Collinear Cluster Tripartition Channel in the Reaction $^{235}\text{U}(n_{th}, f)$

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Abstract—Investigation of the $^{235}\text{U}(n_{th}, f)$ reaction using the miniFOBOS double-arm time-of-flight spectrometer of fission fragments confirmed manifestations of the earlier unknown many-body, at least ternary, decay involving almost collinear decay-product escape, which were first observed in the spontaneous fission of $^{252}\text{Cf}(sf)$. The use of variables sensitive to the nuclear charge of fission fragments allowed the reliability of identification of decay events to be increased and new decay modes to be revealed.

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1. INTRODUCTION

Some experiments that studied spontaneous $^{252}\text{Cf}$ fission revealed multiple manifestations of an earlier unknown many-body (at least ternary) decay, which we called collinear cluster tripartition (CCT) [1]. To obtain better insight into the physics of the effect, it was planned to investigate various fissile systems at various excitations up to the survival threshold for nuclear shells. The reactions chosen for this investigation included $^{238}\text{U} + ^4\text{He}$ (40 MeV). The respective experiment was performed at the Accelerator Laboratory of Jyväskylä University (Finland) [2, 3]. Here, we present the results of the next experiment, in which we studied the reaction $^{235}\text{U}(n_{th}, f)$ [4–5].

2. EXPERIMENTAL FACILITY

The experiment was carried out at the miniFOBOS spectrometer [6, 7] developed at the Flerov Laboratory of Nuclear Reactions at the Joint Institute for Nuclear Research (JINR, Dubna). The spectrometer was installed in the beam line of the IBR–2 pulsed neutron reactor at the Frank Laboratory of Neutron Physics at JINR. Neutron pulses of duration 320 µs followed one another at a frequency of 5 Hz. In order to reduce the fast-neutron and gamma-ray background in the experimental box, a bent reflector neutron guide 20 m long was used to extract thermal neutrons. To increase the neutron transportation efficiency, the neutron guide was filled with argon at a pressure slightly higher than atmospheric pressure, and an almost factor of two increase in the neutron flux at the target was attained owing to this. The neutron beam was formed using a cadmium collimator with a pass-through hole of area not larger than 1 cm$^2$. The thermal-neutron flux incident to the target was close to $10^6$ neutron cm$^{-2}$ s. Fission events were recorded within the first 20 ms of each pulse—that is, within the time interval of the arrival of neutrons at the target.

The layout of our experimental facility is shown in Fig. 1. The miniFOBOS is a double-arm time-of-flight spectrometer of fission fragments (FFs) based on the standard detector modules of the FOBOS $4\pi$ spectrometer [8].

Each detector module consists of a position-sensitive avalanche counter (PSAC) and an axial Bragg ionization chamber (BIC), which records the

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Fig. 1. Layout of the experimental facility: (1) start avalanche counter, (2) Bragg ionization chambers, (3) stop position-sensitive avalanche counters, and (4) neutron guide with a collimator at the exit.
Fig. 2. Relationship between the average mass $\langle M_{TE} \rangle$ (○) and the assumed $M_{TT}$ value, $v(M_{TT})$ (●) [16] [where $v(M_{TT})$ is the average number of neutrons emitted by a fragment of mass $M_{TT}$], for the reaction $^{235}\text{U}(n_{th},f)$.

total distribution of the energy loss (Bragg curve) suffered by fragments in the working gas of the chamber. The amount of the energy loss (residual energy) in the chamber is found by the method of the on-line digital processing of the signal [9]. The system for processing the BIC signal comprises a charge-sensitive preamplifier, a unit for Bragg curve digitization, and a digital processor. It is important that the constant “pedestal” before the signal is automatically subtracted by the processor in accordance with its operation algorithm. As a result, only one unknown parameter, the energy scale of the channel, appears in the channel—residual energy linear calibration dependence.

