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SHELLS IN SUPERASYMMETRIC NUCLEAR FISSION

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The multimodal approach to fission and the macroscopic-microscopic method for the calculations of charge distribution parameters for isobaric chains have been used to analyze fission product yields. In order to describe the peculiarities of fragment mass curve at very asymmetric mass split, the two narrow fission modes related to the magic numbers Z = 28 and N = 50 were introduced. The reliability of the model's predicting power was demonstrated by the agreement between calculated and experimental data on the thermal-neutron-induced fission of actinides. It was found that weight of the fission modes related to the spherical doubly-magic clusters (^{132}Sn or ^{78}Ni) depends on the neutron-to-proton ratio of a compound system.

After the first publications [1 - 3] where V. Strutinsky had proposed and formulated his famous shell-correction method, it becomes clear that this shell-correction approach was a powerful method for study of a large-scale collective motion in nuclei, and in particular, in the nuclear fission process. Quantitative description of the mass asymmetry in nuclear fission and yields of fission fragments is still a challenging problem for the nuclear theory. Application of the shell-correction method for calculations of the potential-energy surface (PES) near the fission barrier leads to the prominent result, namely, the second barrier is unstable against the mass-asymmetry collective coordinate for all actinides [4, 5]. However, the calculations of PES near the scission point within Strutinsky's shell-correction method gave explanations of the nuclear fission characteristics in terms of the nuclear shells in fragments [6, 7]. Indeed, the main features of the nuclear fission of actinides can be explained by influence of the spherical nuclear shells with Z = 50 and N = 82 (^{132}Sn nuclear cluster). Other important combinations of the shells, for instance, with Z = 82 and N = 126 (^{208}Pb nuclear cluster), may also play a significant role in fission and quasi-fission of superheavy nuclei. The combination of the Z=50 and N=50 nuclear shells (^{100}Sn nuclear cluster) does not influence the fission of nuclei, because of too exotic N/Z ratio. The manifestation of the double-magic ^{78}Ni nucleus (Z = 28 and N = 50) in the nuclear fission process was not proved until now. PES of a fissioning nucleus in the collective-coordinate space consists of the saddle and the bifurcation points defined by shells in the compound nucleus, with valleys and ridges depending on the fission-fragment properties. The dynamical calculations even in the minimal collective-coordinate space are still an open problem.

In this report, we suggest the theoretical approach for analysis of the recent experimental data and with the purpose of searching a manifestation of the Z = 28 and N = 50 nuclear shells (^{78}Ni fission mode) in fission of actinides. The superasymmetric mass region (A < 80) is difficult to access experimentally because the yield of very light fission products decreases rapidly and at about A ≈ 70, it becomes comparable with that of heavy particles from ternary fission. In the fission-fragment mass interval corresponding to $A_L \approx 20 - 70$ the data are rather scarce; this region is really a "terra incognita".

A great deal of interest in the investigation of very asymmetric fission is connected with a search for the superasymmetric fission mode, the synthesis and the exploration of superheavy elements and with clarifying the perspectives to produce exotic neutron-rich nuclides for future Radioactive Nuclear Beam (RNB) facilities. Experimental studies on thermal-neutron-induced fission performed at the Lohengrin mass separator in the Institut Laue-Langevin (ILL) of Grenoble [8] during recent years, for a number of fissile nuclei, have extended the experimental knowledge of isotopic and fragment-mass behavior down to approximately A = 70. This brought to light a discontinuity in the fission-product mass-yield curve around mass A = 70, which was explained by the influence of the Ni shell in this mass region [9]. The systematic experimental and the theoretical studies of extreme asymmetric fission at the intermediate energy are being carried out at the Accelerator Laboratory of the University of Jyväskylä, Finland. Enhancement of the fission product yields at A < 80 in the proton-induced fission of ^{238}U at the energy $E_p = 25$ MeV, as compared to the low-energy fission, was established using IGISOL measurements [10, 11]. This enhancement in a far asymmetric mass region was also supported by the time-of-flight measurements with HENDES setup, from the proton- and the neutron-induced fission of ^{238}U with using the ^{238}U(p, f) [12] , ^{238}U(d, pf) [13] and ^{242}Pu(p, f) [14] reactions. Below we will follow our recent publications [15, 16] and will demonstrate of the superasymmetric fission mode (^{78}Ni mode) for the case of the thermal-neutron-induced fission of actinides.

