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Isoscaling studies of fission: A sensitive probe into the dynamics of scission

M. Veselsky,[1,2,*] G. A. Souliotis,[2] and M. Jandel[1]
[1]*Institute of Physics of the Slovak Academy of Sciences, Dubravska 9, Bratislava, Slovakia*
[2]*Cyclotron Institute, Texas A&M University, College Station, Texas 77843, USA*

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The fragment yield ratios were investigated in the fission of ^{238,233}U targets induced by 14 MeV neutrons. The isoscaling behavior was typically observed for the isotopic chains of fragments ranging from the proton-rich to the most neutron-rich ones. The observed high sensitivity of neutron-rich heavy fragments to the target neutron content suggests fission as a source of neutron-rich heavy nuclei for present and future rare ion beam facilities, allowing studies of nuclear properties towards the neutron drip line and investigations of the conditions for nucleosynthesis of heavy nuclei. The breakdowns of the isoscaling behavior around $N=62$ and $N=80$ manifest the effect of two shell closures on the dynamics of scission. The shell closure around $N=64$ can be explained by the deformed shell. The investigation of isoscaling in the spontaneous fission of ^{248,244}Cm further supports such conclusion. The Z dependence of the isoscaling parameter exhibits a structure which can be possibly related to details of scission dynamics. The fission isoscaling studies can be a suitable tool for the investigation of possible new pathways to synthesize still heavier nuclei.

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I. INTRODUCTION

Nuclear fission, first observed in 1938 [1,2], has been investigated extensively for many decades [3–5]. The model description of scission ranged from the early statistical model of Fong [6] to the advanced statistical model of Wilkins [7] and, more recently, to the dynamical description of the random-neck rupture model of Brosa [4]. Despite the obtained level of understanding, there are still many open questions and challenges remaining (for an overview see, e.g., Ref. [8]). Fissionlike phenomena have been observed recently also in metallic clusters [9]. Typically, nuclear fission produces a wide range of fragments with different atomic and mass numbers. Similar wide mass and charge distributions of reaction products can be observed in nuclear reactions leading to the production of hot nuclei which deexcite by emission of charged fragments. It has recently been observed [10] that for two similar reactions occurring at the same temperature, which differ only in the isospin asymmetry, the ratio $R_{21}(N,Z)$ of the yields of a given fragment (N,Z) exhibits an exponential dependence on N and Z of the form $R_{21}(N,Z)\propto\exp(\alpha N+\beta Z)$, this scaling behavior being termed isoscaling [10]. Initially, the investigations of isoscaling focused on the yields of light fragments up to oxygen, originating from the massive hot systems produced at intermediate energies [10], or high energies [11]. The isoscaling behavior was attributed to the difference of statistical deexcitation of two massive hot systems with different isospin asymmetry. In recent papers [12,13], the investigation of isoscaling is reported using the heavy residue data from the reactions of 25 MeV/nucleon ^{86}Kr projectiles with ^{124}Sn, ^{112}Sn and ^{64}Ni, ^{58}Ni targets. The global isoscaling behavior is observed for heavy residues originating from the most damped collisions, while the dependence of the isoscaling parameters α and β on deposited excitation energy can be used to extract information on the level of N/Z equilibration between the projectile and target, ranging from no equilibration at the most peripheral collisions to full equilibration at the most damped collisions. The investigation of isoscaling on reconstructed hot quasiprojectiles with mass $A=20-30$ from the reactions of 30 and 50 MeV/nucleon ^{28}Si with ^{124,112}Sn targets was reported in Ref. [14]. The observed dependence of the isoscaling parameter on excitation energy of the quasiprojectile was independent of the projectile energy and the resulting difference of inclusive isoscaling parameters at different projectile energies reflects different excitation energy distributions. Furthermore, due to the known level of N/Z equilibration [14], the temperature dependence of the isoscaling parameter was used to extract information on the onset of chemical separation. In general, the isoscaling studies appear to be a rather global method to investigate nuclear processes, manifesting the response of the system to the variation of the isospin asymmetry. The exponential scaling is typically a signal that some degree of equilibrium was reached.

It is of interest to further explore the isoscaling across the wide range of nuclear processes, even the most traditional ones explored in most detail. In this study, we present the results of an isoscaling analysis of the fragment yield data from fission induced by fast neutrons. The evaluated independent fragment yields from the fission of ^{233,238}U targets induced by 14 MeV neutrons [15] were used.

