RARE FISSION MODES: STUDY OF MULTI-CLUSTER DECAYS OF ACTINIDE NUCLEI.

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We present a brief review of the results obtained by our collaboration in the frame of the program aimed at searching for new type of multibody decay of actinides, which was arbitrarily called as “collinear cluster tripartition” (CCT). First indications of new decay mode obtained for $^{248}$Cm (sf) and $^{252}$Cf (sf) let one to suppose that at least ternary almost collinear decay of the initial nucleus into the fragments of comparable masses appear to occur with the probability of about $10^{-5}$ per binary fission. The process is strongly influenced by shell effects in the decay partners. The results under discussion were obtained by the “missing mass” method i.e. only two of the decay products were detected in coincidence while the conservation laws indicate a presence of at least third partner.

1. Experiment at the modified FOBOS setup

First indications onto unusual multibody decays of $^{248}$Cm (sf) and $^{252}$Cf (sf) we have obtained in the experiments performed at the $4\pi$-spectrometer FOBOS [1-3]. In order to improve reliability of identification of the CCT events the ordinary FOBOS setup has been modified and covered by the belt of neutron detectors. The experimental layout of the modified FOBOS spectrometer is shown in Figure 1.

Due to the low cross-section of the process and some additional requirements addressed to the spatial arrangement of the detectors involved the two-arm configuration containing five big and one small standard FOBOS modules in each arm was used. Every module consists of position-sensitive avalanche counter (PSAC) and Bragg ionization chamber (BIC). Such scheme of the double-armed TOF-E (time-of-flight vs. energy) spectrometer covers ~29% of the hemisphere in each arm and thus the energies and the velocity vectors of the coincident fragments could be detected. In order to provide “start” signal for all the modules only wide-aperture start-detector capable to span a cone of ~100° at the vertex could be used. Even more essential requirement for the proper detection of the multibody events consists in combination of “start” detector with the radioactive source. Such three-
electron wide-aperture avalanche counter was especially designed for providing a “start” signal.

According to the model of the CCT process, which could be referred from the initial experiments, the middle fragment of the three-body pre-scission
chain borrows almost the whole deformation energy of the system. Being presumably in rest it would be an isotropic source of post-scission neutrons of a high multiplicity (~10) in the lab system. On the contrary, the neutrons emitted from the moving fission fragments are focused along the fission axis. In order to exploit this phenomenon for revealing the CCT events the “neutron belt” was assembled in a plane being perpendicular to the symmetry axis of the spectrometer, which serves as the mean fission axis at the same time. The centre of this belt coincides with the location of the FF source. The neutron detector consists of 140 separate hexagonal modules [4] comprising a $^3$He-filled proportional counters which cover altogether ~35% of the complete solid angle of $4\pi$. The number of tripped $^3$He neutron counters was added to the data stream as an additional parameter for each registered fission event.

According the mathematical model of the neutron registration channel worked out [5, 6] the registration efficiency for those neutrons emitted from an isotropic source was found to be very closed to its geometrical limit, while the registration efficiency for neutrons emitted from the fission fragments registered by the FOBOS modules amounted to ~4% because they are focused along the fission axis which is perpendicular to the plane of the neutron counter belt. The registration probability for more than one neutron from ordinary spontaneous fission in this geometry amounts to 1%, however, the same probability for the CCT events runs up to 85%. The registration probabilities for more than two neutrons are 0.3% and 62%, respectively. Thus the neutron belt proves to be an effective instrument for revealing fission events accompanied by the isotropically emitted neutrons.

The mass-mass plot of the coincident fragments with the high multiplicity of neutrons (at least 3 of them should be detected) is shown in Figure 2a. It is easy to recognize the rectangular-shaped structure below the locus of conventional binary fission. This structure becomes more conspicuous (Figure 2b) if the velocity cut shown in Figure 3a is applied to the distribution.