The independent yield of fission fragments Y_{ind} is defined as the yield of the specific fission product after a prompt neutron emission from the excited primary fragments emerging in fission of a compound nucleus with mass A_c, charge Z_c, and excitation energy E_c:

$$Y_{ind}(A,Z,A_c,Z_c,E_c) = \sum_n P_n(A+n,Z,A_c,Z_c,E_c)P_{pre}(Z,A+n,A_c,Z_c,E_c)Y_{pre}(A+n,A_c,Z_c,E_c), \quad (1)$$

where $P_n(A+n,Z)$ is the probability of a prompt neutron emission from the fragment with mass $A+n$ and charge Z, and $P_{pre}(Z,A+n)$ a charge distribution of the $(A+n)$ isobaric chain, and $Y_{pre}(A+n)$ a primary fission-fragment mass distribution. We omit here and below some arguments for simplicity.

At low excitation energies, the primary fission-fragment mass and the charge distributions show an odd-even staggering. The primary distributions can be presented in the factorized form

$$P_{pre}(Z) = \widetilde{P}_{pre}(Z)F_{oe}(Z), \quad Y_{pre} = \widetilde{Y}_{pre}(A)F_{oe}(A), \quad (2)$$

where $\widetilde{P}_{pre}(Z)$ and $\widetilde{Y}_{pre}(A)$ are smoothed distributions, $F_{oe}(Z)$ and $F_{oe}(A)$ are functions which describe the odd-even staggering. The method for modeling the smoothed mass distribution is based on the multimodal nature of nuclear fission [17], depicting the influence of the nuclear-shell structure on PES of fissioning nucleus. The fission process is the most probably guided by valleys and bifurcation points of the PES from the equilibrium shape to the scission point. For heavy actinides (from Th to Cf), the so-called standard fission modes (symmetric, spherical ^{132}Sn, and deformed $N = 86-90$ shells) have been used. In the case of spontaneous and thermal-neutron-induced fission, these standard modes have to be supplemented with the two additional modes, in order to evaluate better the smoothed primary-mass distribution in the superasymmetric mass region:

$$\widetilde{Y}_{pre}(A) = C_{SY}Y_{SY}(A) + C_{SI}Y_{SI}(A) + C_{SII}Y_{SII}(A) + C_{SA1}Y_{SA1}(A) + C_{SA2}Y_{SA2}(A). \quad (3)$$

Here Y_{SY}, Y_{SI}, Y_{SII}, Y_{SA1}, and Y_{SA2}, are the symmetric and the asymmetric components, respectively. They stand for the contributions from different fission modes. Each asymmetric component normalized to unity consists of the two peaks representing the heavy- and the light-fragment-mass groups. The component Y_{SI} is related to the magic numbers $Z = 50$ and $N = 82$ in heavy fragments, and the superasymmetric components Y_{SA1} and Y_{SA2} are influenced by the $N = 50$ and $Z = 28$ nuclear shells in light fragments. The asymmetric mode Y_{SII} is supposed to be associated with a "deformed" nuclear shell at $N = 86-90$. The competition between fission modes is determined by the fission dynamics and the nuclear shells in fission fragments. The coefficients C_i in (3) were obtained by comparison with experimental data for the thermal-neutron-induced fission of heavy nuclei, and for higher excitation energies the phenomenological approaches were proposed in Ref. [16]. In the previous analysis of the fission-product yields at intermediate energies [16, 18], both light and heavy peaks were approximated by Gaussian distributions. However, our analysis of Lohengrin data [9, 15], which are very precise, shows that such an approximation fails at a large deviation from the peak center. The Gaussian distribution corresponds to the harmonic approximation of the free energy near the bottom of the valley. In order to take into account the anharmonicity correction, the mass dependence of the variation σ_A was introduced for two asymmetric fission modes (SI and SII, see Ref. [15]). This method enables to suppress the contributions of the standard asymmetric modes in symmetric and superasymmetric mass regions.

