Asymmetric and symmetric fission of excited nuclei of 180,190Hg and 184,192,202Pb formed in the reactions with 36Ar and 40,48Ca ions


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Asymmetric and symmetric fission of excited nuclei of $^{180,190}$Hg and $^{184,192,202}$Pb formed in the reactions with $^{36}$Ar and $^{40,48}$Ca ions


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Background: Observation of asymmetric fission of $^{180,190}$Hg has led to intensive theoretical and experimental studies of neutron-deficient nuclei in the lead region.

Purpose: The study of asymmetric and symmetric fission modes of $^{180,190}$Hg and $^{184,192,202}$Pb nuclei.

Methods: Mass-energy distributions of fission fragments of $^{180,190}$Hg and $^{184}$Pb formed in the $^{36}$Ar + $^{144,154}$Sm and $^{40}$Ca + $^{144,154}$Sm reactions, respectively, at energies near the Coulomb barrier have been measured using the double-arm time-of-flight spectrometer CORSET and compared with previously measured $^{192}$Pb isotopes produced in the $^{48}$Ca + $^{144,154}$Sm reactions. The mass distributions for $^{180,190}$Hg and $^{184,192,202}$Pb together with old data for $^{187}$Ir, $^{195}$Au, $^{198}$Hg, $^{201,205}$Tl, $^{205,207}$Bi, $^{210}$Po, and $^{213}$At [J. Nucl. Phys. 53, 1225 (1991)] have been decomposed into symmetric and asymmetric fission modes. The total kinetic-energy distributions for different fission fragment mass regions have been analyzed for $^{180,190}$Hg and $^{184}$Pb.

Results: The stabilization role of proton numbers at $Z \approx 36, 38, 45, 46$, and $Z = 28/50$ in asymmetric fission of excited preactinide nuclei has been observed. The high ($\approx 145$ MeV) and the low ($\approx 128$ MeV) energy components have been found in the total kinetic-energy distributions of $^{180,190}$Hg fission fragments corresponding to the fragments with proton numbers near $Z = 46$ and $Z = 36$, respectively. In the case of fission of $^{184}$Pb only the low-energy component ($\approx 135$ MeV) for the fragments with masses corresponding to the proton numbers $Z \approx 36$ and 46 has been found.

Conclusions: The studied properties of asymmetric fission of $^{180,190}$Hg and $^{184,192,202}$Pb nuclei point out the existence of well deformed proton shell at $Z \approx 36$ and less deformed proton shell at $Z \approx 46$.

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I. INTRODUCTION

Fission of atomic nuclei is a complex process of directed collective motion of nucleons characterized by a significant change in shape, rearrangement of the nucleon configurations, redistribution of the nucleus excitation energy between its various types (vibrational, thermal, etc.). To describe fission, Strutinsky proposed considering the potential energy of a fissionable nucleus as the sum of the liquid-drop (macroscopic) potential and the shell correction [1]. The shell correction, being an oscillating function of deformation [2,3], noticeably transforms the shape of the potential-energy surface.

For actinides, the macroscopic component of the potential energy of the fissionable nucleus is comparable with the shell correction ($\sim 5$–$7$ MeV), which results in a strong influence of shell effects in their fission. To date, the spontaneous and low-energy fission of actinides has been fairly well studied. It was found that they divide mainly asymmetrically with a heavy fragment mass of 142–144 u due to the proton and neutron shells [4–6]. The contribution of the symmetric mode increases with increasing excitation energy and becomes dominant at excitation energy greater than 40–50 MeV.

In the case of preactinides ($Z < 89$), the liquid-drop part of the potential energy of the fissioning nucleus is
significantly higher than the shell correction. Thus, the symmetric fission mode prevails. Until the end of the 1980s, all experimental data on preactinides were in good agreement with the liquid-drop model (LDM) calculations, assumed only one mass-symmetric fission valley \[7,8\]. Hence, the observation of asymmetric fission of \(^{181}\)Ir, \(^{195}\)Au, \(^{198}\)Hg, \(^{201}\)Tl, \(^{205,207}\)Bi, \(^{210}\)Po, and \(^{213}\)At \([9–11]\) was quite unexpected. The most pronounced asymmetric fission mode was found for nuclei with the fissility parameter \(x \approx 0.7\). To describe the mass distributions, the transition-state method was elaborated with an empirical positive shell correction for the mass-symmetric fission valley. This approach allowed to estimate the value of the shell correction of about 1 MeV to the fission barriers of preactinides but did not answer the questions concerning, first, the nature of the shell corrections and, second, the proton and neutron numbers playing a role in shell effects and the formation of fission fragments.