It is well known that the time over which the track produced by a fragment in the gas of the chamber drifts to the Frisch grid is related to the nuclear charge of the fragment [10]. The corresponding parameter was measured as the difference between the time signal from the PSAC and the signal of the time reference to the front of the pulse from the Frisch grid in the BIC.

The linear PSAC signal, which is proportional to the specific ionization loss $(dE)$ of FFs in the counter gas, was used as another charge-sensitive variable.

The specially designed start detector is a symmetric avalanche counter (SAC) with a target within its volume. Owing to this design, the start detector generates a correct start signal even for many-body events. Indeed, only the velocity of the fastest fragments from many-body decay is measured correctly if the source of fragments and the start detector are separated by a certain distance.

The target with an active layer 100 $\mu$g cm$^{-2}$ thick from the isotope $^{235}\text{U}$ was made by deposition onto an Al$_2$O$_3$ backing 50 $\mu$g cm$^{-2}$ thick.

The apparatus also comprised a remotely computer-controlled system for evacuation and supply of a gas (pentane) to the avalanche counters and a standard mixture of 90% Ar and 10% CH$_4$ to the ionization chambers. Note that, in the experimental hall, the pressure in the ionization chambers and the temperature should be constant in order to ensure the required stability of the above-mentioned drift time parameter. Conditioners were used to guarantee thermal stability in the experimental hall and in the box containing the electronics.

3. DATA PROCESSING

Thus, five parameters were detected in each arm of the spectrometer—namely, the time of flight ($T$) as the time difference between the SAC and PSAC signals, the energy ($E$) lost by FFs in the BIC, the drift time, and the coordinates of the point $(X,Y)$ where the fragment traversed the PSAC. The processing of the initial data for each FF yielded the primary mass (before neutron emission) if the 2V method was used.
and the postneutron mass if the V–E method was used (these two masses were denoted in what follows by $M_{TT}$ and $M_{TE}$, respectively), the velocity vector, and the estimate of the nuclear charge.

In all, about $9 \times 10^6$ fission events were detected in the experiment. Data stability was checked by mean values and variances calculated for the main parameters, such as the time of flight, the BIC signal amplitude, and the drift time, over each 1000 events. It turned out that, for all parameters, the instability was on the same order of magnitude as the expected experimental resolution; therefore, no corrections were introduced in the data.

The next stage of the data processing consisted in the coordinate calibration of the PSACs by comparing the shadow image arising in them from the grid supporting the foil of the BIC entrance window with the known geometric parameters of the actual grid. The PSAC spatial resolution did not exceed 1.5 mm.

After that, the time of flight $T$ was calibrated. This procedure consists in determining two calibration coefficients in the linear conversion of the channel time code into nanoseconds,

$$T_{\text{expt}} = T_{\text{chan}} \frac{dT}{dk} + T_0,$$

where $T_{\text{chan}}$ are the experimental times of flight (TOFs) in the channels, $T_{\text{expt}}$ are the corresponding TOFs in nanoseconds, and $dT/dk$ and $T_0$ are the calibration parameters.
The slope of the calibration straight line, \(dT/dk\), was determined using the time calibrator at each arm of the spectrometer. As to the coefficients \(T_0\), they were adjusted in such a way that both the experimental mass spectrum \(Y(M_{TT})\) and the positions of the peaks in the velocity spectrum agreed with the values known from the literature [11–13]. The thicknesses of the SAC films were parameters to be refined within the same procedure. The developed computer code is described in [14].

The next stage of the data processing is calibration of the circuit for energy (\(E\)) measurements. It is a nontrivial procedure because only less than half of the initial fragment energy is released in the BIC. The large energy loss is mainly due to a large thickness of the BIC entrance window. It is an obvious price to be paid for the high aperture of the spectrometer. The idea of \(E\) calibration and mass reconstruction consists in fitting the current spectrum of quasimasses \(M_{TE}\) obtained by the \(V-E\) method to the known spectrum. The algorithm is described in detail in [15].

