investigations of the fission fragments by resonance laser spectroscopy methods are discussed. Research in this field is currently being carried out as part of the DRIBs project, which is under development at the Laboratory of Nuclear Reactions, JINR. The fission fragments under study are mainly very neutron-rich nuclei near the proton ($Z = 50$) and neutron ($N = 50$ and 82) closed shells, nuclei in the region of strong deformation ($N > 60$ and $N > 90$) and nuclei with high-spin isomeric states. Resonance laser spectroscopy is used successfully in the study of the structure of such nuclei. It allows one to determine a number of nuclear parameters (mean-square charge radius, magnetic dipole and electric quadrupole moments) and to make conclusions about the collective and single-particle properties of the nuclei.

Обсуждаются перспективы исследования структуры радиоактивных ядер — продуктов фотоделения методами резонансной лазерной спектроскопии. Исследования в этом направлении выполняются в рамках проекта DRIBs, реализуемого в Лаборатории ядерных реакций ОИЯИ. Исследуемыми продуктами фотоделения являются сильно нейтронообогащенные ядра вблизи замкнутых протонной ($Z = 50$) и нейтронной ($N = 50$ и 82) оболочек, ядра в областях сильной деформации ($N > 60$ и $N > 90$), высокоспиновые изомеры. Применение методов лазерной спектроскопии позволяет определить ряд важнейших ядерных параметров (среднеквадратичный зарядовый радиус, магнитный дипольный и электрический квадрупольный моменты) и судить о роли коллективных и одночастичных свойств в этих ядрах.

NUCLEAR STRUCTURE PECULIARITIES OF THE FISSION FRAGMENTS

The fission fragments of heavy nuclei ($Z > 90$) are neutron-rich isotopes of the elements from Zn ($Z = 30$) to Nd ($Z = 60$) with a neutron number of 50–95. The large neutron excess in the fission fragments under study (in some cases there are 10–15 more neutrons than in the nuclei situated in the β-stability valley) could lead to an essential change in their structure and radioactive decay characteristics. The abnormal ratio of protons and neutrons in such nuclei reflects on spin-orbit interactions and can lead to another order of nucleon shell filling. This change will manifest itself in the appearance of new magic numbers of protons or neutrons, new regions of deformation and new islands of isomerism. A striking example of this phenomena has already been observed at the shell closure $N = 20$ in the light neutron-rich nuclei of ^{31}Na and ^{32}Mg. In contrast to our knowledge about nuclear structure, these nuclei are strongly deformed [1, 2]. The same situation could occur in the case of very neutron-rich isotopes of Cu and Zn around $N = 50$, as well as Ag and Cd around $N = 82$.

One can also expect an unusual change in nuclear structure at the boundary of deformation regions. At the beginning of the known region around $N = 90$ the quadrupole deformation parameter increases in a different way for various elements: there is a sharp jump in the β-stability valley (isotopes of Nd, Sm, Eu, and Gd) and a smooth growth outside it (neutron-rich isotopes of Ba and neutron-deficient isotopes of Yb [3]). At the same time the octupole deformation parameter decreases in these nuclei. The nuclei on the boundary of other deformation regions are studied much less. It is only known that there is a sharp jump in quadrupole deformation at $N = 60$ in the isotopes from Kr ($Z = 36$) to Mo ($Z = 42$). It manifests itself by the strong change in their charge radii [3] and the reduced probabilities of electric quadrupole transitions.

Peculiarities of the fission fragment decay are the high energy of the β decay and low neutron binding energy of the daughter nuclei $Q_\beta > 5$ MeV and $B_n < 5$ MeV in the primary fragments formed directly in fission. Therefore the wide spectrum of excited levels of different nature is populated in the β^- decay of fission fragments. Neutron emission accompanies γ radiation in the de-excitation of these levels. The high energy of β decay can result in the appearance of new, more rare, modes of radioactive decay. Here refers the emission of a neutron pair ($\beta 2n$) or of an α particle after β decay ($\beta\alpha$). These decay modes are also an important source of new information about nuclear structure.