The nuclei in the regions of Ra \([12,13]\), Ac \([14,15]\), and the light isotopes of Th \([16–18]\) are transitional cases between symmetric and asymmetric modes of fission inherent to actinides. In this case the mass distributions are a superposition of symmetric and asymmetric modes with comparable contributions. The trace of asymmetric modes typical for fission of actinides have been observed starting from Po and At with a yield of about 0.1% \([19]\).

Recently, in experiments on the \(\beta\)-delayed fission of \(^{180}\)Tl \([20]\), an asymmetric mass distribution of the fission fragments of its daughter nucleus \(^{180}\)Hg with an excitation energy \(E^* < 10.8\) MeV was found. Note that the formation of two fragments—semimagic \(^{90}\)Zr \((N = 50, Z = 40)\)—should be expected in the symmetric fission of this strongly neutron-deficient nucleus. However, the formation of a light fragment with a mass of 80 u and a heavy one of 100 u was found most likely for \(^{180}\)Hg. The observation of the asymmetric fission of \(^{180}\)Hg \([20]\) has renewed interest in the study of the fission properties of nuclei lighter than lead both theoretically \([21–28]\) and experimentally \([29–36]\). However, there is still no unambiguous approach to understanding the fission of preactinides.

This paper is aimed at studying the asymmetric and symmetric nuclear fission modes in the lead region in dependence on the excitation energy and the nucleon composition of fissioning nuclei. The mass and energy distributions of fission fragments of \(^{180,190}\)Hg formed in the \(^{36}\)Ar + \(^{144,154}\)Sm reactions and \(^{184}\)Pb produced in the \(^{90}\)Ca + \(^{144}\)Sm reaction were measured using the double-arm time-of-flight spectrometer CORSET \([37]\). The previously measured fission fragments mass-energy distributions for \(^{192,202}\)Pb obtained in the \(^{48}\)Ca + \(^{144}\)Sm reactions \([38]\) were also included in our analysis.

The mass distributions of the fission fragments of \(^{180,190}\)Hg, \(^{184,192,202}\)Pb, as well as \(^{187}\)Ir, \(^{195}\)Au, \(^{198}\)Hg, \(^{201}\)Tl, \(^{205,207}\)Bi, \(^{210}\)Po, and \(^{213}\)At from Refs. \([9–11]\) were analyzed in the framework of a multimodal approach, successfully applied to describe the fission of actinides \([6,17,39]\). A generalized analysis of the data performed in this paper, as well as data from Refs. \([9–11]\), significantly expanded the studied region of nuclei and allowed us to determine more accurately the proton and neutron numbers that affect the asymmetric fission properties in the lead region. The properties of the studied reactions are given in Table 1.

