Clusterization in heavy cold nuclei

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Abstract. A cluster is broadly understood to be an object which keeps its identity in a larger scale system. Manifestations of clustering can be revealed in very different size systems from exotic nuclei to galaxies. In our work we search for cluster effects in low energy fission of heavy nuclei. In the series of experiments carried out with the spectrometers based both on the gas filled (FOBOS spectrometer and its modifications) detectors and mosaics of solid-state detectors (setups at the beams of alpha-particles and deuterons, COMETA spectrometer) using the “missing mass” method and with the direct detection of three decay partners, we have discovered a new type of the ternary decay of heavy nuclei dubbed by us “collinear cluster tri-partition (CCT)”. The most interesting aspects of both the original methodic used and the physics of the effect observed are discussed in the presented report.

1. Introduction

Our research of the rare multibody decays has a long history. The first experiments more than fifteen years ago were motivated by two significant discoveries in nuclear physics. In 1980 A Sandulesku, D N Poenaru, and W Greiner described the calculations indicating the possibility of a new type of decay of heavy nuclei intermediate between alpha decay and spontaneous fission [1]. It was called heavy ion radioactivity or cluster decay. In 1984 H J Rose and G A Jones observed the decay: $^{223}$Ra → $^{209}$Pb + $^{14}$C. It was first experimental confirmation of the heavy ion radioactivity [2]. From the very beginning it was clear that the double magic Pb core plays an essential role in the process.

Another important example came from fission. Figure 1(a) depicts the fission fragments (FFs) mass spectra from spontaneous and neutron induced fission of Fm isotopes [3]. Only two extra-neutrons or minimal excitation of the fissioning system change the FFs mass-distribution radically. Due to the two components seen in the mass and total kinetic energy (TKE) spectra, the fission process of such kind was called “bimodal fission”. By its nucleon composition the isotopes of Fm are very close to pair of the magic Sn nuclei. Thus a preformation of magic nucleus (nuclei) gives rise to the cluster decay and bimodal fission. According to the calculations of V Pashkevich [4], both decay modes manifest themselves even in $^{253}$Cf nucleus (figure 1(b)).

Keeping in mind these two examples we asked ourselves a question whether the decay of such kind ($^{246}$Cm → 3 $^{82}$Ge) is possible. If yes, it would be an unknown processes of true ternary fission or ternary cluster decay. We have tried to answer the question during all these years.
Since the discovery of the cluster radioactivity, the following definition of the cluster has been adopted: “Any light partner of the cluster decay heavier than α-particle is called “cluster”. On the other hand: “Historically, the observation of clustering started with the α-particle, which presents a large binding energy and therefore tends to keep its own identity in light nuclei” [5]. Using this broader definition one can find clusters in the systems of different scales from nucleons up to galaxies. The latter definition seems to be more physical. We follow it below.

**Figure 1.** Comparison of the FFs mass spectra for spontaneous and neutron induced fission of Fm isotopes [3] – (a). Potential energy of a fissioning $^{252}$Cf nucleus, corresponding to the bottoms of the potential valleys, as a function of $Q$, proportional to its quadrupole moment. The valleys of the Lead radioactivity (1) and partially overlapping two Sn clusters (2) in $^{252}$Cf nucleus [4] – (b).

Short review of the theoretical background of the problem under discussion should start from ref. [6]. The author of this work W J ’Swiatecki gives a quote from a notebook of Niels Bohr with the following reasoning concerning ternary fission: “…what if the strong electric repulsion would stretch out the post-saddle shape into a sufficiently long cylinder that would actually prefer to divide into three rather than two pieces? This would not be unexpected, because for Uranium the energy released in a division into three equal fragments is actually greater than into two.” The original Bohr’ notes are presented in figure 2.

**Figure 2.** Niels Bohr. The notes made by him 7th October 1950, his 65th birthday.

Later theoretical indications of the true ternary and even quaternary fission were presented in [7, 8]. Two-neck and three-neck shapes marked by the arrows (figure 3(a)) were predicted in [7]. The points
are shown as most favorable for the formation of a “quasi-molecule”. The low probability of such configurations was underlined. The calculations made using the Strutinsky approach demonstrate two-neck shape of the $^{252}\text{Cf}$ nucleus at the descent from the fission barrier [8] (figure 3(b)). The chain-like shape consists of 3 magic clusters.

**Figure 3.** Symmetrical shapes of equilibrium for a liquid drop model [7]. Here $X \sim z^2/A$ is the conventional liquid drop model parameter, $W(x)$ is the difference of the potential energy of the deformed nucleus and that of the initial sphere – (a). The shape of the $^{252}\text{Cf}$ nucleus at the bottom of the “symmetric” valley of the potential energy surface [8] – (b). See text for details.

In our key publications [9–11], we have summarized the experimental evidences of the existence of a new at least ternary decay channel of low excited heavy nuclei dubbed by us collinear cluster tri-partition (CCT). Most of the results were obtained by the “missing mass” method. It means that two decay products (fragments) were detected in coincidence using a double armed time-of-flight spectrometer, while the significant difference between their total mass $M_s = M_1 + M_2$ and the mass of a mother system served as a sign of at least ternary decay. A fragment mass was calculated by the energy $E$ and the velocity $V$. Mainly a scattering of fragments at the entrance of an $E$-detector gives the background events simulating ternary decay. Selection of the “true” events was provided by applying the gates on the fragments momenta, velocities, experimental neutron multiplicity and the parameters sensitive to the fragment’s nuclear charge. Statistical reliability of the typical structures against a random background was estimated to exceed 98% [10]. The structures were reproduced using both spectrometers based on the gas filled detectors (modules of the FOBOS setup [4]) and the solid-state detectors, namely timing detectors on the microchannel plates and the mosaics of PIN diodes (COMETA setup [10] and the similar ones [11]). Even though mass reconstruction procedures for these two types of spectrometers differ significantly, the obtained results are in good agreement. All the structures revealed are somehow related to the magic fragments, such as $^{128}\text{Sn}$, $^{134}\text{Te}$, $^{72,68}\text{Ni}$ and the others. Thus, now we have a whole collection of different CCT manifestations observed through the linear structures in the mass correlation distributions of the decay products [11].