The examples discussed above do not exhaust the variety of peculiarities of the fission fragments. They are relatively poorly studied, and the measurement of their nuclear moments, the level spectra, the decay schemes, etc. allows one to establish the way in which nuclear structure changes with increasing neutron excess. A wide set of experimental devices should be used to obtain this information.

PROJECT DRIBs — DUBNA RADIOACTIVE ION BEAMS

The nuclear structure study of fission fragments is one of the main directions of the project DRIBs, being developed at the Flerov Laboratory of Nuclear Reactions, JINR. The aim of this project is the production of intense beams of accelerated radioactive nuclei in a wide range of Z and A — from He to rare-earth elements. Light neutron-rich nuclei (up to Na) will be produced in the fragmentation of bombarding ions on the 4-m isochronous cyclotron U-400M, and nuclei of a medium mass number — in the fission of uranium on the electron accelerator microtron MT-25. The total yield of fission fragments up to 10^{11} s^{-1} is expected at the irradiation of ^{238}U-target 10 g/cm^2 with the electron beam of 25 MeV and 20 μA. Nuclei chosen for study will be mass-separated and transported to be accelerated in another 4-m isochronous cyclotron U-400.

Study of reactions induced by neutron-rich or neutron-deficient nuclei essentially enlarges information about their structure. It is impossible to judge some details of this structure from radioactive decay characteristics. A striking example is the observation of an unusual wide space distribution of neutrons in some neutron-rich nuclei (neutron halo), first in ^{11}Li and then in others [4]. These data were obtained from measurement of cross sections for different reactions (fusion, stripping, and nucleon exchange) with neutron-rich nuclei. Such multidirection investigations of the properties of nuclei far from the β-stability valley definitely widen our knowledge about the changes in nuclear structure with the growing neutron excess and about the appearing of new phenomena. Reactions with neutron-rich nuclei can also be used for obtaining more neutron-rich nuclei. Really, the compound nuclei formed in these reactions contain the neutron excess, and the evaporation of charged particles increases this excess. By this technique it is possible to get the most neutron-rich nuclei and to draw near the boundary of nucleon stability.

THE YIELDS OF FRAGMENTS IN PHOTOFISSION

The successful study of the structure of fission fragments, especially of the most neutron-rich fragments, depends to a great degree on their yields. These are determined by the mass and atomic number (A and Z) distribution of the produced fission fragments. However, there is poor information about these parameters in photofission as compared with neutron fission. Worthy to be referred are only the investigations performed in Gent (Belgium) [5, 6].

Fission fragments of a medium mass number will be produced in fission of uranium on the electron accelerator microtron MT-25. At maximum energy of 25 MeV of the microtron bremsstrahlung the main contribution to the photofission fragment yield is provided by the 10–15 MeV energy range of its spectrum. This is just the position of the giant dipole resonance in heavy nuclei. This energy range also determines the excitation energy of nuclei in fission. At such an excitation energy the mass spectra of the photofission fragments are asymmetric with an average mass numbers of about 100 and 140 for the light and heavy groups of fragments, respectively. This spectrum and analogous ones for neutron fission of U (thermal and 14.7 MeV neutrons) are presented in Fig. 1. It is seen the yield dependence on the mass number for the case of photofission lies between those for the cases of the neutron fission (the ratio of peak to valley is about 100). Each mass number in these spectra corresponds to some set of nuclides (4–6) with different atomic numbers Z formed directly at the scission point or after β decay.

To get more detailed information about the isotopic yields, the mass number distributions of Kr and Xe photofission fragments of heavy nuclei ^{232}Th, ^{238}U, ^{237}Np and ^{244}Pu induced by bremsstrahlung with a boundary energy of 25 MeV have been measured [7]. A method of transporting the fission fragments by a gas flow and stopping in a cryostat with liquid nitrogen was used [8]. The isotopic distributions of Xe fission fragments of ^{238}U obtained by this technique are presented in Fig. 2 (the same distributions for fission of ^{238}U by 14.7 MeV neutrons and of ^{235}U by thermal neutrons are shown for comparison [9]). These distributions are fitted by Gauss curves:

$$Y\left(A\right) = Y\left(\bar{A}\right)\,\exp\left\{-\frac{\left(A-\bar{A}\right)^2}{2\sigma^2}\right\}, \tag{1}$$

where $\bar{A}$, mean mass number, and σ, dispersion, are the parameters of these curves. These parameters for different fission reactions are presented in Table 1. It is seen that the distributions for the fission of ^{238}U by γ rays and of ^{238}U 14.7 MeV neutrons are similar. But an enhanced yield of the most neutron-rich nuclides is observed in photofission as compared with thermal neutron fission of ^{235}U, and this difference becomes larger with increasing neutron excess.

