A key role of multi-chance fission for the description of fission-fragment mass distributions at high energies

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Fission-fragment mass distributions were measured for 235−240U, 239−242Np and 241−244Pu populated in the excitation-energy range from 10 to 60 MeV by multi-nucleon transfer channels in the reaction 19O+238U at the JAEA tandem facility. Among them, the data for 240U and 240,241,242Np were observed for the first time. It was found that the mass distributions for all the studied nuclides maintain a double-humped shape up to the highest measured energy in contrast to expectations of predominantly symmetric fission due to the washing out of nuclear shell effects. From a comparison with the dynamical calculation based on the fluctuation-dissipation model, this behavior of the mass distributions was unambiguously attributed to the effect of multi-chance fission.

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At present, about 11% of the world’s electricity is produced by thermal-neutron-induced fission in nuclear power reactors. Management of nuclear waste, and in particular, of long-lived minor actinides produced in these reactors, is one of the most important issues in the use of nuclear power. For further public acceptance of nuclear power, it is essential to reduce the already-existing and newly-produced nuclear waste. The use of accelerator-driven systems (ADS), for example [1], is considered as one of the viable options for the incineration and/or transmutation of the long-lived minor actinides into shorter-lived fission products. In the ADS approach, energetic spallation neutrons, produced via high-energy proton impact on a heavy target material such as lead and/or bismuth, could be used to irradiate the fissionable minor actinides. This leads to fission with higher, and more broadly distributed, excitation energies in comparison to those in the thermal-neutron-induced fission in a traditional power reactor. Thus, understanding of fission at high excitation energy is important for nuclear-data evaluations related to ADS developments.

The fission process is usually described as an evolution of a nuclear shape on a potential-energy surface, resulting from the interplay of macroscopic nuclear properties and microscopic shell effects. The shape of fission-fragment mass distributions (FFMDs) is directly influenced by nuclear shell effects, a well-known example being the asymmetric FFMD in the thermal-neutron-induced fission of 235U, whereby the compound nucleus, 236U, fissions at the excitation energy of 6.55 MeV. The asymmetric FFMD, in this case, is attributed to the influence of strong shell effects in the fission fragments in the vicinity of doubly-magic 132Sn.

With increasing excitation energy, two competing processes are expected to occur. First of all, due to a reduced importance of shell effects, the transition to predominantly symmetric (liquid-drop) type fission should occur, which is indeed demonstrated by many experiments [2]. The other process is multi-chance fission (MCF), or fission after consecutive neutron evaporations, where the fissioning nuclei with less neutrons will have lower excitation energy, thus showing stronger shell effects than in the initial compound nucleus. The latter effect is then supposed to favor the asymmetric fission of typical actinides after neutron evaporation. The MCF concept itself is well-known from studies of the fission probability in high-energy neutron-induced reactions, whereby step-like behavior is observed in the fission cross-sections at the energies corresponding to 1n, 2n, . . . neutron emission (see for example Fig. 17 in [3]). It was also reported that the effects of MCF can be seen in the average total kinetic energy [4, 5], and in the average energy of the prompt fission neutrons [6], as a function of the excitation energy of the compound nuclei. In contrast to these
In this Letter, we present our investigation of the effects of MCF on FFMDs, by measuring the mass distributions in a wide range of nuclides and excitation energies, using the novel experimental method recently developed at JAEA [11]. By exploiting multi-nucleon transfer (MNT) channels in the reaction $^{18}$O+$^{238}$U, FFMDs of twelve isotopes of O, Np, and Pu were obtained in the excitation energy range of $E^* = 10-60$ MeV, some of which cannot be populated by other experimental methods. A persistence of predominantly asymmetric FFMDs was observed for all the studied nuclides. To understand this behavior, the fluctuation-dissipation model was used. It was shown that a reliable understanding of the observed FFMDs can be obtained only by invoking MCF.