The rectangle in Figure 2b, which is bounded by the clusters from at least three sides. Corresponding magic numbers are marked in this figure at the bottom of the element symbols. More complicated structures (marked by the arrows a, b, c in Figure 2c are observed in the mass-mass plot if the events with two fired neutron counters are also taken into play. Omitting for a moment physical treating of the structures observed, we attract ones attention to the specific peculiarity of some lines constituted the structures “b” and “c”. The sum of the masses along them remains constant; see the dashed line in the lower left corner of Figure 2c for comparison.
Figure 2. a) The mass-mass plot of the complementary fragments with at least 3 neutrons detected; b) The same plot after filtering the fragment velocities in the rectangular box shown in Figure 3; c) The same as (a), but the lowest number of tripped neutron counters let down to 2.

Figure 3. Velocity matrix of the complementary fragments (a) and momentum-momentum plot (b). The events falling into the rectangular boxes in Figures 3a, b were used to compose the final mass-mass plot in Figure 4.
Figure 4a represents a similar structure to that shown in Figures 2a, b except that it is not gated by neutrons and both the velocity and the momentum windows are used here to reveal the mass-symmetric partitions. The corresponding momentum distribution of the fragments and the selection applied are shown in Figure 3b. The plot in Figure 4b obtained on conditions of the momentum selection solely is not so clear. However like in the previous case the rectangular structure observed is bounded by the magic fragments, namely $^{68}$Ni (the spherical proton shell $Z=28$ and the neutron sub shell $N=40$) and, probably, $^{84}$Se (the spherical neutron shell $N=50$). Each structure revealed maps an evolution of the decaying system onto the mass space.

![Figure 4. Mass-mass matrix of the complementary fragments selected by requirement of their approximately equal velocities and momenta (a), the same matrix if only momentum selection is assumed (b).](image)

It should be stressed that the observed neutron multiplicity (the number of tripped neutron counters) for the events from the rectangle in Figure 4 is low. This fact contradicts the expectations put forward earlier that a middle fragment in the chain should be the source of neutrons of high multiplicity. The discrepancy reported may be an indication of more complicated decay scenario to be restored.

The following conclusions can be drawn from the results presented above:
- the multi-fragment (at least ternary) fission is experimentally confirmed;
- clustering of the decaying system, i.e. pre-formation of the magic constituents inside its body is decisive for the effect observed;
- collinear pre-scission configuration predicted by theory is proved to be a preferable one for true tripartition.
2. Comparative study of the effect at different spectrometers

The next series of results to be reported were obtained in three different experiments [7] devoted to searching for collinear tripartition of the $^{252}\text{Cf}$ nucleus.

In the first experiment (Ex1, Figure 5 a), performed at the FOBOS spectrometer installed in Flerov Laboratory (JINR, Dubna), about $1.3 \times 10^6$ coincident binary fission events were recorded. The TOF of the fragments was measured over a flight path of 50 cm between the “start” detector (3) based on the micro-channel plates (MCP) placed next to the $^{252}\text{Cf}$-source (1) and the “stop” position-sensitive avalanche counters (PSAC, 4). The energies of the coincident fragments, which passed through the PSACs were measured in the Bragg ionization chambers (BIC, 5) with entrance windows supported by a grid (6) with 70% transparency. The geometrical structure of the grid is hexagonal, the side view is shown in the insert “a” of Figure 5.

![Figure 5. The scheme of coincident measurements of two fragments of the collinear tri-partition partners for the three experiments. First experiment Ex1 (a) was performed at the FOBOS setup. Here 1 – Cf source, 2- source backing, 3- microchannel plate (MCP) based timing “start” detector, 4- position sensitive avalanche counter as “stop” detector, 5- ionization chamber with the supporting grid 6 on the entrance window. The side view of the grid is shown in the insert “a”. Second and third experiments Ex2, Ex3 (b and c) were performed at the spectrometers based on MCP detectors 2, 10 and PIN diodes 8 bounded by frame 9. Insert a) the scheme of detecting of the tripartition partners is shown in the insert “b”. After passage of the dispersion foil two light fragments (L1 and L2) obtain a small angle divergence due to multiple scattering. One of the fragments (L1) can be lost hitting the separating block, while the fragment L2 reaches the energy detector.](image)

This mechanical structure of the detectors is essential for the registration of the effect described below. In Figure 5 (insert b) the primary heavy fragment (H) is emitted to the left from the free side of the $^{252}\text{Cf}$-source; the two light fragments (L1 and L2) are emitted into the same direction. As explained below scattering processes will separate the two light fragments in a small angular
separation, and only one of them is likely to be registered. If both fragments enter only the total energy is measured.