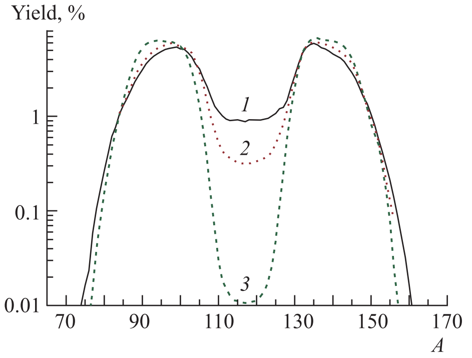

Fig. 1. The mass spectrum of the fragments in the fission of uranium induced by γ rays and neutrons: *1* — ^{238}U(n,f), 14.7 MeV; *2* — ^{238}U(γ,f), 25 MeV; *3* — ^{235}U(n,f), thermal

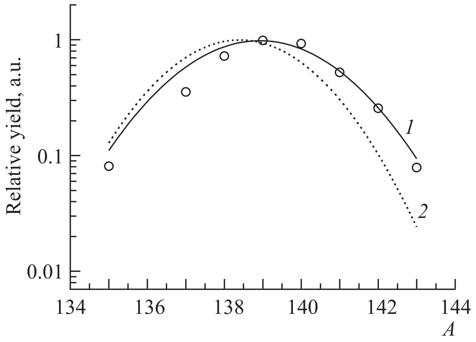

Fig. 2. The isotopic distributions of Xe fission fragments: *1* — ^{238}U(n,f), 14.7 MeV; ○ — ^{238}U(γ,f), 25 MeV; *2* — ^{235}U(n,f), thermal

Table 1

Reaction	$\frac{N-Z}{N}$	Kr		Xe		References
		$\bar{A}$	σ	$\bar{A}$	σ	
^{232}Th(γ,f)	0.366	91.3(2)	1.1(1)	138.9(2)	1.2(1)	[7]
^{238}U(γ,f)	0.370	91.1(2)	1.3(1)	139.4(2)	1.5(1)	[7]
				138.9(3)		[6]
^{237}Np(γ,f)	0.354	90.3(1)	1.3(1)	137.1(1)	1.3(1)	This work
^{244}Pu(γ,f)	0.373			139.7(2)	1.8(2)	[7]
^{235}U(γ,f)	0.357	89.4(3)	1.3(1)	137.4(4)	1.4(1)	[6]
^{233}U(n,f)	0.352	89.3(1)	1.5(1)	137.8(1)	1.5(1)	[9]
^{235}U(n,f)	0.364	90.1(1)	1.5(1)	138.4(1)	1.6(1)	[9]
^{238}U(n,f)	0.379	91.5(1)	1.6(1)	139.5(1)	1.8(1)	[9]

From the results presented in Table 1 and in Fig. 2, it is possible to conclude that we can use a large amount of available information about neutron fission of heavy nuclei to obtain the isotopic distributions for photofission. One of the parameters of these distributions — mean mass number of fission fragments $\bar{A}$ — is presented in Fig. 3 for the range of their atomic number $Z = 28{-}62$. The independent yields of fragment for each Z and $\bar{A}$ are close to the cumulative yields of fragment with the same A in Fig. 1.

It is possible to obtain the independent yields of fragments at another A for each element using expression (1). The mass number of fragments with the yields 10^{-8} of the total ones is also presented in Fig. 3 (the parameter $\sigma = 1.5$ was used). These yields are close to experimental limit of the produced fission fragments. The nuclides with these yields are unknown as a rule, and their study will bring the new and very interesting information about their nuclear structure.