### Table I. The reactions under study, \(E_{\text{lab}}\) : bombarding energy in the laboratory system; \((l)\) : mean angular momentum of the compound nucleus (CN); \(v_{\text{pre}}\) : prescission neutron multiplicity; \(E_{\text{pre}}\) : energy of prescission neutrons; \(B(l)\) : angular momentum dependent fission barrier \([40]\); \(E_{\text{CN}}^*\) and \(E_{\text{SP}}^*\) : excitation energies of the formed CN and fissioning nucleus at the saddle point.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>(E_{\text{lab}}) MeV</th>
<th>(E_{\text{CN}}^*) MeV</th>
<th>(E_{\text{pre}}) MeV</th>
<th>(v_{\text{pre}})</th>
<th>(B(l)) MeV</th>
<th>(E_{\text{SP}}^*) MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{36})Ar + (^{144})Sm (^{180})Hg</td>
<td>158</td>
<td>34.1</td>
<td>0</td>
<td>0</td>
<td>8</td>
<td>10.0</td>
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<tr>
<td></td>
<td>181</td>
<td>52.5</td>
<td>0.85</td>
<td>12.0</td>
<td>31</td>
<td>7.1</td>
</tr>
<tr>
<td>(^{36})Ar + (^{154})Sm (^{190})Hg</td>
<td>158</td>
<td>56.7</td>
<td>1.10</td>
<td>14.1</td>
<td>8</td>
<td>13.2</td>
</tr>
<tr>
<td></td>
<td>181</td>
<td>75.3</td>
<td>2.22</td>
<td>29.5</td>
<td>36</td>
<td>9.3</td>
</tr>
<tr>
<td>(^{40})Ca + (^{144})Sm (^{184})Pb</td>
<td>180</td>
<td>35.3</td>
<td>0</td>
<td>0</td>
<td>8</td>
<td>8.0</td>
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<tr>
<td></td>
<td>190</td>
<td>43.1</td>
<td>0.29</td>
<td>4.1</td>
<td>17</td>
<td>7.3</td>
</tr>
<tr>
<td>(^{40})Ca + (^{154})Sm (^{192})Pb</td>
<td>182</td>
<td>33.4</td>
<td>0</td>
<td>0</td>
<td>9</td>
<td>10.8</td>
</tr>
<tr>
<td></td>
<td>188</td>
<td>37.9</td>
<td>0</td>
<td>0</td>
<td>9</td>
<td>10.8</td>
</tr>
<tr>
<td>(^{40})Ca + (^{154})Sm (^{202})Pb</td>
<td>173</td>
<td>41.2</td>
<td>0.17</td>
<td>1.9</td>
<td>9</td>
<td>17.7</td>
</tr>
<tr>
<td></td>
<td>183</td>
<td>48.8</td>
<td>0.63</td>
<td>7.3</td>
<td>9</td>
<td>17.7</td>
</tr>
<tr>
<td></td>
<td>194</td>
<td>57.2</td>
<td>1.13</td>
<td>13.4</td>
<td>31</td>
<td>15.1</td>
</tr>
</tbody>
</table>

### II. EXPERIMENT

The measurements were performed using the U400 cyclotron at the Flерov Laboratory of Nuclear Reactions, Dubna, Russia. The 235-μg/cm\(^2\) \(^{144,154}\)Sm targets deposited on 30-μg/cm\(^2\) carbon backings were irradiated with the 180- and 190-MeV \(^{40}\)Ca and 158- and 181-MeV \(^{36}\)Ar beams. The target backings faced the beam. The details of measurements for the reaction with \(^{48}\)Ca ions are given in Ref. \([38]\). The energy resolution was \(\sim 1\%\). Beam intensities on targets were 1 to 2 pnA. The enrichment of the \(^{144,154}\)Sm targets was 93.8% and 98.9%, respectively.

The binary reaction products were measured in coincidence by the double-arm time-of-flight spectrometer CORSET \([37]\). Each arm of the spectrometer consists of a compact start detector and a position-sensitive stop detector based on microchannel plates. The angular acceptance of the spectrometer arms in the reaction plane was \(\pm 10^\circ\) and \(\pm 19^\circ\). In these experiments, the main attention was focused on detecting symmetric fragments formed in the complete fusion reaction. Thereby, according to the kinematics of the symmetric reaction fragments, the spectrometer arms were positioned symmetrically with respect to the beam axes at the angles \(\pm 60^\circ\) corresponding to \(90^\circ\) in the center-of-mass (c.m.) system. The position resolution of the stop detectors was 0.3 μm, and the time resolution was about 150 ps. The mass and energy resolutions of the CORSET setup were deduced from the full width at half maximum of the mass and energy spectra of elastic particles, respectively. The mass and total kinetic energy (TKE) resolution of the spectrometer under these conditions was \(\pm 2\) u and \(\pm 6\) MeV, respectively.

Data processing assumed standard two-body kinematics \([37]\). Primary masses, velocities, energies, and angles of...
FIG. 1. The mass-energy distributions of binary fragments formed in the $^{36}$Ar + $^{144,154}$Sm reactions leading to the formation of mercury ($Z = 80$) isotopes at energies near the Coulomb barrier.

reaction products in the c.m. system were calculated from the measured velocities and angles using the momentum and mass conservation laws, assuming that the mass of the composite system is equal to $M_{\text{target}} + M_{\text{projectile}}$. Corrections for fragment energy losses in the target material and the foils of detectors were taken into account. The extraction of the binary reaction channels exhibiting full momentum transfer was based on the analysis of the kinematical diagram (see Refs. [37,41] for details).