Previous experiments showed that CCT was mainly affected by shell effects and that at least one of the decay products was a magic nucleus. To identify a fragment as magic (in mass number), the mass calibration should be kept unbiased. This requirement seems more important than the requirement of having a high mass resolution.

A good test for both the time and energy calibration to be unbiased is a comparison of the masses \(M_{TT}\) and \(M_{TE}\). The results of this comparison are shown in Fig. 2. The closed circles are the results obtained by subtracting, from each mass number, the average number of emitted neutrons, which is known from the literature [16]. By definition, the ordinate of each point is therefore the average mass \(\langle M_{TE}\rangle\) for the value \(M_{TT}\), which is the abscissa of the point. To compare this assumed dependence with the experimental one, we accumulated the \(M_{TE}\) mass spectrum for each mass \(M_{TT}\) and calculated the average over the spectrum. Only events with approximately equal momenta were selected. This guarantees rejection of scattered fragments and improves the mass resolution for selected events. The corresponding experimental values are shown in Fig. 2 by open circles. As is evident from the figure, the expected linear dependence is well reproduced.

To evaluate the nuclear charge of an FF from the measured drift times of the track in the BIC, \(D_{\text{expt}}\), and the FF mass and energy, a special procedure was developed [17, 18]. The charge resolution for fragments of the light peak in the mass distribution was (root-mean-square deviation) \(\sigma = \pm 1.6\) ch.un.

As was already indicated, the major background for the effect under investigation comes from fragments scattered by the BIC grid. They are characterized by a smaller mass \(M_{TE}\) yet by a correctly measured velocity and, accordingly, mass \(M_{TT}\). The drift time of the track in the BIC, \(D_{\text{expt}}\), will also be correctly measured. In view of the aforesaid, it is generally possible to separate the effect of interest from the scattered events of binary fission. On the basis of the formula used in [17, 18] to relate the drift time, mass, and charge of an ion, we introduce, for this purpose, a new variable, the estimated drift time,

\[
D_{\text{est}} = \alpha - \beta \sqrt{E_{\text{BIC}} M_{TT}^3} Z^{-2/3} + \gamma M_{TT},
\]

where \(M_{TT}\) is the mass corrected for the number of emitted neutrons (that is, the “postneutron” mass); \(Z\) is the fragment charge number consistent with the hypothesis of a constant charge density \(Z_{\text{UCD}}\); \(E_{\text{BIC}}\) is the fragment energy in the BIC; and \(\alpha, \beta,\) and \(\gamma\) are the parametrization coefficients found in advance during the calibration.

From the definition of \(D_{\text{est}}\) in (1), it follows that, for scattered events, we have \(D_{\text{est}} = D_{\text{expt}}\) within the resolution and that, for CCT events, we have \(D_{\text{est}} > D_{\text{expt}}\). One can readily verify this by comparing \(D_{\text{est}}\) for a typical light fragment detected in a correct way and after scattering. The result of employing the variable introduced in this way will be discussed below.

4. RESULTS

In our previous experiments devoted to studying the spontaneous fission of \(^{252}\text{Cf}(\text{sf})\), a distinct feature was observed in the mass–mass distribution of FFs without any selection of detected events. This is a
specific two-dimensional bump below the locus of ordinary binary fission events [19, 20] (Fig. 3a).

The inner structure of the bump, which included a ridge \( M_1 = \text{const} \sim 70 \text{ amu} \) and a system of slantwise ridges \( M_s = M_1 + M_2 = \text{const} \), where each constant presumably corresponds to the mass of a pair of magic clusters (light/heavy) Ni, Ge, Sn, and Te was discussed in those studies, and the reasons why the bump was seen in only one arm of the spectrometer were analyzed there.