II. ISOSCALING ANALYSIS OF FISSION FRAGMENT YIELDS

In Fig. 1, we present the ratios of the fragment yields from the fission of ^{238,233}U targets induced by 14 MeV neutrons, obtained using the recommended independent fission fragment yields from the evaluated nuclear data file ENDF-349 [15]. The recommended yields are obtained as a result of

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[*] Electronic address: fyzimarv@savba.sk

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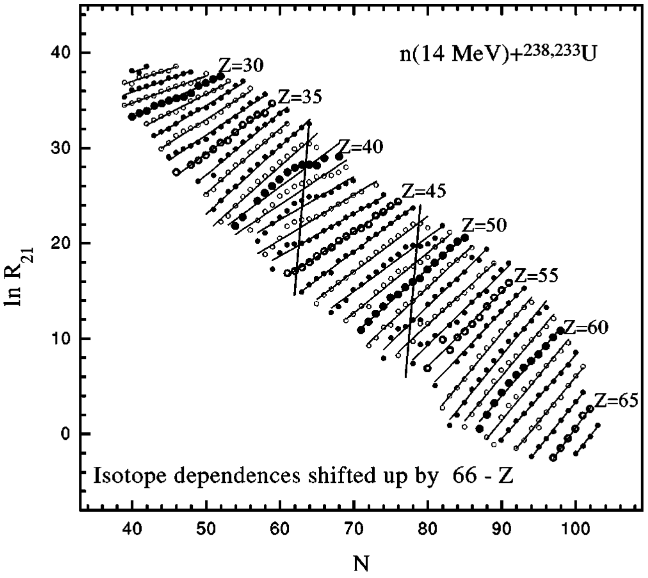

FIG. 1. Ratios of the fragment yields from the fission of ^{238,233}U targets induced by 14 MeV neutrons [15]. The data are shown as alternating solid and open circles. The labels apply to the larger symbols. The lines represent exponential fits. For clarity, the R_{21} dependences are shifted from element to element by one unit. Nearly vertical lines mark major isoscaling breakdowns.

the evaluation procedure and comprise both the fission yield measurements reported in the literature and the calculated Gaussian extrapolations of charge distributions for given masses. Despite limitations, such data possess a predictive power worth to explore, partially also due to the immensity and diversity of the existing experimental data. The study was restricted to recommended independent yields larger than 10^{-5}%.

In Fig. 1, one can observe, in the majority of the cases, a remarkable isoscaling behavior of isotopic chains with more than ten members. As mentioned above, some of the last data points are the results of extrapolation using the calculated Gaussian charge distributions. Nevertheless, since the evaluations have been done on a case-by-case basis, independent for each mass (and not for each element), the continuation of the trend they represent is possibly related to the physical behavior of the system without being excessively biased by the evaluation procedure. The available neutron multiplicity data from photofission of ^{238}U at comparable excitation energy [16] show that the fragments typically emit only two neutrons on average and thus the effect of emission is rather limited. Thus the observed behavior in Fig. 1 may imply that the isoscaling behavior is mainly determined by the dynamical evolution of the system prior to scission.

Of special interest for investigation are the mass regions corresponding to the most asymmetric mass splits. Practically all fragments below Zn exhibit weak dependence of R_{21} on the neutron number, strongly contrasting the enhanced sensitivity of R_{21} for their partners, the heaviest observed fission fragments. Such a weak dependence may imply a higher temperature of the scissioning light prefragment, since the isoscaling parameter is typically, within the grand-canonical approximation, considered inversely proportional to the temperature ($\alpha=\Delta\mu_n/T$, $\Delta\mu_n$ being the difference of neutron chemical potentials of the two fragmenting systems). It is, however, not *a priori* obvious why such grand-canonical formula could be applied to fission. On the other hand, one can arrive at a formula of similar structure using the nucleon transport theory appropriate for fission. Typically the process of fission can be divided into two phases. The fissioning system must first overcome the saddle point (the peak of the fission barrier), thus entering the irreversible path towards scission. In the second stage, the properties of the scission configuration are determined during the long descent from the saddle to the scission configuration. The process of fission (as well as the closely related process of deep-inelastic nucleus-nucleus collision) can be described in terms of collective motion using the transport theory [17–20]. The dynamics of the collective degrees of freedom is typically described using the Langevin or Fokker-Planck equation. From a practical point of view, the isoscaling occurs when the two mass distributions for a given Z from two processes with different isospin are Gaussian distributions of the same width with different mean masses. Isotopic yield distributions can be, according to transport theory, considered as the solutions of the Fokker-Planck equation (Gaussian mass distributions). A necessary condition for isoscaling, the assumption of equal width (typically a sign of equal temperatures) leads in the context of the Fokker-Planck equation to equal values of the diffusion coefficients. Then the isotopic dependence of the yield ratio R_{21} will assume the form