III. RESULTS

Mass-total kinetic energy ($M$-TKE) distributions of the primary binary fragments obtained in the $^{36}$Ar + $^{144,154}$Sm, $^{40}$Ca + $^{144,154}$Sm, and $^{48}$Ca + $^{144,154}$Sm reactions leading to the formation of $^{180,190}$Hg and $^{184,192,202}$Pb isotopes at energies close to the Coulomb barrier are shown in Figs. 1 and 2. In the $M$-TKE distributions, the reaction products with masses close to those of the projectile and target and energies around $E_{\text{cm}}$ are associated with elastic and quasielastic events and can be separated well enough from other reaction channels. The fissionlike products located between the quasielastic peaks within the contour lines in the $M$-TKE distributions in Figs. 1 and 2 are characterized by large mass transfer and energy dissipation and can originate either from CN fission or QF processes.

A key parameter used to assess QF probability is the mean fissility parameter $x_{m}$ [42]. It is expressed as $x_{m} = 0.75x_{\text{eff}} + 0.25x_{\text{CN}}$, where $x_{\text{CN}}$ is a fissility of CN and $x_{\text{eff}}$ is the effective fissility parameter [43] reflecting the entrance-channel mass and charge asymmetry. QF appears for reactions with $x_{m} > 0.68$ and results in widening of mass distributions and forward-backward asymmetry in angular distributions. The largest value of mean fissility parameter $x_{m} = 0.665$ is reached in the reaction $^{40}$Ca + $^{144}$Sm. This value is lower than the threshold value for QF appearance. So, all studied reactions are favorable for CN formation. For the $^{36}$Ar + $^{144,154}$Sm reactions, QF was not observed in the previous studies [29,33].

However, fusion suppression and the presence of asymmetric QF at energies near and below the Coulomb barrier have been observed for the reaction of $^{48}$Ca with the deformed $^{154}$Sm target [38]. In this case the asymmetric QF results in

FIG. 2. The mass-energy distributions of binary fragments formed in the $^{40}$Ca + $^{144}$Sm and $^{48}$Ca + $^{144,154}$Sm reactions leading to the formation of lead ($Z = 82$) isotopes at energies near the Coulomb barrier.
the asymmetric fragments with masses of 60–75 u for the light fragment and 127–142 u for the heavy one. For more symmetric fragments, the signatures of QF presence have not been observed. For the $^{48}$Ca$^{+}$144Sm reaction with a spherical 144Sm target, no evidence of QF has been found [38]. Therefore, we can expect that for all studied systems the fragments inside the contour lines in Figs. 1 and 2 originate from the CN-fission process, except for 48Ca$^{+}$154Sm. But even for this reaction, the fragments in the symmetric mass region 75–127 u are the products of the CN fission.

The excitation energies of formed CNs vary from 34 up to 75 MeV (see Table I). In this case the precission neutron and proton emissions have to be taken into account. The precission neutron multiplicities estimated using the systematics [44] are listed in Table I. The number of protons emitted before scission was estimated using NRV code [45]. For neutron-deficient $^{180}$Hg and $^{184}$Pb nuclei neutron evaporation is about three times more probable than proton one. For $^{192,202}$Pb and $^{190}$Hg the precission proton emission is found to be less than 1%. We neglected proton evaporation in our paper. Therefore, the actual fissioning isotope may differ from the CN in terms of the neutron number, especially at the highest excitation energies.

IV. ANALYSIS AND DISCUSSION

A. Mass distributions of fission fragments

The mass distributions of fission fragments of excited $^{180,190}$Hg and $^{184,192,202}$Pb (events inside the contour lines in the $E$-TKE distributions in Figs. 1 and 2) normalized to 200% are presented in Figs. 3 and 4. The mass distributions of $^{198}$Hg fission fragments [10] formed in the $^{197}$Au ($p,f$) and $^{194}$Pt ($\alpha,f$) reactions and $\beta$-delayed fission of $^{180}$Tl [20] are also shown in Fig. 3 for comparison.

The investigations of fission fragment angular distributions [46] show that the preactinides have very large deformations in the transition states (saddle point). According to the calculations within the different versions of the LDM [47–49], a dumbbell-like shape with a thin neck close to the scission shape is proper to these large deformations of fissioning nuclei. Therefore, we may expect that the influence of dynamic effects on the descent from fission barrier to scission point is comparatively small, and the fragment formation is generally defined by the deformation potential-energy landscape in the vicinity of the saddle point. This is opposite to the fission of actinides characterized by a long descent from the saddle to scission in which the dynamical effects may strongly affect the fission process.

