Evidence for a New Compact Symmetric Fission Mode in Light Thorium Isotopes

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Taking benefit of the $R3B$/SOFIA setup to measure the mass and the nuclear charge of both fission fragments in coincidence with the total prompt-neutron multiplicity, the scission configurations are inferred along the thorium chain, from the asymmetric fission in the heavier isotopes to the symmetric fission in the neutron-deficient thorium. Against all expectations, the symmetric scission in the light thorium isotopes shows a compact configuration, which is in total contrast to what is known in the fission of the heavier thorium isotopes and heavier actinides. This new main symmetric scission mode is characterized by a significant drop in deformation energy of the fission fragments of about 19 MeV, compared to the well-known symmetric scission in the uranium-plutonium region.

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The nuclear fission process has been thoroughly studied since its discovery in 1938, and the very first fission description, based on the liquid drop model [1], was proposed only one year after its discovery. Considering the fissioning nucleus as a charged liquid drop, the liquid drop model successfully explained why the competition between the repulsive Coulomb force and the attractive surface energy, causes heavy nuclei to deexcite through fission. However, this approach failed to reproduce the experimentally observed mass asymmetry of the two fission fragments in the fission of actinide nuclei. The observed asymmetric mass distributions could only be understood when models started to include microscopic shell corrections [2], resulting in the appearance of asymmetric valleys in the potential energy surface (PES), characterizing the energy of the deforming nucleus on its way to scission. The structure of the nascent fission fragments plays a major role in the description of these asymmetric paths and scission configurations. Experimentally, to characterize these configurations, the fission fragment mass was combined with an additional observable: either the total kinetic energy (TKE), or its anticorrelated observable, the prompt-neutron multiplicity (ν_{tot}). The bulk of the energy released in fission is passed on to the fission fragments at scission, either as kinetic or excitation energy, the latter is released by neutron evaporation and to a lesser extent, by γ-ray emission. Since the deformation energy of the nascent fragments at scission is converted into excitation energy of the primary fragments, the more deformed the scission configuration is, the higher the ν_{tot} becomes. In the opposite situation, a more compact scission configuration results in higher Coulomb repulsion and, correspondingly, higher TKE and lower neutron multiplicity. Data on fission fragment masses correlated to TKE and $\langle\nu_{\mathrm{tot}}\rangle$ describes the fission process as a competition between different fission modes [3–5]. Each fission mode corresponds to a given path in the PES, reaching a specific scission configuration. For the near stable actinide region, three main fission modes have been proposed. A first asymmetric mode, the so-called standard I (ST1) mode, is mainly governed by the doubly magic shell closure around ^{132}Sn, leading to an almost spherical heavy fragment. A second asymmetric mode, referred as the standard II (ST2) mode, is characterized by a heavy fragment stabilized around $Z = 54$ [6,7], which has been recently related to a proton shell in octupole-deformed fragments [8]. Finally, a last path leads to two, on average, mass-symmetric fission fragments, which are both highly deformed. This fission mode is often referred to as superlong (SL). The latter is characterized by a larger prompt-neutron emission and a lower TKE value, compared to the asymmetric modes. This SL mode prevails at higher excitation energies of the fissioning nucleus. One important exception to this symmetric mode is the sudden transition from asymmetric fission in the spontaneous fission (sf) of ^{256}Fm to symmetric fission in ^{258}Fm(sf) with an abnormally high TKE [9,10]. This high-TKE mode is explained as a very compact mode caused by the influence of the fragment shell structure near the doubly magic ^{132}Sn, possible only for fermium isotopes with masses larger than 257. Another transition from asymmetric to symmetric fission has been observed along the thorium isotopes, which is not yet fully explained [6]. In this Letter, we report on a dedicated study, probing the evolution of scission configurations along this transition.