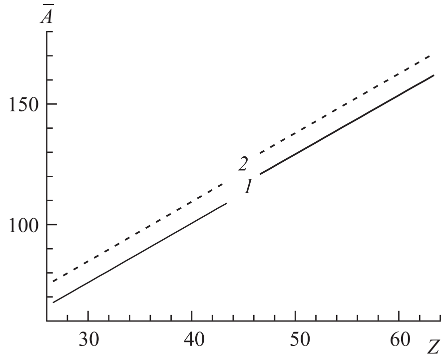

Fig. 3. The mean mass number of fission fragments versus their atomic number at the photofission of ^{238}U: *1* — mean mass number; *2* — the mass number of fragments with yields 10^{-8} of the total ones

Some examples of these fragments and their yields are presented in Table 2. They are interesting from the point of view of their nuclear structure having number of protons or neutrons close to the magic numbers, large quadrupole deformation, and unusual mode of radioactive decay. The yields are large enough, and so they allow one to perform successful investigations of such nuclides.

Thus, the photofission reactions of heavy nuclei are very convenient and promising way for the production of intense beams of nuclides with maximum neutron excess. The large penetrating power of the γ rays allows one to use thick targets. But the low excitation energy of the nuclei in fission and of the fission fragments results in a small number of evaporated neutrons. This compensates for the low photofission cross sections which are smaller (0.5 b for the case of ^{238}U) in comparison with the cross sections for fission induced by charged particles and neutrons. Moreover, electron accelerators are simpler and much cheaper than charged particle accelerators or atomic reactors.

Table 2

Fission fragment	Peculiarities	$Y, 1/f$	$Y, 1/s$ (DRIBs)
^{78}Ni	Double magic nucleus $Z = 28$, $N = 50$	10^{-8}	10^{3}
^{132}Sn	Double magic nucleus $Z = 50$, $N = 82$	$3 \cdot 10^{-3}$	$3 \cdot 10^{8}$
^{134}Sn	Two neutrons over closed shell	$8 \cdot 10^{-4}$	10^{7}
^{104}Zr	Strongly deformed nucleus	$5 \cdot 10^{-4}$	$5 \cdot 10^{7}$
^{160}Sm	Strongly deformed nucleus	10^{-4}	10^{7}
^{134}Sb	Delayed two-neutron emitter	10^{-6}	10^{5}
^{140}J	Delayed α emitter	10^{-5}	10^{6}

TRANSPORT AND SEPARATION OF FISSION FRAGMENTS

Fission fragments which are the farthest from the β-stability valley and most interesting from the point of view of their nuclear structure are very short-lived as a rule (their half-lives are less than 1 s). Therefore, to be successfully investigated, fission fragments should be fast and efficiently transported to measuring devices. Although at the present time a number of science centers use spectrometers of undelayed fission fragments (a review of these facilities is given in [10]) with a transport time of less than 1 ms for a distance of several meters, their efficiency is very low ($< 10^{-4}$) and the possible targets are thin (< 1 mg/cm^{2}). So their capabilities in study of fission fragment structure are limited and it is necessary to search for other ways.

One of such methods is transport of the fission fragments with a gas flow. Fragments which have escaped from the target are slowed down in a noble gas, adsorbed by aerosols and carried through a capillary to the measuring devices. In a set-up of this kind, developed at FLNR, JINR, the transport time is 0.2 s for a distance of 1 m and 3.5 s for a distance of 30 m at the efficiency up to 70% [8]. The target thickness corresponds to the stopping range of fission fragments in uranium (~ 10 mg/cm^2), but it is possible to use several targets (up to 10), placed in parallel. It is much larger than in spectrometers of undelayed fragments, but smaller than the stopping range of bombarding particles in a substance, and their capabilities are not used entirely. Another disadvantage of this method is the absence of separation for the chosen Z and A fission fragments (it is necessary for their successful study).

The most efficient method of fission fragment separation by mass number A is to use electromagnetic mass-separators. In on-line experiments the irradiated target is usually a part of the ion source. This target must be thick enough (up to several tens of grams) to increase the yield of fission fragments. This thickness is much larger than the stopping range of fragments, and they can leave the heated target only by a thermal diffusion. The extraction efficiency of fragments from the target and the ionization probability are determined by their thermal properties (melting and boiling points, saturated vapor pressure) and by the ionization potential. For this reason it is impossible to develop a universal ion source that is efficient enough for all fission fragments. Ion sources of various types are used for different groups of elements: surface ionization sources for alkaline, alkaline-earth and rare-earth elements and gas discharge sources for noble gases and for a number of volatile elements. For the group of refractory elements, IGISOL ion source was developed [11]. The ion recoils are stopped in noble gas and then carried into the electromagnetic separator.