$$R_{21}(N) \propto \exp\left(\frac{V_{N2}-V_{N1}}{2D_{NN}}N\right), \tag{1}$$

where $V_{N1,2}$ are the drift coefficients characterizing the two processes with different isospin and D_{NN} is the common diffusion coefficient. The transport theory typically complies with the fluctuation-dissipation theorem $V_N T=D_{NN}F_N$ which after expressing the driving force as $F_N=\Delta\mu_{nab}$ [18] leads to the following expression for the isoscaling coefficient

$$\alpha = \Delta(\Delta\mu_{nab})/2T, \tag{2}$$

where $\Delta\mu_{nab}$ is the difference of neutron chemical potentials of interacting nuclei a,b determining the drift velocity. Thus apparently the isoscaling behavior is consistent with statistical transport theory. Within the statistical transport theory, the two interacting nuclei (fission fragments) can both be treated statistically using a grand-canonical approach [18], with two different temperatures, with the stochastic force depending in the general case on both temperatures. The temperature T thus possibly can be related to the Fermi gas temperature of the fragments in the given isotopic chain. In the photofission of ^{238}U, the observed neutron multiplicities for the lightest and heaviest fragments observed (about two neutrons on average [16] for $A\approx 90$ and $A \approx 150$) correspond to different fragment temperatures, higher temperature for the lightest fragments and lower for the heaviest fragments. In Fig. 1 the light fragments typically exhibit weak logarithmic slopes while their heavy partners exhibit much larger logarithmic slopes. Equation (2), based on the transport theory, suggests that the logarithmic slopes (isoscaling parameters) can indeed be related to temperature and excitation energy, thus possibly establishing an analogy with Ref. [16]. A quantitative test of Eq. (2) would be helpful in order to establish such analogy.

The high sensitivity of neutron-rich heavy fragments to the neutron content of the fissioning system, combined with observation from Ref. [16] that the multiplicity of neutrons emitted from heavier fragment is less dependent on photon energy, is of considerable interest for the production of neutron-rich nuclei, since it suggests that neutron-rich heavy fragments can be created in the fission of neutron-rich uranium isotopes, thus leading to the production of neutron-rich nuclei which are of crucial importance for future rare isotope beam facilities, e.g., Refs. [21–23]. From a nuclear reaction standpoint, a promising candidate for the production of very neutron-rich fissile nuclei would be the peripheral collision of heavy fissile nuclei at energies between the Coulomb barrier and the Fermi energy [24] leading to the moderately excited heavy uranium-like nuclei which would further fission into neutron-rich fragments. Some encouraging results have been obtained already using a ^{238}U beam at 20 MeV/nucleon [25,26]. Of special interest for future experiments is the reaction of ^{238}U beam with targets exhibiting the effect of neutron skin, such as ^{64}Ni and ^{124}Sn [27,28] or ^{208}Pb and ^{238}U.