Experiments correlating information on yields to the TKE or the ν_{tot}, in order to study the scission configuration, are scarce. This statement holds particularly true for exotic fissioning systems where the availability of targets is a strong limitation. Therefore, a new generation of experiments, based on inverse kinematics coupled with a recoil spectrometer, has been developed. At GANIL, transfer- and fusion-induced reactions of a ^{238}U beam on ^{12}C are used to populate fissioning nuclei from ^{238}U up to ^{250}Fm [11]. From this experiment, the scission configuration of ^{240}Pu was studied in detail [12]. At the GSI facility, a pioneering experiment based on inverse kinematics was developed in the 1990s [6]. There, electromagnetic excitation is exploited to induce nuclear fission of radioactive isotopes at relativistic energies. At such high kinetic energies, fission fragments are fully stripped, allowing for their unambiguous identification in terms of their nuclear charge over the full fission-fragments range. This experiment highlighted the previously mentioned transition along the thorium chain. However, since only nuclear charges were measured, the limited number of observables make this transition difficult to be characterized precisely.

The $R3B$/SOFIA experiment at GSI, also based on electromagnetic induced fission of actinides at relativistic energies, studied fission of several compound nuclei, including some thorium isotopes. For the first time, both fission fragments were identified in coincidence, in nuclear charge, and in mass [13,14]. The results obtained confirmed the transition from asymmetric fission to symmetric fission seen in the measurement of the elemental yields and complete the previous data with the isobaric and isotonic yields [14]. In addition, the average total prompt-neutron multiplicity ($\langle \nu_{\mathrm{tot}} \rangle$) could also be measured as a function of the nuclear charge asymmetry. Indeed, since the masses of the compound nucleus (A_{CN}) and fission fragments (A_{FF_1} and A_{FF_2}) are measured on an event by event basis, the total prompt-neutron multiplicity (ν_{tot}) is trivially obtained by ($\nu_{\mathrm{tot}} = A_{\mathrm{CN}} - A_{\mathrm{FF}_1} - A_{\mathrm{FF}_2}$). For each pair of nuclear charges (Z_{FF_1},$90 - Z_{\mathrm{FF}_1}$), the total prompt-neutron emission probability is measured as shown in Fig. 1. The mean value of each distribution gives $\langle \nu_{\mathrm{tot}} \rangle (Z)$. The negative component is caused by the experimental uncertainty in the measurement of the mass number. The correlation of the nuclear charge asymmetry with the mean neutron multiplicity, both obtained with a good accuracy, is sufficient to provide us with a new insight in the transition from asymmetric to symmetric fission in the thorium isotopes. The results are shown in Fig. 2 for the nuclides ^{222,226,230}Th and compared to the results obtained for the fission of ^{236}U [15] in a $R3B$/SOFIA experiment performed in 2014. Fission in ^{230}Th(γ, f) behaves similarly to ^{236}U(γ, f), however the evolution of the prompt-neutron multiplicity at symmetry towards the light thorium isotopes is striking. Indeed in Ref. [5], the calculated neutron multiplicity related to the SL mode is stable over a large range of fissioning isotopes, almost 20 mass units wide. For fission of ^{232}Th, ^{227}Ac, and ^{213}At, the calculated values of $\langle \nu_{\mathrm{SL}} \rangle$, 5.7, 5.4, and 4.9, respectively, show a weak dependence on the mass of fissioning isotopes. The opposite is observed experimentally, with a significant decrease of $\langle \nu_{\mathrm{sym}} \rangle$ by 2.7 neutrons over 8 mass units, from ^{230}Th ($\langle \nu_{\mathrm{tot}} \rangle_{\mathrm{sym}} = 6.2$) to ^{222}Th ($\langle \nu_{\mathrm{tot}} \rangle_{\mathrm{sym}} = 3.5$). Taking into account the effective neutron binding energies in the fragments of these systems (S_n around 8 MeV) and their kinetic energy ($\langle E_n \rangle$ around 2 MeV), this drop translates to a loss of 27 MeV in the available excitation energy of the fragments for the symmetric split.