The smoothed charge distribution of a primary isobaric chain is approximated by a Gaussian function

$$\widetilde{P}_{pre}(Z) = \frac{1}{\sigma_Z(A)\sqrt{2\pi}} \exp\{-\frac{(Z-\overline{Z}(A))^2}{2\sigma_Z^2(A)}\}, \tag{4}$$

where $\overline{Z}(A)$ is the averaged charge of the primary isobaric chain. This value deviates a little from an estimation which can be obtained from the hypothesis of a unchanged charge density distribution during the fission process

$$\overline{Z}(A, A_c, Z_c, E_c) = A\frac{Z_c}{A_c} + \delta\overline{Z}(A, A_c, Z_c, E_c) \ . \tag{5}$$

The deviation $\delta\overline{Z}$ is determined by global liquid-drop properties of potential-energy surface near the scission point and also by nuclear shell effects. In order to calculate $\sigma_Z(A_c)$ we shall consider the isobaric charge width as a result of a frozen quantal fluctuation at the scission point. [19]. Fissioning nucleus at this point is described by two slightly overlapping fragments. The radius of the aperture (neck radius) through which the two fragments may exchange nucleons is equal to r_{neck}. The proton-to-neutron degrees of freedom are assumed to be much faster than the deformation and mass ones. Therefore it is possible to fix the fragment masses and study only the variation of the fragment charge.

Potential energy at the scission point, relative to energy of two fragments in the ground states at the infinite separation, consists of the interaction energy between them and their deformation energies

$$V_{scp} = V_{Coul} + V_{nucl} + E_{def}^{L} + E_{def}^{H}, \tag{6}$$

where V_{Coul} and V_{nucl} are the Coulomb and the nuclear interaction energies, E_{def}^{L} and E_{def}^{H} are the deformation energies of the light (L) and the heavy (H) fragments, respectively. The deformation energy was calculated by making use the Strutinsky macroscopic-microscopic method [1 - 3]. The single-particle spectra were obtained for the axially deformed Woods-Saxon potential with "universal" nuclear-potential parameters proposed in Ref. [20]. For a given compound nucleus, after minimization of the potential relative with respect to the deformation parameters of both fragments, its two-dimensional function $V_{scp}^{\min}(A,Z)$ at the minimum was computed. For a fixed fragment mass, the last function was approximated by the parabolic dependence

$$V(Z) = V(\overline{Z}) + \tfrac{1}{2}C_{ZZ}(Z-\overline{Z})^2 \tag{7}$$

and parameters $\overline{Z}$ and $C_{ZZ} = \frac{\partial^2 V_{scp}^{\min}(A;Z)}{\partial^2 Z}\Big|_{Z=\overline{Z}}$ were determined.

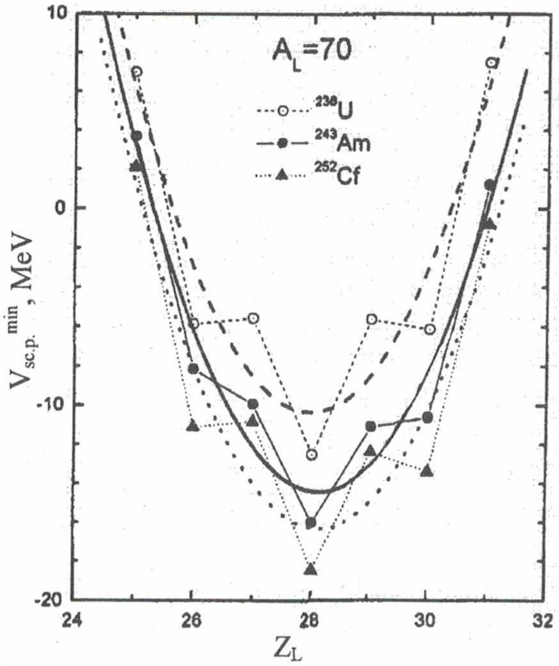

Fig. 1. Potential energy dependence on the light fragment charge for the isobaric chain $A = 70$ calculated for ^{236}U, ^{243}Am, and ^{252}Cf. Their approximations are shown by continuous lines.