Each mass number in the spectrum of fission fragments corresponds to a number of different elements (up to 5–6). They are formed both directly in the fission and after the β decay. For detailed study of chosen nuclei it is important to remove the unnecessary contamination. Sometimes it is possible to use a difference in their properties, for instance, in the pressure of the saturated vapors. But the most effective way seems to be selective laser ionization [12, 13]. Under definite conditions, a source contains predominantly neutral atoms. Absorbing 2–3 photons, they are ionized by laser radiation. Since different elements have quite different atomic levels schemes, the resonant absorption of laser photons occurs only for a chosen element, and a very high degree of separation can be obtained.

LASER METHODS IN THE INVESTIGATION OF FISSION FRAGMENTS

One of the directions of nuclear structure research is obtaining the information about the space distribution of the nuclear matter and of the electric charge and current in nuclei. The radial distribution of these quantities is determined by mean radii and the azimuthal distribution by multiple moments. The high-resolution laser resonance spectroscopy methods are used efficiently for measurement of these nuclear parameters [14, 15]. These methods are based on excitation of electron levels of an atom and on precise measurement of the transition energies between these levels. The electron shell of atoms or ions is very sensitive to a number of nuclear parameters. The finite dimension of the nucleus results in a shift of the atomic levels and nuclear moments — in their splitting. Although these changes are very small (their relative values are less than 10^{-5}–10^{-6}), the contemporary optical methods allow high-accuracy measurement. These methods are the most efficient when tunable dye laser is used. The main peculiarity of the laser methods is the resonance excitation of transitions between atomic levels which is characterized by an enormous, for an elementary system, probability and explains the efficiency of laser spectroscopy. It is possible to observe the appearance of the resonance by a number of signs: a sharp increase in resonance laser fluorescence, occurrence of nuclear radiation anisotropy, laser ionization by stepwise resonant excitation with two or more laser quanta the sum of which exceeds the ionization energy of the atom. The use of tunable lasers characterized by high intensity and very narrow spectral lines allows simultaneous achievement of high resolution and especially high sensitivity which are impossible for the traditional optical methods. This permits one to perform precise measurements with extreme small quantities of atoms. Thus, nuclei at the nucleon drip-lines obtained with a very low yield still could be studied. The following examples show the ultimate capabilities of laser spectroscopy methods:

1. *The sensitivity:* the lowest yield in the successful measurements was reached for fission isomers of Am — the yield of isomer was only 10 s^{-1} [16].
2. *The expressivity:* the most short-lived studied nucleus was the isomer ^{87}Rb with a half-life of 10^{-6} s (obtained in the β decay of ^{87}Kr) [17].
3. *The accuracy:* the best value of 10^{-12} was obtained in the measurement of hydrogen–deuterium isotopic pair frequency difference [18].

These examples confirm the efficiency of laser methods for the study of a wide number of nuclei in ground and isomeric state with half-lives up to the microsecond range.

OPTICAL PROPERTIES OF FISSION FRAGMENTS

The success of laser methods in investigation of fission fragments is determined essentially by their optical characteristics. Since laser methods are based on excitation of atomic levels, the initial state must be the ground state or a long-lived metastable state. Moreover, it is necessary that one of the states of transition is sensitive to the measured nuclear parameters. In measurement of charge radii these transitions with s-electron jump (or to a much smaller extent with p-electron jump) or also transitions in which the screening of the inner closed electron shells is changed are used. In the case of nuclear multipole moments the requirements to atomic level configurations are not so restricting. However, the orbital moment l of level must correspond to the multipolarity of the measured moment (for instance, for the electric quadrupole moment it is necessary that $l \geqslant 1$).