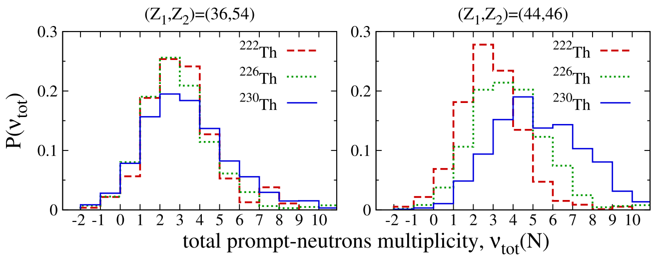

FIG. 1. Prompt-neutron emission probability for two different combinations of fission fragment charge pairs produced by electromagnetic induced fission of ^{222,226,230}Th, respectively, in dashed red line, dotted green line, and full blue line.

These results must be linked to the excitation energy distribution of the compound nuclei which cannot be measured by our setup. However, we could estimate it, as shown in Fig. 3, based on the total electromagnetic differential cross-section calculation and the fission probability obtained from the general description of fission observables model (GEF-2019.V1.1 [16]). Actually, a decrease of the prompt-neutron multiplicity is expected at symmetry, since the distribution of excitation energies

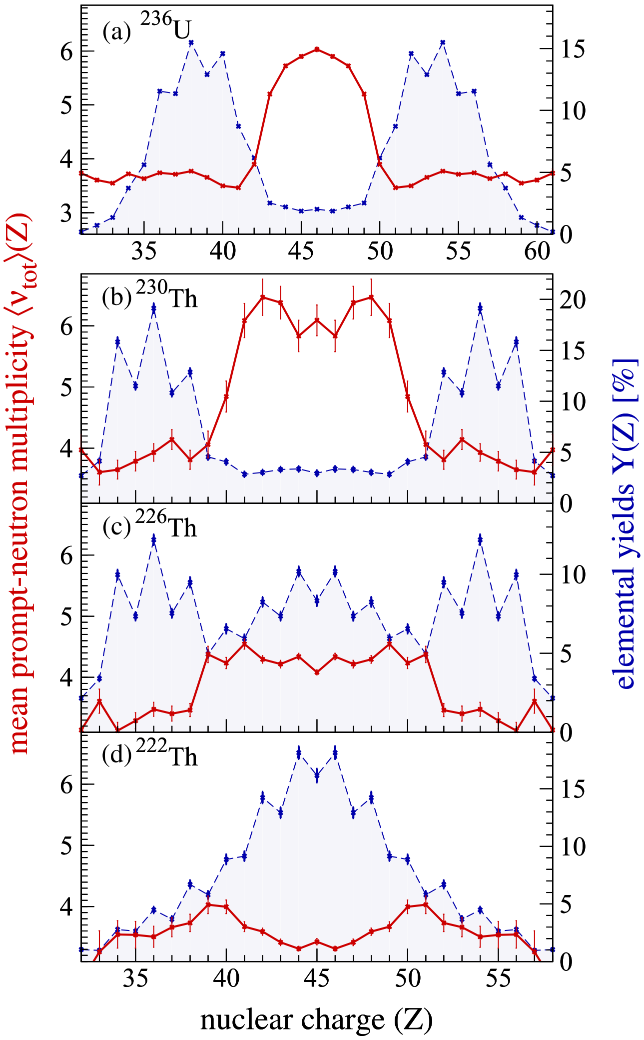

FIG. 2. For the ^{236}U(γ,f) [(a) [15]] and ^{230,226,222}Th(γ,f) [(b)–(d)] the mean prompt neutron multiplicities measured at SOFIA as a function of the nuclear charge of the fission fragments are represented with the red full lines and correspond to the left y axis. The elemental yields are represented with the blue dashed lines and the shadowed areas and correspond to the right y axis.