Thus, the fission properties of preactinide nuclei are mainly determined at the saddle point and depend on its excitation energy in the simplest approximation defined as

$$E^{*}_{SP} = E^{*}_{CN} - B_{f}(l) - E_{pre},$$

where $B_{f}(l)$ is the angular momentum depended fission barrier and $E_{pre}$ is the energy loss due to precission neutron emission. Since the angular momenta introduced by the Ar and Ca projectiles are relatively large (see Table I), we used a fission barrier for the rotating liquid drop [40]. The
ground-state shell corrections are less than 2.5 MeV for the studied nuclei [50]. Moreover, they decrease with increasing CN excitation energy [51], and we neglected them in the fission barrier calculation. In proton- and α-induced fission, the angular momentum introduced to the CN is negligibly small. The values of the interaction energies, excitation energies of formed CNs, mean angular momenta of the CN calculated with the PACE4 code [52], fission barriers at these momenta and excitation energies at the saddle point for the studied reactions are listed in Table I.

It is revealed from Fig. 3 that the mass distributions of fission of neutron-deficient nuclei $^{180,190}$Hg have a pronounced asymmetric component even at rather high excitation energies. The positions of the maxima in mass distribution for $^{180}$Hg are the same as observed in the low-energy fission of $^{180}$Hg ($E^* < 10.8$ MeV) obtained as a daughter nucleus after β decay of $^{180}$Tl [20]. For the nucleus $^{198}$Hg located near the β-stability line, the fission has asymmetric mode at $E_{sp} = 7.4$ MeV, nearly disappearing at $E_{sp} = 26.2$ MeV, and the shape of mass distribution becomes close to a single Gaussian. For $^{184}$Pb, the mass distributions are flattopped in the symmetric mass region, whereas for the more neutron-rich $^{192,202}$Pb isotopes the mass distributions are close to a single Gaussian, although at $E_{sp} < 30$ MeV the deviations from a single Gaussian shape are observed (see Fig. 4).

In the analysis of fission of excited nuclei from $^{176}$Os to $^{206}$Pb [29,33], the mass distributions were proposed to be a sum of two Gaussians centered at complementary fragment masses, and the symmetric fission mode was not considered. However, in the case of excited nuclei, the contribution of the symmetric mode, described by the LDM, which increases with increasing the excitation energy of fissioning nucleus, has to be taken into account. The analysis of fragment mass distributions of fission of $^{205,207,209}$Bi at different excitation energies [34] showed that triple-Gaussian fits (a sum of symmetric and asymmetric modes) give a better agreement with experimental mass distributions than double-Gaussian fits (only one asymmetric mode).

Therefore, a question about the number of fission modes of preactinide nuclei arises. For instance, for actinides the four main fission modes were distinguished both theoretically and experimentally. According to the model of Brosa et al. [53], the modes are as follows: the superlong symmetric mode (S); the standard I (S1) mode caused by the influence of proton $Z = 50$ and neutron $N = 82$ shells; the standard II (S2) mode determined by deformed neutron shell with $N \approx 88$; the standard III (S3) superasymmetric mode caused by the influence of proton $Z = 28$ and neutron $N = 50$ shells; and the supershort mode (SS), which manifests itself only when both the light and the heavy fission fragments are close in their nucleon composition to the double magic tin with $A \approx 132$.

Based on the analysis of fragment mass yields from the fission of preactinides from $^{187}$Ir to $^{213}$At, located near the β-stability line, at low excitation energies at the saddle point ($E_{sp} \approx 7–13$ MeV) [54], the existence of two strongly deformed neutron shells in the formed fragments with $N \approx 52$ and $N \approx 68$ was supposed. However, it is impossible to explain the asymmetric fission for neutron-deficient nuclei by these neutron numbers. For instance, in the present paper and in Ref. [20] for $^{190}$Hg the maximum yields in mass distributions for the light and the heavy fragments are observed at 80 and 100 u. For $^{190}$Hg the maxima were found at 83 and 107 u. The same values of the most probable fragment masses for the asymmetric fission of $^{180,190}$Hg have been found in Ref. [29]. According to the simple assumption of the unchanged charge density of $N/Z$ equilibration [55], the most
The sensitivity of the variance to the angular momentum is much weaker, although not negligible. To estimate the $\partial \sigma_M^2/\partial l^2$ coefficient, we used similar systematics presented in Ref. [56].