A similar bump is also well seen here in the mass distribution of FFs from the reaction \( ^{235}\text{U}(n_{th}, f) \) (Fig. 3b). As in the previous case, the bump is observed in only one spectrometer arm (with index 1) facing the target backing. In Fig. 3c, the projections of the bump onto the \( M_1 \) axis for the aforementioned reactions are compared. The peaks are centered in the mass range 68–70 amu associated with the magic Ni isotopes. The projections onto the \( M_s = \text{const} \) direction are presented in Fig. 3d. Although the masses of the corresponding fissioning systems differ by 16 amu, the projections of the bump onto this direction are shifted by not more than 6 amu.

The position of the peak for Cf corresponds to the sum of the masses of the magic Sn and Ge isotopes; for \( ^{236}\text{U} \), it corresponds to the sum of the masses of Sn and Ni.

A comparison of the nuclear-charge spectra of FFs detected in the corresponding arm of the spectrometer confirms the hypothesis that the bump is based on the elements in the range from Ni to approximately Ca (Fig. 4).

The presence of the slantwise ridges \( M_s = \text{const} \) in the bump was revealed for \( ^{252}\text{Cf} \) FFs by a special
mathematical treatment of the mass–mass distribution [19]. The \( dE \) coordinate, which is sensitive to the nuclear charge, allowed this to be directly verified for \(^{230}\text{U}^*\) FFs. Figure 5a shows the distributions of FFs that fell within the window for increased fragment velocities \( V \) and the specific energy loss \( dE \) in the stop PSAC-1. This kind of selection discriminates events where a vee formed by two tripartition fragments goes to the first arm of the spectrometer (where the bump is observed). Their overall energy loss in the PSAC turns out to be higher than \( dE \) of a “normal” fragment from the light peak of binary fission. It is worth mentioning that, in this case, the experimental variables \( V \) and \( dE \) used to select CCT events are not distorted by scattering of fragments by the BIC entrance grid (Fig. 1), this being the main source of background events.

The bump region in Fig. 5a shows a sort of sloping dip between the ridges \( M_s = \text{const} \), which is indicated by a slantwise arrow. The projection onto this direction is given in Fig. 5b. The observed peaks correspond to the sum of the masses of the magic isotopes \(^{128,132}\text{Sn}\) and \(^{68,70,72}\text{Ni}\) (left bracket above the peaks) and \(^{128,132}\text{Sn}\) and \(^{80,82}\text{Ge}\) (right bracket). The peak centered at 68 amu is well distinct in the projection onto the \( M_1 \) axis (see Fig. 5c). To the left of it, the peak is also bounded by the magic \(^{62}\text{Cr}\) isotope [20]. It can be inferred from Fig. 5a that heavy fragments forming the bump are within the mass–number range 128–144, which is associated with the magic nuclei \(^{128}\text{Sn}\) and \(^{144}\text{Ba}\). This inference is confirmed by the projection of the bump onto the corresponding axis (Fig. 5d).

When events characterized by approximately equal momenta and velocities are selected, the mass–mass distribution of FFs reveals a specific structure in the form of a right angle whose vertex lies on the diagonal of the plot in the vicinity of the points (68, 68) amu (Fig. 6a). We observed earlier a similar structure (rectangle) in the mass–mass distribution of fragments from the spontaneous fission of the \(^{252}\text{Cf}\) nucleus [21].

The assumption that the sides of the right angle in Fig. 6a are connected with magic Ni isotopes is confirmed by Fig. 6b. It depicts the distribution of FFs with approximately equal momenta, velocities, and nuclear charges. With allowance for a small shift toward larger values in the charge calibration in arm 1 [17], a charge of about 28 does indeed correspond to the points at the vertex of the angle. Of interest in this figure are also points that lie on the slantwise straight line \( M_s = \text{const} \) and the “lost” mass corresponds here to the deformed magic nucleus \(^{118}\text{Pd}\). The middle of the points corresponds in mass and charge to a symmetric precession configuration of three magic clusters: \(^{59}\text{V}–^{118}\text{Pd}–^{59}\text{V}\).