Apart from the typically regular isoscaling behavior across the periodic table, the isoscaling behavior is violated in some regions, most prominently around $N=62$, around $N=80$, and possibly around $N=86-88$. The violation of the isoscaling in the regions $N=80$ can be possibly related to the neutron shell closure around $N=82$. The isoscaling behavior around another strong neutron shell closure around $N=50$ does not seem to exhibit irregularities exceeding the statistical ones. The breakdown of isoscaling around $N=62$ can not be directly related to the effect of any spherical shell closure in the final fission fragments. On the contrary, in this region the nuclei are supposed to be typically deformed. However, according to the fission model of Wilkins *et al.* [7], it may be related to the effect of the deformed neutron shell at $N=64$ with quadrupole deformation $\beta_{quad}=0.6$ (point C in Fig. 1 of Ref. [7]) which plays a crucial role in the dynamics of scission. In a similar manner the violation of isoscaling in the region $N=80$ and possibly $N=86-88$ can be related to points G ($N=82$ and $\beta_{quad}=0$) and H ($N=88$ and $\beta_{quad}=0.65$). The smooth dependence for $Z=50$ can be explained by the influence of spherical proton shell $Z=50$, again in accord with the conclusions of Ref. [7]. Thus the observed isoscaling behavior appears to be generally consistent with the model calculations of Wilkins *et al.* concerning the scission configuration which allows to assume that it may reflect the real properties of the scission configuration. On the other hand, the probability of a given fission channel can be considered as a function of the scission configuration as a whole. Within the grand-canonical picture of isoscaling which describes deexcitation phenomena [10], the effect of binding energy (and thus of the ground state shell correction) of the final product is canceled out and only the properties of the whole excited system are reflected. In analogy, since the scission configuration is essentially binary, one can assume that the irregularities exhibited for final fragments around some neutron number may be caused by the shell structure of the other fragment. Then the observed structure around $N=62$ can be possibly related to the effect of $N=82$ shell closure in the other fragment. However, in the same sense the observed structure around $N=80$ should not be attributed to the shell closure around $N=82$ but to the presence of shell closure around approximately $N=64$ of the other fragment. Thus, again the observed behavior points to the presence of the dominant shell closures in the scission configuration, one around $N=82$ and another one around $N=62-64$. The former one can be either the ground state spherical shell closure or the deformed shell closure as calculated by Wilkins *et al.* [7]. The latter shell closure can be only deformed (as calculated by Wilkins *et al.* [7]). In any case the mass of the fissioning systems ^{234,239}U causes that the two observed effects are exhibited by complementary fission fragments.

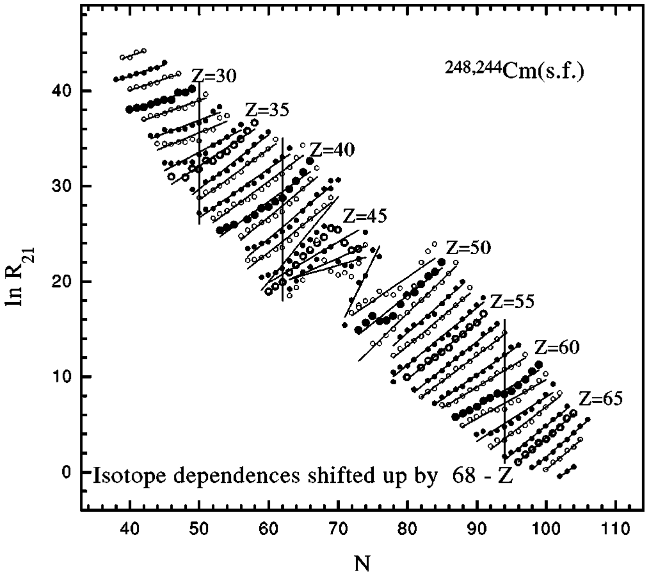

FIG. 2. Ratios of the fragment yields from the spontaneous fission of ^{248,244}Cm [15]. The data are shown as alternating solid and open circles. The labels apply to the larger symbols. The lines represent exponential fits. For clarity, the R_{21} dependences are shifted from element to element by one unit. Vertical lines mark major isoscaling breakdowns.

In order to separate the two effects, a logical step is to continue the analysis by investigating a fissioning system of different mass, for practical reasons heavier. The systematic fission data for nuclei heavier than uranium are rather scarce, one nevertheless can find a good candidate in evaluated fragment yield data from spontaneous fission of heavier nuclei such as ^{248,244}Cm [15]. The spontaneous fission is a much colder process where additional phenomena such as barrier penetration probability and potential energy surface can play a crucial role, it is nevertheless interesting to explore it in the context of isoscaling. The isoscaling plot from the spontaneous fission of ^{248,244}Cm is shown in Fig. 2. In the spontaneous fission of ^{248,244}Cm, the overall isoscaling behavior can be observed from the most populated region of mass distributions towards asymmetric mass splits. The breakdowns featuring rather the slope change than the structures observed in the previous case are observed around $N=50$ and $N=62$. A significant effect is observed around $N=68-70$ (with complementary fragment around $N=82$), which nevertheless can be a sign of transition into symmetric region around $N=74-76$ where the behavior is rather irregular. Further the isoscaling behavior is quite regular up to the heaviest fragments (with the exception around $N=94$ where a hint of isoscaling breakdown can be observed). It is remarkable that the breakdown of isoscaling (slope change) is again observed around $N=62$ (along with $N=50$) which possibly points to the role of shell structure, which may influence the potential energy surface and thus the probability of a given fission channel. While around $N=50$ the shell structure can be identified with spherical fragment, for the region around $N=62$ the influence of the deformed shell is the most plausible. The observed isoscaling behavior in the spontaneous fission of ^{248,244}Cm, despite the more complex dynamics of the cold process, again appears to point to the role of the deformed shell closures (in particluar around $N=64$) in the fission of actinides.