A similar source of $^{252}$Cf was used in further experiments performed in the Accelerator laboratory of the University of Jyväskylä, Finland (JYFL). In the second experiment (Ex2, Figure 5 b) we used a different TOF-E-spectrometer based on one MCP “start” detector and two PIN diodes (8), the latter provided both time and energy signals. An active area of each PIN diode was bounded by the frames (9). The flight-paths here were 10 cm for each detector arm. An Al-foil (7), 5 μm thick has been placed just near active $^{252}$Cf layer. In this experiment $2\times10^5$ binary events were registered.

In the third experiment (Ex3, Figure 5c) two pairs of the MCP-based timing detectors (10) provided signals for measuring TOF’s with flight paths of 8 cm each. The fragment energy was measured by PIN diodes. The total transparency of each arm amounted to 70% due to the grids of the electrostatic mirrors (four per detector, instead of two as in the Ex1 and Ex2) of the timing detectors. In this third experiment $2\times10^6$ of binary events were collected.

In Figure 6a we show in a logarithmic scale the two-dimensional (2D) distribution of the two registered masses of the coincident fragments in the experiment (Ex1) at the FOBOS set up. The “tails” in the mass distributions marked (3)-(6) in Figure 6a, extending from the loci (1) and (2) used to mark the conventional binary fission, are mainly due to the scattering of the fragments on both the foils and on the grid-edges of the “stop” avalanche counters and the ionization chambers. The only small, but important, asymmetry between the two arms to be emphasized consists in a very thin source backing for the “rear side” and the start detector foil located in the arm “b” only (Figure 6a). An astonishing difference in the shapes of the “tails” (3) and (4) attracts attention. There is a distinct bump, marked (7), on the latter “tail” (4), oriented approximately parallel to the line defining a constant sum of masses, $M_a+M_b=const$, i.e. tilted by $45^\circ$ with respect to the abscissa axis. The explanation of this bump is the essence of our analysis. The bump is located in a region corresponding to a large “missing” mass. The statistical significance of the events in the structure (7) can be deduced from Figure 6b. There the spectra of total masses, $M_{total}=M_a+M_b$, for the “tails” (4) and (3), spectrum “a” and spectrum “b”, respectively, are compared. The difference spectrum of “b” and the tail (3) is marked “c”, the integral of these events is $4.7\times10^{-3}$ relative to the conventional fission events contained in the locus (2), shown in Figure 6a.
In order to explain the differences in the “tails” (4) and (3) mentioned above following scenario is proposed, the geometry is shown in Figure 5 (insert b). In ternary fission the three fragments are emitted collinearly and two of the fragments are emitted in one direction but become separated with an angle less than 10° after passing a dispersing media, due to scattering. These materials are the backing of the source (located only on the side of tail (4) or the Al foil placed deliberately in the path). If both fragments pass on and enter into the (BIC), we register a signal corresponding to the sum of the energies of the two fragments. The event is registered as binary fission with almost usual parameters. In the other scenario only a proper energy (mass) of one of the light fragments is measured, because the second one is stopped (lost) in the supporting grid of the ionization chamber, or for the other cases in the frame of the PIN diode playing the role of the separating element.

Figure 6. Experimental evidence of the collinear tripartition of 252Cf obtained at the FOBOS setup. (a) Contour map (in logarithmic scale) of the mass-mass distribution of the collinear fragments detected in coincidence in the opposite arms (marked by letters “a” and “b”) of the spectrometer. The loci of conventional binary fission events 1, 2 are prolonged by the “tails” marked as 3-6 due to the scattered fragments. Bump 7 located below the line of the sum Ma+Mb=225 amu is analyzed in Figure 6b. There the spectra of total masses for the “tails” (4) and (3), spectrum “a” and spectrum “b”, respectively, are compared. The difference spectrum is marked “c”. Is a polynomial fit Curve “d” is a polynomial fit using the points outside of the gross peak on spectrum “a”.