Note that, although the distribution in Fig. 6b includes just a few events, the reliability of their analysis is quite high. Indeed, there are no other points in the vicinity of the concentration of points under consideration. Each point results from the measurement of six independent experimental parameters (time of flight, energy, and drift time at each arm). Finally, the points are grouped along three specific directions: \( M_1, M_2 = \text{const} \), and \( M_s = \text{const} \).

A less stringent selection permitted revealing a whole family of events similar to the three-cluster configuration discussed above. Figure 7a shows the distribution of FFs upon momentum and drift–time selections. The momentum selection was applied to
Fig. 7. (a) Mass–mass distribution of FFs selected by the momentum and the parameter $D_{\text{est}}$. The arrow points to the group of events with approximately equal masses. (b) Window (rectangle) for selection in the momentum–momentum distribution of FFs. (c) Events of collinear cluster tripartition with equal masses of detected decay products. The numbers label the following cluster configurations: (1) $^{78}\text{Zn}_{30}$–$^{80}\text{Ge}_{50}$–$^{77}\text{Zn}$, (2) $^{82}\text{Ge}_{50}$–$^{72}\text{Ni}_{28}$–$^{82}\text{Ge}$, (3) $^{80}\text{Se}_{54}$–$^{60}\text{Fe}_{24}$–$^{80}\text{Se}$, (4) $^{95}\text{Rb}_{58}$–$^{95}\text{Rb}$, (5) $^{103}\text{Zr}_{40}$–$^{30}\text{Zr}$, and (6) $^{108}\text{Mo}_{66}$–$^{20}\text{O}$–$^{108}\text{Mo}$. (d) As in Fig. 7c, but for total statistics.

5. CONCLUSIONS

Here, we list the basic results of the present study.

(1) We have proven that there exists a two-dimensional region of products resulting from CCT of heavy nuclei with an extremely high (in relation to the known case of ternary fission) yield of about $4 \times 10^{-3}$ (binary fission)$^{-1}$. This conclusion was drawn not only from the observation of CCT events in the FF mass–mass distribution, as had been done in previous experiments, but also on the basis of the selection in the nuclear charge, specific ionization
loss, and drift time of the fragment track in the ionization chamber.

(2) The hypothesis proposed earlier that concerns the inner structure of the region of high CCT product yields (two-dimensional bump) has been directly confirmed by the following:

(i) In the mass–mass distribution, this region is bounded at least on three sides by the mass numbers corresponding to magic nuclei.

(ii) It includes the slantwise ridges \( M_2 = M_1 + 2M_2 = \text{const} \), where \( M_1 \) and \( M_2 \) are the masses of detected fragments and the constant takes several values corresponding to the sum of mass of magic nuclei, \( 128, 132 \text{Sn} + 86, 82 \text{Ge} \) and \( 128, 132 \text{Sn} + 68, 70, 72 \text{Ni} \).

This conclusion was drawn from an analysis of the region in question, which was separated from background events by using selections in the nuclear charge, specific ionization loss, and drift time of the fragment track in the ionization chamber.

The fact that the position of the ridges \( M_2 = \text{const} \) does not change as one goes over from \( ^{252}\text{Cf} \) to \( ^{236}\text{U}^* \), which differ in mass by 16 amu, confirms the hypothesis that they are due to the aforementioned magic-cluster pairs.

(3) For the system \( ^{236}\text{U}^* \) not studied previously, the rectangular structure associated with the magic nuclei \( 68, 70 \text{Ni} \) has been proven to exist in the region of equal masses of detected fragments.

(4) The \( ^{236}\text{U}^* \) CCT mode, which has a yield of about \( 8 \times 10^{-6} \) (binary fission)\(^{-1} \) and within which the system before scission is a chain that is formed by three nuclei with commensurate masses and where the end clusters are identical magic nuclei, has been observed for the first time.

Possible physical scenarios behind the observed manifestations of the effect have not been considered in this article for reasons of space.

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