The above isoscaling analysis was carried out using the recommended fission fragment yields obtained as a compilation of the existing fission fragment yield data complemented with model based extrapolations. The uncertainty of the extrapolated data depends on the detailed extrapolation procedure and its magnitude is not expressed quantititively. It is of interest to establish whether the observed behavior could be influenced by the uncertainty of the extrapolation. Since both the overall isoscaling behavior and the irregularities in particular regions around the neutron numbers $N=62$ and $N=80$ are observed across many isotopic chains with values of yields ranging across several orders of magnitude, one can conclude that the observed behavior corresponds to the details of the fission process rather than to the uncertainty of the data.

For the fission of ^{238,233}U targets induced by 14 MeV neutrons, we present in Fig. 3(a) the systematics of the isoscaling parameters α, obtained by exponential fits of the isotope chains, as a function of the atomic number Z. One line was fitted for each Z (see Fig. 1). As already discussed, the isoscaling parameter α decreases below $Z=32$, while one can observe an increase above $Z=58$, thus possibly signaling a relatively hot light fragment and a colder heavy fragment in the asymmetric fission channels. In the central part of the fragment mass distribution, one can observe an increase up to the maximum around $Z=38$, then an abrupt discontinuity around $Z=40$ and a linear increase above. According to formula (2), the decrease of the isoscaling parameter α around $Z=42$ signals rather hot (and possibly initially deformed) fragments which is a reasonable result when considering that the second fragment for such a mass split is in the vicinity of $Z=50$ proton shell and it can be expected to be spherical and possibly colder, as one can conclude from the increase of values of α around $Z=50$. This is again consistent with the model calculations of Wilkins *et al.* [7] which obtained a decrease of distance of centers for corresponding mass splits (see Fig. 27 of Ref. [7]). Also the recent results from ternary fission suggest that typically the low energy light charged particle in the spontaneous fission of ^{252}Cf is correlated with a ^{132}Sn fragment and a strongly deformed light complementary fragment at scission [29,30], which may deexcite by secondary emission of the low energy ternary particle. One can also attempt to relate the discontinuous Z dependence of the isoscaling parameter α to the sawtooth dependence of the neutron multiplicity on fragment mass [16,31,32], which motivated the model of random neck rupture [4]. However, in the partially complete data from the photofission of ^{238}U [16], the minimum in the neutron multiplicity, signaling the position of the discontinuity, appears at the mass approximately $A=130$ rather than at masses $A=100-108$, corresponding approximately to $Z=42$. The way for detailed comparison of the isoscaling parameters α with the neutron multiplicity, determined in Ref. [16], is not trivial, since there the measured neutron multiplicity for a given fragment pair was divided between the fragments according to the model calculation. In any case, the observed discrepancy deserves further investigations.

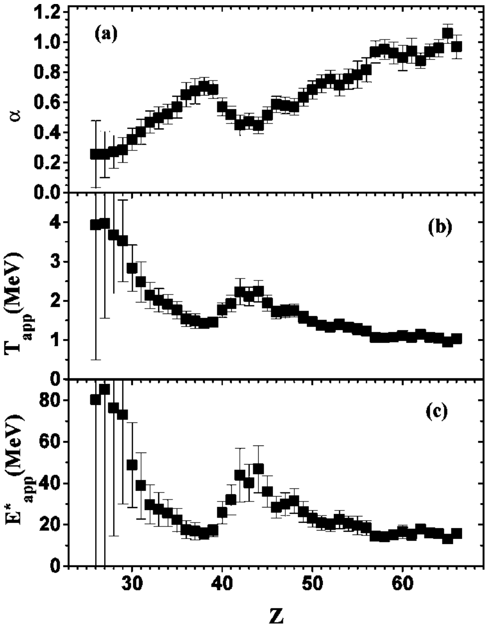

FIG. 3. The systematics of (a) isoscaling parameter α in the fission of ^{238,233}U targets induced by 14 MeV neutrons (b) estimates of apparent fragment temperatures obtained using Eq. (2) (for details see text), and (c) corresponding fragment excitation energies as a function of the atomic number Z.