An example of a potential energy calculated relative to ground state of the compound nucleus, as function of the fragment charge at A = 70 for ^{236}U, ^{243}Am, and ^{252}Cf is shown in Fig. 1. In Fig. 1, the points represent computed values and the continuous lines are approximating functions. As seen from Fig. 1, for a wide range of compound nuclei, the minimum of potential energy falls on the nuclear shell $Z = 28$. For the inertia parameter M_{ZZ} we used an expression derived in Ref. [21]

$$M_{ZZ} = \frac{16}{9} r_0^3 m \frac{A_c^2}{Z_c N_c} \frac{L + 2r_{neck}}{r_{neck}^2}, \qquad (8)$$

where r_0 is the nuclear radius parameter ($r_0 = 1{,}16$ fm), m the nucleon mass, and L the neck length (L = 2 - 4 fm). Estimation for the radius neck is $r_{neck} = 2$ fm or $r_{neck} = 0{,}5(A(A_c - A))^{1/6}$ fm.

The energy of these charge oscillations or the giant dipole resonance energy along the symmetry axes for the expanding configuration at the scission point defines the standard deviation of charge distribution of the isobaric chain in the low energy fission

$$E_{GDR}^{\parallel} = \hbar\omega_Z = \hbar\sqrt{\frac{C_{ZZ}}{M_{ZZ}}},$$

$$\sigma_Z(A) = \frac{1}{2}\frac{E_{GDR}^{\parallel}}{C_{ZZ}}. \qquad (9)$$

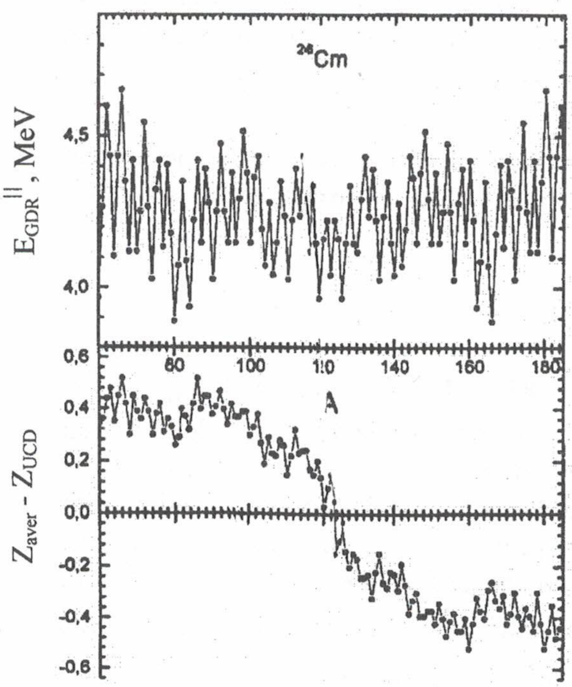

Fig. 2. Calculated values of the giant dipole resonance energy along the symmetry axes $E_{GDR}^{\parallel}$ and charge displacement at the scission point for ^{246}Cm as a function of the primary fragment mass.

The calculated $E_{GDR}^{\parallel}$ and $\sigma_Z(A)$ for ^{246}Cm are presented in Fig. 2. As shown in Fig. 2, the odd-even staggering and the nuclear shell effect manifest themselves in the dependence of $\overline{Z}(A)$ in contrast to the values predicted by the liquid drop model.

Since the smoothed pre-neutron emission isobaric-charge distribution is approximated by a Gaussian function, the odd-even structure can be described by a parameter defined as a third difference of the natural logarithms of the fractional yields. If we consider the proton and the neutron odd-even effect separately, one can write

$$F_{oe}(Z) \propto \exp((\Pi_Z^H + \Pi_Z^L)\delta_Z(A_c, Z_c, A_c)), \quad (10)$$

where Π_Z^H and Π_Z^L are parities of the proton number in heavy and light primary fragments;

$\Pi_Z^{H(L)} = 1$ if Z is even and $\Pi_Z^{H(L)} = -1$ if Z is odd. The proton odd-even difference parameter $\delta_Z(A_c, Z_c, E_c)$ is parameterized in accordance with experimental data [22]. Odd-even staggering in the primary-mass distribution is described by a combination of proton and neutron odd-even effects.