The experiment was performed at the JAEA tandem accelerator facility using a 157.5 MeV $^{18}$O beam with an intensity of 0.5 nA. The target was prepared by electrodeposition of an 80 $\mu$g/cm$^2$ layer of $^{238}$U on a 90 $\mu$g/cm$^2$ nickel backing. The experimental setup, consisting of a multi-detector $\Delta$E-E silicon telescope and four multi-wire proportional counters (MWPCs) for fission observables, to our knowledge, no experimental study of the effects of MCF on mass distributions has been reported to date. It was only recently that the effect of MCF on mass distributions was introduced in theoretical studies [7–10]. However, the validity of the calculated FFMDs for each fission chance was not shown because of the lack of experimental data. Thus, an elaborated and well justified interpretation of experimental FFMDs at high excitation energies has not been yet established.

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of 6\%, 3\% and 2\% on average, for $^{16-19}$O, $^{14-17}$N and 
$^{12-15}$C, respectively. There were two cases with a less 
evident separation, i.e., $^{19}$O ($^{237}$U) and $^{18}$O ($^{238}$U) at 
$E^*=50$–$60$ MeV, where the admixture of neighboring iso-
topes was as high as $\sim 25\%$. To obtain the correct FFMD 
of $^{238}$U, the FFMD of the major contaminant $^{239}$U (23\%) 
was subtracted from the initially derived FFMD for $^{238}$U. 
The data for $^{237}$U was obtained in the same manner, by 
subtracting the contribution of $^{238}$U (25\%) which is the 
only background source. In the analysis, it was found 
that the change in the FFMDs by the background cor-
rection remained within the statistical errors and did not 
alter the shapes of the FFMDs. Hence, the background 
subtraction was not applied to the rest of the data. 

The momentum of the recoiling compound-nucleus, 
which should be shared by both fragments in fission, was 
determined from the energy $E_{\text{tot}}$ and the direction of 
the ejectile. The excitation energy of the compound nu-
cleus was then deduced from the recoil momentum, $E_{\text{tot}}$, 
and the reaction $Q$-value [13]. It is assumed that no ex-
citation energy is given to the ejectile, thus the excita-
tion energies quoted in this study should be considered 
as the upper limit. The measured resolution for $E_{\text{tot}}$ is 
$\sim 1.0$ MeV (FWHM), which determines the uncertainty of 
the excitation energy of a compound nucleus. Coincident 
fission fragments produced in MNT fission were detected 
by using position-sensitive MWPCs, which allow deter-
mination of the directions of the fission fragments. The 
time of flight difference between two fragments was mea-
sured to determine the pre-neutron-emission masses. 

As an example of benchmarking of the method and new 
data, in Fig. 2 FFMDs for (a) $^{237}$U ($-1n$ transfer) and 
(b) $^{239}$U ($+1n$ transfer) from our experiment are com-
pared with the existing data [11, 12]. The closed cir-
cles in Fig. 2(a) show FFMDs for $^{237}$U observed in our 
previous measurement of the $+2p3n$-transfer channel in 
the reaction $^{18}$O+$^{232}$Th [11] using the same experimental 
setup. A fairly good agreement, both for mass asymme-
try and for peak-to-valley (P/V) ratio, at all measured 
excitation energies may imply the independency of the 
FFMDs from the transfer channel. In Fig. 2(b), FFMDs 
for $^{239}$U are compared to those deduced in the neutron-
induced fission of $^{238}$U [12] (closed circles) at the simi-
lar excitation energy ranges of $E^*=$13.8–15.8 MeV, 18.8– 
23.8 MeV and 30.8–44.8 MeV. They have a mass resolu-
tion of 3.5 u in average, estimated from their spectra [14]. 
To allow for comparison with the present data (mass resolu-
tion $\sigma=6.5$ u), their FFMDs were broadened and the 
results are shown by the red curves in Fig. 2(b). For 
the two lowest energy regions, the P/V ratios obtained 
from their broadened FFMDs showed a good agreement 
with those from the present data within the statistical 
errors. Their P/V ratio at $E^*=20$–$30$ MeV was found 
to be about 20\% larger than our value of 2.3 $\pm$ 0.2, but 
still agrees within two sigma. As shown in Fig. 2, the 
MNT reaction provides the opportunity to study poorly-
understood spin dependence in fission. Although detailed 
studies are needed to find the conditions for which the 
present MNT approach could be used as a surrogate for 
neutron-induced fission, the reached agreements with the
other experimental methods indicate that a set of FFMDs shown later have enough quality to discuss the effect of MCF.