The fission fragments mass distributions of nuclei from $^{187}$Ir to $^{213}$At can be described with one symmetric mode with Gaussian parameters given above, and two asymmetric modes associated with the proton shells at $Z \approx 36, 38$, and $44$.

It is to be noted that the detailed experimental study of low-energy fission of actinides [5,6] also revealed the stabilization role of proton numbers at $Z = 52$ for the S1 mode and $Z = 55$ for the S2 mode instead of $N = 82$ and 88 usually attributed to the S1 and S2 modes.

By analogy with actinides, we assume that the fission fragment mass distributions of the preactinide nuclei can be described as a sum of symmetric and several asymmetric modes, each asymmetric mode being a sum of two Gaussians centered at the complementary masses. The modes overlap, and, therefore, the variance of the symmetric mode should be fixed. Since we connect the symmetric mode with the macroscopic part of the potential energy to set its variance we use the empirical systematics based on the LDM [56].

The variance $\sigma_M^2$ increases linearly with $l^2$ and with the nuclear temperature $T$ and can be written as [56]

$$\sigma_M^2 = \frac{\partial \sigma_M^2}{\partial T} \cdot T + \frac{\partial \sigma_M^2}{\partial l^2} \cdot l^2. \quad (2)$$

The first term in Eq. (2) corresponds to the variance of the mass distribution at zero angular momentum and can be calculated (with some assumptions) as

$$\sigma_M^2(l = 0) = \frac{A_{CN}^2}{16} \cdot \left[ \left( \frac{d^2V}{d\eta^2} \right)_{\eta=0} \right]^{-1}. \quad (3)$$

where $A_{CN}$ is a mass number of the CN, $(d^2V/d\eta^2)_{\eta=0}$ is the stiffness of a nucleus at symmetric mass division ($\eta = 0$) and at zero angular momentum, which is derived from the corresponding systematics [56] for the temperature at the saddle point,

$$T = \sqrt{\frac{8.5E_{SP}/A_{CN}}{}}. \quad (4)$$

The experimental data in Refs. [9–11] were measured at different...
excitation energies of CNs. With increasing excitation energies, the shape of mass distribution becomes close to a single Gaussian with the parameters derived from the LDM at the saddle-point temperature $T_{\text{SP}} > 1.5 \text{ MeV}$.

Mass distributions of neutron-deficient CNs of Hg and Pb isotopes also demand to involve one more asymmetric fission mode connected with the influence of $Z \approx 50$ and $Z \approx 28$ proton shells ($A_3$ asymmetric mode) (see Figs. 3 and 4). This mode is observed in the excited fission of these nuclei as well as in the fission of $^{180}\text{Hg}$ at low excitation energy [20]. For the lead isotopes, the best fits of experimental data were found at $Z \approx 36$ for the $A_1$ mode and at $Z \approx 46$ for the $A_2$ mode. In this case the $A_1$ mode is complementary to the $A_2$ mode. As was mentioned above, the asymmetric fragments in the 60–75 u mass region for the light fragment and 127–142 u for the heavy one formed in the reaction $^{48}\text{Ca} + ^{154}\text{Sm}$ leading to the formation of $^{202}\text{Pb}$ are associated with the asymmetric QF process [38]. Therefore, it is impossible to distinguish the $A_3$ mode in the fission of $^{202}\text{Pb}$.

In Figs. 3–5 one can see that the modes $S$, $A_1$, $A_2$, and $A_3$ overlap to a rather significant extent. Therefore, the description of the resulting mass distributions depends on the width of the Gaussian corresponding to the symmetric mode. Since the estimation of proton and neutron numbers of fission fragments is based on the unchanged charge density assumption, the precission neutron evaporation may also add uncertainties to the obtained results of about 0.4 u. Moreover, the experimental resolution of our mass measurements is about ±2 u. For these reasons, we estimate the accuracy of proton numbers for the $A_1$–$A_3$ modes found at the fitting procedure of mass distributions as ±1 u, at least.