As a further step in the isoscaling analysis, we test the potential of the Eq. (2), derived from transport theory, to provide quantitative information on the properties of the scission configuration. In Figs. 3(b) and 3(c) we present an estimate of the apparent fragment temperatures obtained using Eq. (2) and of the corresponding excitation energies. The quantity $\Delta(\Delta\mu_{nab})$ was approximated using the difference of neutron separation energies of two composite systems. Such approximation can be based on the assumptions that $\Delta(\Delta\mu_{nab})$, reflecting the difference of neutron transfer driving forces for two fissioning systems, will track with the difference of neutron chemical potential of the two composite systems. Thus we assume that the average driving force for a given fissile system is a property of the whole system and that its relative change between two fissiong systems is equal to the difference of chemical potentials, which can be roughly approximated by their neutron separation energies. However rough such approximation is, it provides an interesting quantitative test of Eqn. (2) and thus also of the underlying theory. As one can see in Figs. 3(b) and 3(c), the extracted values of the apparent temperature and excitation energy indeed lead in some mass regions to reasonable agreement with the experimental neutron multiplicities of photofission of ^{238}U [16] at comparable excitation energies. Specifically for the most asymmetric splits ($A \approx 90$ vs 150, corresponding roughly to $Z \approx 36$ vs 56 in the present work) observed in Ref. [16] approximately two neutrons are emitted from each fragment, thus suggesting the fragment excitation energies of about 15–20 MeV. For more central mass splits some similarity of trends can be observed, especially the excitation energy increase with increasing mass of the light fragment. The observed rapid increase of estimated temperatures and excitation energies for the lightest fragments in Figs. 3(b) and 3(c) may be caused by statistical uncertainties. While the above estimate is indeed rough, it nevertheless suggests a possibility to study details of the fission dynamics, e.g., to map the transfer driving force using the fragment temperatures measured independently. The specific observation that the estimated fragment temperatures are different for complementary light and heavy fragments can be understood naturally within the transport theory, where the number of transfers in both directions is typically comparable, thus leading to comparable excitation energies, which in the case of very asymmetric mass splits lead to a hotter light fragment and colder heavy fragment. Such a situation is routinely observed in peripheral mass-asymmetric nucleus-nucleus collisions (see, for example, Ref. [33]), where the light quasiprojectile can get very hot while the quasitarget remains relatively cold.

In general, the isoscaling analysis of the fission data appears to be a sensitive tool to investigate the fission dynamics. While it can not substitute the traditional experimental investigations of fission, its simplicity can prove useful in situations where such studies are not possible. For instance, as suggested in Ref. [34], the knowledge of the fission dynamics of superheavy nuclei is essential for understanding of the possibility to synthesize still heavier nuclei. In this aspect, the isoscaling analysis of reactions with two targets, possibly isotopes of the same element, can allow investigations of the fission dynamics of very heavy nuclei, using either a designated experimental setup or radiochemical methods. From a theoretical point of view, the capability to reproduce the isoscaling behavior can be a simple but, nevertheless, crucial test of any fission model. Also, as shown in the above analysis, the isoscaling properties can provide essential information on the possibility to produce heavy neutron-rich nuclei which are essential to understand the nuclear properties in general and, in addition, the astrophysical processes leading to the nucleosynthesis of the heaviest elements.

III. SUMMARY AND CONCLUSIONS

In summary, the fragment yield ratios were investigated in the fission of ^{238,233}U targets induced by 14 MeV neutrons. The isoscaling behavior was typically observed for isotopic chains ranging from the most proton-rich to most neutron-rich ones. The high sensitivity of the neutron-rich heavy fragments to the target neutron content suggests the viability of fission (possibly following a peripheral collision with another neutron-rich nucleus) as a source of very neutron-rich heavy nuclei for future rare ion beam facilities, thus allowing studies of the nuclear properties of such nuclei and exploration of the conditions for the nucleosynthesis of heavy nuclei. The observed breakdowns of the isoscaling behavior around $N=62$ and $N=80$ indicate the effect of two major shell closures on the dynamics of scission, one of them being the deformed shell closure around $N=64$. The isoscaling analysis of the spontaneous fission of ^{248,244}Cm further supports such conclusion. The values of the isoscaling parameter appear to exhibit a structure which can be possibly related to details of scission dynamics, e.g., for the charge split $Z=42$ vs $Z=50$. The isoscaling studies present a suitable tool for investigation of the fission dynamics of the heaviest nuclei, which can provide essential information about possible pathways to the synthesis of still heavier nuclei.

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