As a summary of all the experimental results, Fig. 3 shows the FFMDs for the twelve compound nuclides $^{237-240}U$, $^{239-242}Np$ and $^{241-244}Pu$ obtained by gating on the different ejectiles (i.e., different MNT channels) in Fig. 1. A 10-MeV interval of the excitation energy was chosen as a compromise between the available statistics and a reasonable increment of $E^*$. It should be noted that the FFMDs for $^{240}U$ and $^{241,242}Np$ were observed for the first time. It is evident that the FFMDs for all nuclides in Fig. 3 have predominately asymmetric shape at the lowest excitation energy. A growing contribution of symmetric fission can be observed with increasing excitation energy, however the double-humped shapes are still clearly preserved.

To understand these trends, the experimental FFMDs are compared with a calculation based on the fluctuation-dissipation fission model developed in [15, 16]. In this model, the evolution of a nuclear shape, defined by three parameters (charge-center distance, mass-asymmetry and fragment deformation), is traced from the compound state to the scission point by solving the Langevin equations, and FFMDs are calculated with the Monte Carlo method. The potential energy is defined as the sum of the liquid-drop part and excitation-energy ($E^*$)-dependent shell-correction energy given by: 

$$\delta W(0) \times \exp(-E^*/E_d),$$

where $\delta W(0)$ is the zero-excitation shell-correction energy.

The shell-damping energy was chosen to be $E_d=20\text{ MeV}$, as in [11, 16]. The calculation reproduces well the global shape the FFMDs, both for the peak-to-valley ratio of the double-humped shape and the position of the light and heavy-fragment peaks, for $n+^{233,235}U$, $^{239}Pu$ [16] as well as our recent MNT fission data for $^{231-237}Th$, $^{232-236}Pa$ and $^{234-238}U$ [11] within the limit of low excitation energies $E^* \leq 20\text{ MeV}$, for which the effects of MCF should be small.

As a first step in the present calculations, MCF was not taken into account, which means that calculated FFMDs are due only to fission of the initial compound nucleus at each specific excitation energy. The results are shown by the thin blue curves in Fig. 3. Under this assumption, the mass asymmetry, i.e., the peak positions of the double-humped FFMD, for all isotopes are reproduced below $E^* \approx 20\text{ MeV}$, with clear deviations seen for higher energies. At the highest energy, the calculation shows structure-less symmetric fission in contrast to the measurement. With regard to the $P/V$ ratios of the FFMDs at $E^*=10-20\text{ MeV}$, the calculation which reproduced those for $^{231-234}Th$, $^{232-236}Pa$ and $^{234-237}U$ from the MNT fission of $^{18}O+^{232}Th$ [11] agrees well also with the present data for the heavier uranium isotopes $^{238-240}U$. On the contrary, the calculation gives a smaller $P/V$ ratio for heavier neptunium ($^{241,242}Np$) and plutonium ($^{241-244}Pu$) isotopes. One of the possible reasons for this deviation could be in the treatment of the neck parameter $\varepsilon (0 < \varepsilon < 1)$ [17], which defines the depth of the potential at the neck of the dumbbell-shaped nucleus, used in our two-center shell calculation. In this work, we adopted $\varepsilon=0.35$ derived as an optimal value in [16] to explain the FFMDs of compound nuclei with mass of $234-240$. For heavier nuclei, this value could thus be slightly different. This deviation, however, does not influence our conclusion which was drawn from the discussion on the excitation-energy dependence of the FFMDs. The evolution of the $\varepsilon$ parameter in heavier nuclei will be the topic of a future investigation.