There are still some difficulties with the precise calculations of the prompt neutron multiplicity and additional studies are needed to improve the theory. Therefore a simplified statistical description with parameters fitted to experimental neutron-multiplicity data for the thermal-neutron-induced fission of ^{235}U and spontaneous fission of ^{252}Cf was used in our calculations [15]. The averaged prompt-neutron multiplicity is proportional to the excitation energy of the fragment, which was calculated by using a scission-point model. A linear approximation of the dependence of the standard variation on the mean neutron multiplicity was applied too.

This theoretical model was applied for analysis of the thermal-neutron-fission characteristics to search the superasymmetric fission mode. As example, the comparison between the experimental and the theoretical fission-product yields for the ^{239}Pu and ^{245}Cm nuclei is shown in Fig. 3. As seen from Fig. 3, there is a pronounced structure on the mass curves at $A \approx 70$ and a change of the curve slope near A = 80, which can be explained by the stabilization due to the influence of the nuclear shells with Z = 28 and N = 50. It was also observed [23] that at A < 80 the mass yields (in per cents) from various fissioning systems are practically the same. The mass-curve proximity at A < 80 takes place over a broad fragment-mass interval and persists over a large range of mass yields (approximately three orders of the magnitude). Unlike the double-magic ^{132}Sn case, the two shells Z = 28 and N = 50 do not belong to the same fragment but are displaced from each other by approximately 12 masses, due to the extreme neutron richness of the ^{78}Ni cluster (N/Z = 1,786) as compared to the actinides studied at present (N/Z = 1,54 – 1,57). This displacement can be a reason for such a prolonged overlap in mass yields observed so far for all compound nuclei at Lohengrin. The proximity of the yield means that the formation probability of very light fragments is not sensitive to the difference in excitation energy, which appears from difference in the neutron-binding energy for compound nuclei under consideration. This implies that, most likely, such fragments are formed in a cold state and have nearly spherical shapes at the scission point. A physical quantity related to the fragments excitation and the deformation is the corresponding prompt-neutron multiplicity. Unfortunately no reliable experimental data on prompt-neutron emission in the superasymmetric mass region are available to make a definitive conclusion on this subject. The independence of the mass yield from the fissioning system, established in the superasymmetric mass region for thermal-neutron-induced fission, as well as the anomalous behavior of the fission-yield curve around A = 70, strongly supports the necessity of involving additional modes in the multimodal approach as introduced earlier in equation (3). The fission-mode parameters have been obtained for ^{244}Cm and ^{246}Cm compound nuclei [15] and the work is in progress for other actinides. Though the mode at A = 70 is narrow and its contribution to the total yield is small, the structure at A = 70 is important for understanding the superasymmetric fission dynamics.

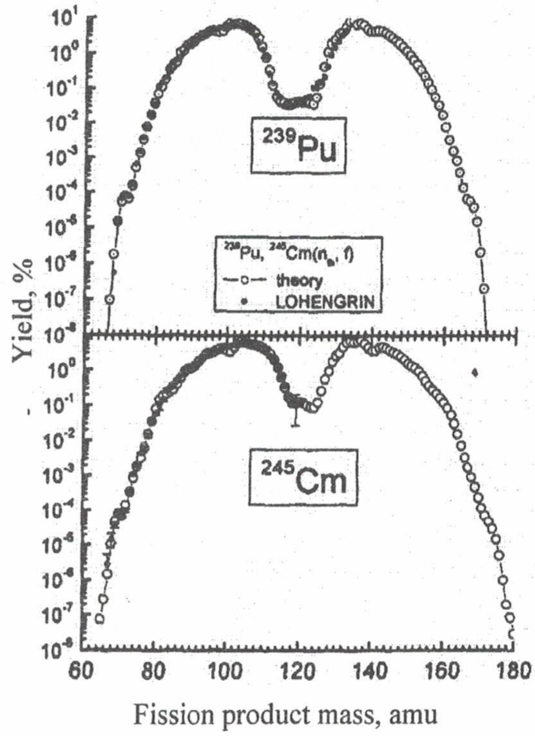

Fig. 3. Comparison between experimental Lohengrin data (solid points) and calculated (open circles) mass yields for the ^{239}Pu and ^{245}Cm targets.