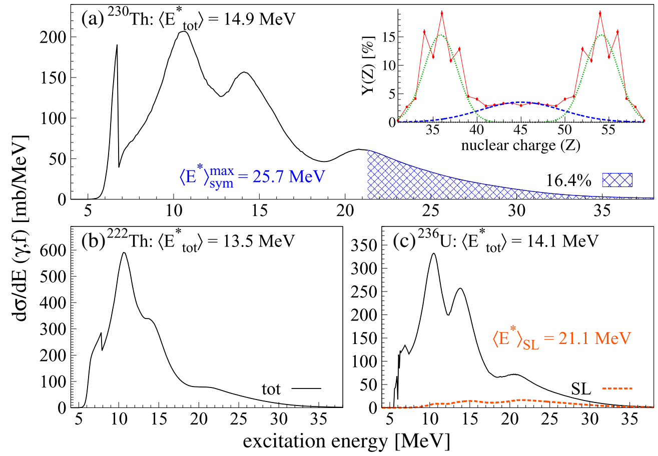

FIG. 3. The calculated excitation functions are represented with black full lines for the ^{230}Th(γ,f), ^{222}Th(γ,f) and ^{236}U(γ,f) reactions in (a), (b), and (c), respectively. The blue hatched area in (a) represents the upper-16.4% part of the ^{230}Th(γ,f) excitation function. The orange dashed line in (c) shows the SL component obtained from GEF for ^{236}U(γ,f). Additionally, the elemental yields measured with SOFIA for the ^{230}Th(γ,f) [14] reactions are reproduced in the inset of (a) in order to illustrate the three-Gaussian fit, from which the asymmetric components (green dotted line) and the symmetric one (blue dashed line) are extracted.

leading to symmetric scission, has a lower mean value for the ^{222}Th(γ,f) reaction than for ^{230}Th(γ,f) (see Fig. 3). The question is whether this change of mean excitation energy can explain the 27 MeV difference observed in the fragments excitation, or not. Since the fission of ^{222}Th is predominantly symmetric [see Fig. 2(d)], the mean excitation energy of the symmetric contribution $\langle E^*_{^{222}\mathrm{Th}}\rangle_{\mathrm{sym}}$ can be considered equal to 13.5 ± 0.5 MeV, the average value of the full distribution [reproduced in Fig. 3(b)]. In contrast, as previously discussed, symmetric fission of ^{230}Th corresponds to the SL mode, dominantly fed by the high energy part of the excitation energy spectrum. In order to evaluate the value of $\langle E^*_{^{230}\mathrm{Th}}\rangle_{\mathrm{sym}}$ we take two approaches: a crude approximation which will provide an upper limit, and a more refined calculation based on GEF [16].

First, as illustrated in the inset of Fig. 3(a), the elemental yields are fitted with three Gaussian functions to extract the light, symmetric, and heavy components. Since the symmetric splitting represents $16.4 \pm 1.0\%$ of the fission events, an extreme upper limit of $\langle E^*_{^{230}\mathrm{Th}}\rangle^{\mathrm{max}}_{\mathrm{sym}}$ is found to be 25.7 ± 0.3 MeV by using the upper 16.4% of the excitation function. This, in turn, means that the change in excitation energy between ^{222}Th and ^{230}Th is around 12.2 ± 0.6 MeV.

A second and more realistic calculation is performed based on the GEF code which reproduces the elemental yields along the uranium chain [16]. A reliable value of the mean excitation energy of the ^{236}U compound nucleus for the SL mode is therefore obtained from GEF at $\langle E_{^{236}\mathrm{U}}\rangle_{\mathrm{SL}} =$ 21.1 MeV [see Fig. 3(c)]. This value can be considered close to the one of ^{230}Th for the same fission mode, given that both systems, ^{230}Th and ^{236}U have similar prompt neutron multiplicity at symmetry. From this calculation, we deduce that the change in excitation energy between ^{222}Th and ^{230}Th is 7.6 ± 0.5 MeV.

Both approaches lead to the same conclusion, however only the realistic value is retained in the following: the change of 7.6 MeV in excitation energy is not at all sufficient to explain the 27 MeV decrease of the excitation energy of the fission fragments produced at symmetry from ^{230}Th to ^{222}Th. The weight of the multichance fission was estimated by GEF. It increases from 10% in ^{222}Th to 30% in ^{230}Th, with a small contribution for the latter case to the symmetric scission. This negligible bias in the prompt-neutron multiplicity measurement can not account for the significant drop observed at symmetry.