In the next step, MCF was introduced into the calculation. Figure 4(a) is a conceptual view of MCF for the case of $^{240}U$ as the initial compound nucleus. The highly excited $^{240}U$ can decay either via first-chance fission, or via single neutron emission, leading to the less excited $^{239}U$. The latter nucleus can decay again either by fission (thus, second-chance fission) or by neutron evaporation; the competition between fission and neutron emission continues until the excitation energy drops below the fission barrier of the corresponding daughter nucleus. The shape of the FFMD at each fission chance is also shown schematically in this panel, with predominantly symmetric fission for the initial highly excited compound nucleus $^{240}U$, and dominant asymmetric fission for subsequent fission chances of daughter nuclides, in particular, for $^{237}U$ (4th-chance fission). The application of this procedure to the calculated FFMDs for $^{240}U^*$ is demonstrated in Fig. 4(b). The calculated FFMDs for respective fission chances are shown by the dashed curves with different colors, where the fraction (probability) of each fission chance is determined using the GEF code [8]. The reduction of the excitation energy of the compound nucleus due to neutron emission was calculated from neutron binding energies [18] and a mean energy for the emitted neutron, ~1.9 MeV, obtained by the PACE2 code [19]. For each MCF step, the potential energy surface of the respective compound nucleus was also adopted. The sum of all the FFMDs obtained from each fission chance (up to 6th-chance fission) is shown by the thin black curve. It reproduces the observed peak positions of the experimental FFMD, but has narrower peaks than the measured ones. However, after introducing the experimental mass resolution, the calculation well reproduces also the $P/V$ ratio as well as the mass asymmetry as shown by the thick solid red curve. The key conclusion which can be drawn from Fig. 4(b) is that the apparent mass-asymmetric fission observed in the data even at high excitation energies originates from the lower-energy 4th-, 5th-, and 6th-chance fissions ($^{235,236,237}U$). On the contrary, the 1st and 2nd-chance fissions lead to predominantly symmetric mass splits, as they occur at high excitation energy.

The same calculation procedure was applied to all the cases displayed in Fig. 3, where the results are shown by thick red curves. In contrast to the results without MCF
The calculation with MCF well explains the excitation-energy dependence of the FFMDs characterized by mass-asymmetry and P/V ratio. With increasing excitation energy, FFMDs contain greater contributions from higher fission chances. Therefore, the agreement in Fig. 3 for all the excitation-energy ranges validates the calculation of the FFMDs. The decreasing P/V ratio of the measured FFMDs from uranium to plutonium (for example, $E^* = 30-40$ MeV) is also explained by introducing MCF, whereas the analysis without MCF predicts almost the same flat-top distributions through all the isotopes.

To conclude, even though MCF is a well-established concept in several fission observables (e.g., fission probability), so far its role for fission-fragment mass distributions has not been experimentally investigated. This is mainly due to the absence of systematic data on the FFMDs in a large span of excitation energies. We overcame this difficulty by exploiting the novel approach of multi-nucleon transfer reactions. Fission of a multitude of nuclides studied in a broad range of excitation energies has allowed us to show that the apparent asymmetric shape of FFMDs for a given initial excitation energy originates from fission of less excited lighter isotopes produced via a chain of MCF. In particular, this finding means that asymmetric shapes in the FFMDs measured at high excitation energies ($E^* > S_a$) should no longer be interpreted as signatures of survival of shell effects in the initial compound nucleus, which would incite one to reexamine existing experimental data measured at high excitation energies. Ignoring multi-chance fission, the asymmetric structure of FFMD observed at high excitation energy would introduce an unexpectedly higher shell-damping energy than the conventional $E_d = 20$ MeV which was also used in this work. The shell-correction energy at high excitation energy is also important for other fields, for example, heavy-ion fusion reaction for the synthesis of super-heavy elements (SHEs). This is because only the shell-correction energy forms the fission barrier of a compound nucleus the height of which significantly alters its survival probability in the competition between neutron evaporation and fission.

Our results also suggest that the consideration of MCF is essential to interpret and evaluate other fission observables. One of the examples is the neutron multiplicity as a function of fragment mass $A$, $v(A)$. An important, but not yet fully understood phenomenon, the increase of the initial excitation energy leads to enhancement of $v(A)$ only for heavy fragments [8, 20]. For a quantitative discussion, $v(A)$ should be also represented as a sum of contributions from each fission chance. As a further development of our MNT approach, we aim to undertake measurements of prompt neutrons correlated with fission fragments by installing a neutron-detector array around the present fission setup.

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