**B. TKE distributions of fission fragments**

The kinetic energy of fragments originates mainly from the Coulomb repulsion at the scission point. In the fission of actinides, the TKE values of the distinct modes were found to differ significantly [4]. The difference between the TKEs of the distinct modes can reach 20–25 MeV. We assume that in the fission of preactinides, the asymmetric modes can manifest themselves also in the TKE of fragments. In Fig. 6 the TKE distributions of fragments for different mass ranges for the fission of $^{180,190}\text{Hg}$ and $^{184}\text{Pb}$ at the lowest measured excitation energies are shown. The top panels show the TKE distributions for symmetric mass split, the middle panels—for the mass range where the yield of asymmetric fragments is maximal, the bottom panels—for the range where the $A_3$ mode prevails according to the mass decompositions (see Figs. 3 and 4).

It is seen from Fig. 6 that the TKE distributions have a complex structure and change significantly with the fragments mass. We connect the $S$ mode with the LDM valley. The most probable TKE value for this mode can be estimated using the Viola systematics [58]. According to the LDM, the TKE has a parabolic dependence on fragment mass,

$$
\text{TKE}(M) = 4 \text{TKE}_{\text{Viola}} \frac{M(A_{\text{CN}} - M)}{A_{\text{CN}}}.
$$

where $M$ is a fragment mass, $\text{TKE}_{\text{Viola}}$ is the most probable TKE.

In the case of $^{180}\text{Hg}$, when the yield of the $S$ mode is minimal compared to the $^{190,198}\text{Hg}$ (see Fig. 3), the TKE distribution for the symmetric mass split has a pronounced
TABLE II. The TKEs of the symmetric (S) and the asymmetric A1 (Z \approx 36), A2 (Z \approx 46), and A3 (Z = 28/50) fission modes for the studied reactions at the lowest measured energies.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>CN</th>
<th>S</th>
<th>A1</th>
<th>A2</th>
<th>A3</th>
</tr>
</thead>
<tbody>
<tr>
<td>36Ar + 144Sm 180Hg</td>
<td>142 ± 2</td>
<td>127 ± 2</td>
<td>145 ± 2</td>
<td>132 ± 2</td>
<td></td>
</tr>
<tr>
<td>36Ar + 154Sm 190Hg</td>
<td>142 ± 2</td>
<td>129 ± 2</td>
<td>146 ± 2</td>
<td>134 ± 2</td>
<td></td>
</tr>
<tr>
<td>40Ca + 144Sm 184Pb</td>
<td>149 ± 2</td>
<td>135 ± 2</td>
<td>135 ± 2</td>
<td>142 ± 2</td>
<td></td>
</tr>
</tbody>
</table>

For this reaction the A1 and A2 modes are complementary to each other.

The TKE values of the fission modes are listed in Table II.

The TKE of fragments depends on the distance d between the centers of the formed fragments at scission,

\[ TKE[\text{MeV}] = 1.44 \frac{Z_1 Z_2}{d[\text{fm}]} \tag{6} \]

where \( Z_1 \) and \( Z_2 \) are the proton numbers of formed fragments. Thus, using the Eq. (6) we may estimate the elongation of the fissioning nucleus at the scission point for different modes. In the case of the fission of \( 180\text{Hg} \) at \( E_{c.m.} = 23.9 \text{ MeV} \) the following values of distance have been found: for the S mode –16.2 fm, the A1 mode –18.0 fm, the A2 mode –15.5 fm, and the A3 mode –16.2 fm. Consequently, the shape at scission for the A2 mode is more compact than the LDM one, whereas for the A1 mode the shape is more elongated.

Recently it was found that the symmetric scission in the light thorium isotopes shows a compact configuration [18]. In this case, the number of protons is \( Z = 45 \) for both fragments that corresponds to the compact A2 mode proposed in our paper. Therefore, our results are consistent with the observed features of neutron-deficient Th nuclei, and the A2 mode may probably manifest itself in the fission of actinides.

In the fusion reaction \( ^{36}\text{Ar} + ^{154}\text{Sm} \), leading to the formation of \( ^{190}\text{Hg} \), the lowest measured energy of 158 MeV is close to the Coulomb barrier \( (E_{c.m.}/E_C = 0.98) \) that corresponds to the CN excitation energy of 56.7 MeV. Unfortunately, the measurements of fission of \( ^{190}\text{Hg} \) at lower energies in this reaction are rather difficult due to the low counting rate. However, even at this relatively large excitation energy, the TKE distributions have a structure similar to the one observed for \( ^{180}\text{Hg} \) (see Fig. 6), whereas the contribution of the S mode is significantly higher. As it follows from Table II, the obtained values of mean TKE are similar to those that we found for \( ^{180}\text{Hg} \) for all fission modes.