We emphasize, see Ref. [15], that the fraction of the ^{78}Ni mode (SA1 and SA2) increases from ^{243}Cm to ^{245}Cm. The most surprising finding is, however, the disappearance of the standard-I (SI) mode in ^{243}Cm. Certainly this mode is too neutron excessive to be competitive in the fission of ^{243}Cm. A good description of the experimental yields of independent fission products was obtained, and hence, the validity of the model of a frozen quantal fluctuation in the calculations of the mean charge and charge dispersion of the isobaric chain was shown.

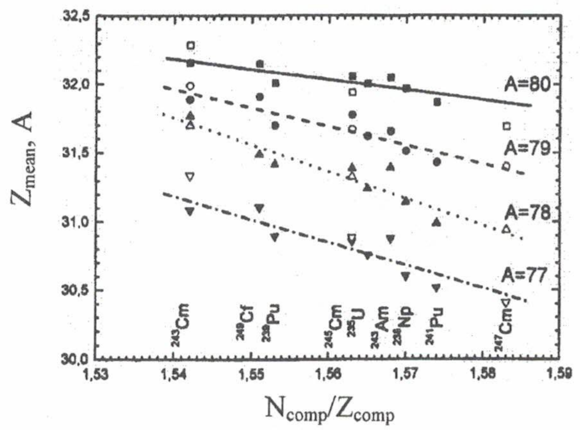

Fig. 4. Experimental (solid points) and calculated (open symbols) mean isobaric charge $\overline{Z}$ for masses A = 77 - 80, obtained from the thermal-neutron-induced fission of different nuclei, as function of the neutron-to-proton ratio of the corresponding compound systems (from Ref. [15]).

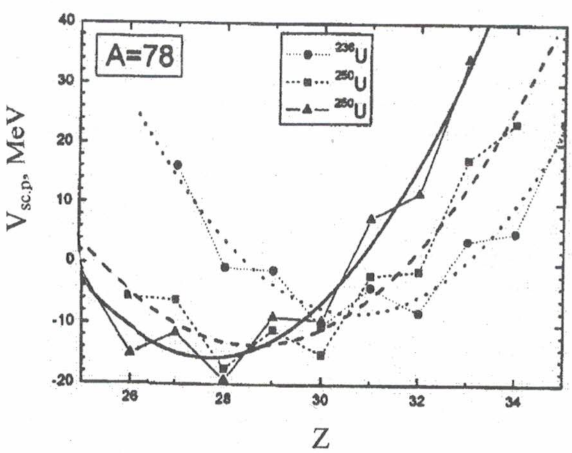

Fig. 5. Calculated potential energy at the scission point for pairs with the A = 78 fragment for ^{236}U, ^{250}U and ^{260}U.

The prediction of the yields of extremely-neutron-rich fission products, such as ^{78}Ni, is a challenging task. The systematic trends in the dependence of the mean charge in the A = 77 - 80 isobaric chains with using the Lohengrin experimental data have been investigated. In Fig. 4, the experimental mean-charge values for the A = 77 - 80 isobaric chains as function of the neutron-to-proton ratio of compound nuclei for the thermal-neutron-induced fission of targets from U to Cf are presented as solid symbols. As seen from Fig. 4, the dependence $\overline{Z}$ for a certain isobaric chain can be approximated by a straight line. The calculated $\overline{Z}$ values for the ^{244,246,248}Cm compound nuclei (open symbols) follow the experimental trend.

On basis of the overall good agreement between the theoretical evaluations and the experimental data (see Figs. 3 and 4) we could predict the yields of neutron-rich Ni isotopes. Independent yields of the neutron-rich Ni isotopes have been calculated from the fission of the ^{243,245,247}Cm targets induced by thermal neutrons [15]. It was shown that the ^{247}Cm(n_{th}, f) reaction is the most suitable for the production of nuclei with extreme neutron excess.

The data for A = 78 in Fig. 4 were approximated with the linear function $\overline{Z} = 61.8 - 19.5 N_c/Z_c$. From extrapolation of this dependence to the extremely neutron-rich U isotopes one finds that the ^{78}Ni fission mode ($\overline{Z} = 28$) will play an important role at $A_c \approx 250$. This result is supported by calculations of the potential energy at the scission point for $^{250,\,260}$U compound nuclei presented in Fig. 5. Nowadays these extremely neutron-rich nuclei are inaccessible for study in laboratories, but the fission of that type of nuclei can be expected in neutron stars.