Measurement of the above-considered nuclear parameters will be most efficient if the exciting laser radiation is from the visible, near-infrared or near-ultraviolet regions of the spectrum. An analysis of fission fragment optical spectra was performed in our previous paper [19]. It is shown that most elements of the I, II, III, IV and VIII groups of the Mendeleev table satisfy these requirements. They make up the largest part of the fission fragment nuclei. For atoms of the V, VI and VII groups the transitions convenient for measurement lie at high excitation energies (as a rule more than 40000 cm^{-1}). But in some of these elements the initial levels of these transitions are metastable, and they can be populated in a charge exchange with, e.g., alkali vapor. Thus, one can conclude that the optical properties of fission fragments allow one to study their nuclear structure in very interesting nuclear regions by laser spectroscopy methods.

PROSPECTS OF FISSION FRAGMENTS NUCLEAR STRUCTURE INVESTIGATION BY LASER METHODS

As discussed earlier, the laser spectroscopy methods are very sensitive and precise tools for nuclear structure investigations. They allow one to understand the nuclear shape evolution, the role and interplay of single-particle (spin, magnetic moment) and collective effects (charge radius, electric quadrupole moment) in nuclei. These values determine the nucleon configurations of ground and isomeric states, the size and shape of a nucleus; they identify the levels in the scheme of a nuclear model. It is very important to establish how these nuclear properties are changed with increasing neutron excess and with the approach to the neutron drip-line.

The experimental set-up was made at FLNR, JINR, for the measurements of these nuclear parameters. It is based on the resonance fluorescence registration of the studied atoms. The combination of the set-up with the mass-separator of the fission fragments is planned in the near future.

The most interesting regions and directions of investigation of fission fragments are the following:

1. Nuclei near neutron shells closures: $N = 50$ (isotopes of Zn, Ge, Ga) and $N = 82$ (isotopes of Cd, In, Sn). The regularity of charge radii changes in an isotope sequence from the β-stability valley or neutron-deficient nuclei is well known: nuclides with a magic neutron number have the smallest radius (or the most compact shape). It will be very interesting to clarify if this trend remains in the case of neutron-rich nuclei. Here it is very important to determine the parameters of lower single-particle states around the shell closure.

2. Nuclei at the boundary of the deformation region with $N > 60$ (isotopes of Zr, Nb, Mo, Tc). In the known deformation region ($N > 90$) a change in a nuclear shape is quite different for different atomic numbers [3]. A sharp jump of deformation is observed at $N = 60$ in nuclei with $Z \leqslant 40$, but it is unclear what will happen in nuclei with $Z > 40$.

3. Shape isomers in fission fragments. In some nuclei there exist excited states with unusual large quadrupole deformation (the deformation parameter $\beta \geqslant 0.6$). In nuclei with $Z \geqslant 92$ these states are isomeric (fission isomers [20]). In other regions of nuclei excited rotational bands are observed with an unusual large quadrupole deformation (the super- and hyper-deformations [21]). However, the lowest states of these bands have not been observed up to now. Probably these states are isomeric and their study could be of great interest. One can identify such states by the abnormal isomeric shift of the optical lines, caused by the large deformation difference between ground and isomeric states. Among fission fragments such states have been predicted for nuclei around ^{78}Zn and ^{114}Ru. Some indications of the existence of shape isomerism in fission fragments are observed in [22].

4. Nuclei with difference in spatial distribution of electrical charge and nuclear matter. The large excess of neutrons in fission fragments can result in an essential difference in the spatial distribution of protons and neutrons. Usually information about this difference is obtained by a comparison of the cross sections for elastic scattering and the charge-exchange reactions induced by such nuclei. Experiments of this kind are planned to be carried out on beams of accelerated fission fragments in the DRIBs project. But it is possible to get similar

information in laser spectroscopy experiments. The method to be used is based on the Bohr–Weisskopf effect [23] — the influence of nuclear magnetism distribution on the hyperfine splitting of atomic levels. Here the deviation of the ratio of the gyromagnetic factors from the ratio of the magnetic dipole constants (the so-called hyperfine magnetic anomaly) gives information about neutron distribution radius.

These examples show how wide the field of activity can be in the study of the nuclear structure of neutron-rich nuclei, and the DRIBs project is the first step on this way.

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