The scenario proposed above is confirmed by the results obtained in the experiments Ex2 and Ex3 (Figures 5b, c). Figure 7 depicts the spectra obtained in the same manner as it was done for the Ex1. As can be referred from the figure the bumps similar to this labeled by number 7 in Figure 6 demonstrate a multipeak structure which manifests itself due to better mass resolution in Ex2.
and Ex3. The origin of these peaks can be understood from the fine structure of the bump 7 in two-dimensional presentation (Figure 8).

Figure 7. Spectrum of sum of masses (Mb+Ma) from experiment Ex2 (a), for two registered fragments for the gate similar to the "tail"4 from Figure 5a, spectrum "b" corresponds to the "clean" opposite arm free from dispersion foil, "c" is the difference spectrum. Spectrum of sum of masses of two detected fragments obtained in our third experiment Ex3 (b): (a) from the arm facing the source backing, (b) the same spectrum smoothed by means of averaging of counts in three adjacent channels (shifted up by 25 counts). (c) The sum of spectrum “a” and a complementary spectrum obtained in the second arm of the spectrometer (shifted up by 35 counts). The sums marked in the panels correspond to different pairs of magic nuclei (see text). The peak in spectrum "b" marked by arrow is due to the doublet of “missing” 70, 68Ni fragments.

Figure 8. a) The figure depicts as a 2D-contour map (Mb versus Ma) the difference between the “tails” 4 and 3, of the events measured with the FOBOS-detector system shown in Figure 5a; note the expanded scale for the lighter mass fragments. Dashed lines tilted by 45° with respect to the Ma axis correspond to the fixed total mass of detected fragments (see the text for more details). Part b) the same as in a, however, passed through a filter which emphasizes the two dimensional structures.

3. Discussion

From the observed mass spectra we will have to consider a ternary fission process with one heavier and two lighter fragments. The missing masses in the
sum spectra of the experiment (Ex1) suggest subsystems with particular masses. The same mass values are observed as distinct peaks in Figure 7; these are also seen as ridges in Figure 8b. We note that from these data the shell closures in proton and neutron number are decisive for the formation of the emitted subsystems. As can be deduced from Figure 8a the ridges (marked by the dashed lines) go through crossing points corresponding to different combinations of two fragments with “magic” nucleon numbers (marked by the dash-and-dot arrows). These marked points could be related to mass values with magic subsystems well known from binary fission [8] as follows: 204→70Ni+134Te or 72Ni+132Sn (“missing” 48Ca), 208→80Ge+128Sn (“missing” 44S28) and for Mtotal=212→80Ge+132Sn or 78Ni+134Te or 68Ni+144Ba.

It should be noted that the central peak in the Figure 7a (marked as Σ = 204-208) is likely a triplet which includes the peak centered at 206 a.m.u. It could be related to a magic subsystem 206→72Ni+134Te (“missing” 46Ar28). Thus, three subsystems from these proposed above consist of three magic clusters each. The ridges discussed are crossed as well by the horizontal ridge (seen via bunching of contour lines in Figure 8a), this effect can be linked with the isotopes of 68,70Ni which are also magic [9]. This observation would imply that the detected light fragment from the two L1, L2 fragments (see Figure 5b) is always a Ni-isotope. Due to the symmetry of the detector setup, namely that the two L1, L2 fragments are always detected in coincidence with the same heavy fragments, one must also observe events with a “missing” Ni-fragment. This is indeed observed, the peak corresponding to “missing” masses of 70 and 68 a.m.u. is well seen in Figure 7 (curve b). Thus the different peaks in the “missing”- mass spectrum consistently correspond all to the ternary decay scenario proposed.

References