In conclusion, the stabilization of the left wing of the light-mass peak in the superasymmetric mass region has been discovered in thermal-neutron-induced fission reactions at the Lohengrin mass separator. This feature of the mass curve, along with discontinuity in the yield disclosed at the mass A = 70 (Fig. 3), strongly supports the hypothesis on an important role of nuclear shells with Z = 28 and Z = 50 in the superasymmetric fission [24]. Contrary to the ^{132}Sn mode with Z = 50 and Z = 82, the influence of the Z = 28 and Z = 50 shells spans a wide range of masses, since any coherent action of the doubly magic ^{78}Ni mode is prohibited by a large deviation of its neutron excess from that of the compound nuclei close to the valley of stability. The multimodal fission approach and the model of frozen quantal fluctuations due to the charge asymmetry at the scission point were used for fission-product yield calculations. In order to describe the lightest slope of the mass peak, the two superasymmetric fission modes (at $A \approx 82$ and $A \approx 70$) were introduced to the calculation. The parameters of fission modes were extracted from the fit of the experimental mass yields in the thermal-neutron-fission measured at ILL with Lohengrin mass separator. It was found that the weight of the standard-I mode (^{132}Sn mode) for the ^{244}Cm compound nucleus is very small in comparison with that for the ^{246}Cm compound system. It is believed to be a common trend that the weight of the ^{132}Sn mode increases with the increasing neutron-to-proton ratio of a compound system N_c/Z_c. This feature was used for prediction of the ^{78}Ni yields in the reaction ^{247}Cm(n_{th}, f), which was shown as very promising for production of nuclei with the extreme neutron excess. The systematics obtained for the mean-product charge of the isobaric chains A = 77 - 80 for compound nuclei from ^{236}U to ^{250}Cf measured at Lohengrin demonstrates a clear decrease of $\overline{Z}$ as the ratio N_c/Z_c increases. This systematics is very useful for testing theoretical models and predicting the formation of very neutron-rich nuclides in the fission of nuclei far away from stability line (up to neutron drip line), which are supposed to be formed in different astrophysical processes. In the case of fission and quasi-fission of superheavy systems, the superasymmetric Ni fission mode is enhanced by influence of the Z = 82 and N = 126 nuclear shells in a heavy fragment. These results are important for the prediction of fission-product-formation cross sections in connection with different RNB projects.

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РОЛЬ ЯДЕРНИХ ОБОЛОНОК У СУПЕРАСИМЕТРИЧНОМУ ПОДІЛІ

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Для аналізу виходу продуктів поділу використано багатомодальне наближення для опису масових розподілів і метод оболонкових поправок для розрахунків параметрів зарядового розподілу ізобарних ланцюжків розпаду первинних уламків поділу. Дві додаткові вузькі моди поділу, пов'язані з впливом ядерних оболонок з магічними числами Z = 28 та N = 50, були введені для опису особливостей у масових розподілах при сильно асиметричному розділенні мас. Успіх запропонованої моделі підтверджується добрим описом експериментальних даних, отриманих для поділу актинідних ядер тепловими нейтронами. При порівнянні з відомими експериментальними результатами зроблено висновок, що внески поділових мод, які визначаються двічі магічними кластерами (^{132}Sn чи ^{78}Ni), залежать від відношення числа нейтронів до числа протонів складеного ядра поділу.

РОЛЬ ЯДЕРНЫХ ОБОЛОЧЕК В СУПЕРАСИММЕТРИЧНОМ ДЕЛЕНИИ

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Для анализа выходов продуктов деления использовано многомодальное приближение для описания массовых распределений и метод оболочечных поправок для расчета параметров зарядового распределения изобарных цепочек первичных осколков деления. Две дополнительные узкие моды деления, связанные с влиянием ядерных оболочек с магическими числами Z = 28 и N = 50, были введены для описания особенностей в массовых распределениях при сильно асимметричном разделении масс. Успех предложенной модели подтверждается хорошим описанием экспериментальных данных, полученных для деления актинидных ядер тепловыми нейтронами. Из сравнения с имеющимися экспериментальными результатами сделан вывод, что вклады делительных мод, которые определяются дважды магическими кластерами (^{132}Sn или ^{78}Ni), зависят от отношения числа нейтронов к числу протонов составного делящегося ядра.

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