Contrary to the fission of \( ^{180}\text{Hg} \) at similar excitation energy where the pronounced asymmetric component is observed, in the case of \( ^{184}\text{Pb} \), the mass distribution for symmetric fragments is flattened due to the dominance of symmetric mode. It is seen from Fig. 6 that the main parts of TKE distributions for the different mass ranges may be attributed with the S mode, described by the LDM. For symmetric mass range of 86–98 u, the low-energy component with TKE \approx 135 MeV together with the S mode is observed. For fragment masses with maximal asymmetric yield (76–86 u), the yield of the low-energy component increases, whereas the high-energy component attributed to the A2 mode is not found. The absence of the high-energy component may be explained in a way that in the fission of Pb isotopes the A1 mode is complementary to the A2 mode (see the mass decomposition in Fig. 4). For the mass range of 50–70 u, the main component is the A3 mode with TKE \approx 142 MeV.

Thus, the analysis of the TKE distributions of fission fragments for \( ^{180,190}\text{Hg} \) and \( ^{184}\text{Pb} \) confirms our assumption of the existence of several asymmetric modes in the fission of preactinide nuclei, namely, A1 and A2 connected with proton numbers \( Z \approx 36 \) and \( Z \approx 46 \), respectively, and the A3 mode caused by the proton shells at \( Z = 28 \) and/or \( Z = 50 \). The TKE values for these modes significantly differ.

C. Discussions and comparison with theory and other experimental data

The proton numbers of fission fragments, deduced in this paper for the A1 and A2 asymmetric modes, are shown as a function of the neutron numbers in Fig. 7. It is seen that for the A1 mode the proton numbers are close to \( Z = 36 \) for the fragments with less than 53 neutrons and about 38 for fragments with more than 53 neutrons. For the A2 mode \( Z = 46 \) for \( N < 64 \) and \( Z = 45 \) for \( N > 64 \).

As was mentioned above, according to the calculations of Wilkins et al. [2], the deformed proton shells at \( Z \approx 38 \) (\( \beta_2 \approx 0.32–0.42 \)), \( Z \approx 44 \) (\( \beta_2 \approx 0.50–0.58 \)), and \( Z \approx 36 \) (\( \beta_2 \approx 0.67–0.77 \)) are expected. Recently, asymmetric fission has been studied theoretically in the sublead region using microscopic mean-field calculations of fission based on the Hartree-Fock approach [23]. Its origin is attributed to the influence of neutron shell gaps at \( N = 52, 56 \) for compact octupole deformations as well as shell gaps associated with large quadrupole deformations at \( Z = 34 \) and \( Z = 42–46 \) protons.

The analysis of experimental mass distributions of fission fragments of 14 nuclides between \( ^{176}\text{Os} \) and \( ^{208}\text{Pb} \) [33] with double-Gaussian fits shows that the deduced proton centroids are \( Z = 36–37 \) for the light fragments and 43–44 for the heavy ones. In the study of mass-asymmetric fission of \( ^{205,207,208}\text{Bi} \) [34], it was found that the light fragments have \( Z \approx 58 \) and \( N \approx 56–58 \) and the heavy fragments have \( Z \approx 45 \) and \( N \approx 66–68 \).
Thus, the proton numbers obtained in this paper, which probably affect the asymmetric fission of preactinide nuclei, are close to those found in other experimental studies. The different theoretical approaches also predict similar proton numbers. In the present analysis, the stabilization role of different theoretical approaches also predict similar proton numbers close to those found in other experimental studies. The proton numbers obtained in this paper, which probably affect the asymmetric fission of preactinide nuclei, are close to those found in other experimental studies. The different theoretical approaches also predict similar proton numbers.
nucleus. For $^{180, 190}$Hg the contributions of symmetric mode
are lower than those for Pb isotopes. $^{198}$Hg and $^{202}$Pb, located
near the $\beta$-stability line, undergoes mainly symmetric fission
at the saddle point excitation energies higher than $20$ MeV.

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