Therefore in the neutron-deficient thorium isotopes, fission fragments are produced at symmetry with lower deformation than in the one closer to stability. As symmetric fission in heavier thorium isotopes can be described by the SL mode, another path leading to more compact symmetric scission configurations appears in the PES and prevails for the fission of the lighter thorium isotopes. The smooth evolution of the observables along the thorium chain (see Figs. 1 and 2) shows that the PES valley corresponding to this compact symmetric mode becomes gradually favored compared to valleys governed by shell effects leading to asymmetric modes. The fission of the ^{226}Th represents an intermediate case between heavy thorium isotopes for which the minor symmetric fission corresponds to the SL mode, to those, where compact symmetric fission dominates. In that sense, this evolution is also different from the one observed in the fermium isotopes, which is sudden and caused by the influence of the strong shell closures at $(Z = 50, N = 82)$ in both fission fragments. Following these results, a theoretical work has been initiated [17] to study the effect of the tensor term in the thorium chain, using the Gogny force D1ST2a [18] built as a standard D1S interaction plus a finite range tensor force. They found that the tensor term is responsible for the appearance of a symmetric valley in the (Q_2, Q_4) PES. This new valley becomes more dominant as the neutron number decreases and reaches scission at lower Q_2 value than the SL mode.

The coincident measurement of the yields and the total prompt-neutron multiplicity permits us to probe the scission configuration of the different modes along the thorium isotopic chain. Whereas the fission for the ^{230}Th isotope is similar to that observed in the fission of the heavier actinides, this is not the case for the lighter thorium isotopes. The dramatic fall of the prompt-neutron multiplicity for the symmetric configuration is a clear signature that this fission mode is compact in contrast to the symmetric SL mode. A new scission mode, compact at symmetry, has thus been observed for the light thorium isotopes, and is characterized by a loss in deformation energy of about 19 MeV.

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[1] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
[2] V. M. Strutinsky, Nucl. Phys. **A95**, 420 (1967).
[3] U. Brosa, Phys. Rev. C **38**, 1944 (1988).
[4] F.-J. Hambsch, H.-H. Knitter, and C. Budtz-Jørgensen, Nucl. Phys. **A491**, 56 (1989).
[5] U. Brosa, S. Grossmann, and A. Müller, Phys. Rep. **197**, 167 (1990).
[6] K.-H. Schmidt, S. Steinhäuser, C. Böckstiegel, A. Grewe, A. Heinz, A. R. Junghans, J. Benlliure, H.-G. Clerc, M. de Jong, J. Müller, M. Pfützner, and B. Voss, Nucl. Phys. **A665**, 221 (2000).
[7] C. Böckstiegel, S. Steinhäuser, K.-H. Schmidt, H.-G. Clerc, A. Grewe, A. Heinz, M. de Jong, A. R. Junghans, J. Müller, and B. Voss, Nucl. Phys. **A802**, 12 (2008).
[8] G. Scamps and C. Simenel, Nature (London) **564**, 382 (2018).
[9] E. K. Hulet *et al.*, Phys. Rev. Lett. **56**, 313 (1986).
[10] D. C. Hoffman, Nucl. Phys. **A502**, 21 (1989).
[11] M. Caamaño, F. Farget, O. Delaune, K.-H. Schmidt, C. Schmitt *et al.*, Phys. Rev. C **92**, 034606 (2015).
[12] M. Caamaño and F. Farget, Phys. Lett. B **770**, 72 (2017).
[13] E. Pellereau, J. Taïeb, A. Chatillon, H. Alvarez-Pol, L. Audouin *et al.*, Phys. Rev. C **95**, 054603 (2017).
[14] A. Chatillon, J. Taïeb, H. Alvarez-Pol, L. Audouin, Y. Ayyad *et al.*, Phys. Rev. C **99**, 054628 (2019).
[15] L. Grente (private communication).
[16] K.-H. Schmidt, B. Jurado, C. Amouroux, and C. Schmitt, Nucl. Data Sheets **131**, 107 (2016).
[17] R. N. Bernard, N. Pillet, L. M. Robledo, and M. Anguiano, Phys. Rev. C **101**, 044615 (2020).
[18] M. Anguiano, M. Grasso, G. Co ’, V. De Donno, and A. M. Lallena, Phys. Rev. C **86**, 054302 (2012).