2. Recent experiments and results

The experiment Ex1 was carried out using the COMETA-F spectrometer at the FLNR (JINR, Dubna, Russia). The layout of the spectrometer is shown in figure 4.

It is double-armed time-of-flight FFs spectrometer consisted of four mosaics of PIN diodes and two timing detectors. The signals of all the detectors are digitized in 0.2 ns increments by the fast flash-
ADC (Amplitude to Digital Convertor) CAEN DT5742. In case the PIN diode is used for the measurement of both FF energy and velocity, for the proper reconstruction of the FF mass, one should take into account the so called “pulse height defect” and “plasma delay” effects [12, 13]. This is exactly what was done in the data processing approach proposed by us in Refs. [14, 15]. In the latter publication a new “parabola time-pickoff” method used in Ex1 is described.

Figure 4. Layout of the COMETA-F setup. A spectrometric source of $^{252}$Cf (sf) is located between two microchannel plates based timing detectors St1 and St2 giving “start” signals. Four mosaics (D1–D4) of eight PIN diodes each provide the signals for measurement of both the FF time-of-flight and energy. The FF mean flight pass does not exceed fifteen centimetres.

FFs mass correlation distributions in the region of the “Ni-bump” obtained in Ex1 and Ex2 [10] are presented in figure 5. Due to the background conditions of the experiment Ex1, the events with the energy of the light fragment in the range $E_2 = (6÷30)$ MeV were selected (figure 5(a)).

Figure 5. FFs mass correlation distributions from $^{252}$Cf(sf) obtained in Ex1 – (a). Specific rhombic-spiral structure in the bottom of the figure (“nuclear rose”) is marked by an arrow. The distribution is compared with that obtained earlier in Ex2 [10] – (b).

The data from Ex1 as well as the presence of the lines at the mass numbers $M_2 = (128, 68, 72)$ u marked in the figure 5 by numbers 1, 2, 3 correspondingly, show some additional structures. We called a rhomic-spiral structure in the lower right corner of figure 5(a) that resembles a rose depiction the “nuclear rose”. It consists of the family of lines $M_1 + M_2 = \text{const}$ and several lines almost perpendicular to them. The lines of the first type (marked by the numbers 1–6 in figure 6(a)) will be analyzed in the next section while a discussion of the latter is below the scope of this paper.

Two structures from the figure 6(a) are presented in figure 7 in a larger scale. Expected prescission cluster composition of the decaying system for the events marked by the arrows is shown in the insets.
Each list of the clusters starts with the “missed” nucleus. Its mass corresponds to the missing mass in the expression $M_1 + M_2 = \text{const.}$

![Figure 6](image1.png)

**Figure 6.** Mass-correlation distribution from Ex1 – (a). The tilted numbered lines in white meet the condition $M_1 + M_2 = \text{const.}$ Projection of the distribution along such direction in the vicinity of line 6 – (b). The peak centred at 218 u marked by the arrow exceeds by more than two sigma the level obtained by smoothing the original spectrum.

![Figure 7](image2.png)

**Figure 7.** Two structures from the figure 6(a) in the larger scale. Cluster compositions for the events marked by the arrows are shown in the insets.

Different projections of the mass correlation distribution from Ex1 are shown in figure 8. As can be inferred from the figure 8(a) the statistics in Ex1 is approximately three times more than that in Ex2. A total yield of two Ni peaks in Ex1 does not exceed $10^{-4}$ per binary fission which agrees with our previously obtained value. The data of Ex2 indicated that the heavy clusters in the ternary prescission configurations are predominantly magic nuclei (table 1 in Ref. [16]). Noticeably large statistics in Ex1 allowed us to confirm previous observation. The projection of the distribution shown in figure 8(b) onto $M_1$ axis for the range of $M_2 = (65–76)u$ clearly demonstrates increased yield of the heavy fragments corresponding to the magic isotopes of $^{128}\text{Sn}$, $^{134}\text{Te}$, $^{140}\text{Xe}$, $^{144}\text{Ba}$, $^{150}\text{Ce}$, $^{154}\text{Nd}$ (their masses are marked in figure 8(b) by the arrows).
Figure 8. Projections of the mass-correlation distributions obtained in Ex1 and Ex2 onto $M_2$ axis – (a). Projection of the mass-correlation distribution from Ex1 onto $M_1$ axis under condition $M_2 = (65–76)$ u – (b).

3. Discussion
For the first time we had observed the linear structures meeting the condition $M_1 + M_2 = \text{const}$ in our experiment (Ex3) at the FOBOS setup based on the gas-filled detectors [9]. The linear ridges corresponding to the constant missing masses were revealed as a fine structure in the mass correlation plot collected with good statistics. Later, using the COMETA spectrometer based on the mosaics of PIN diodes, we also observed the structures under discussion (Ex2), but the data suffered from small statistics [10]. Parameters of all linear structures observed so far in the region of so called “Ni-bump” are presented in table 1.

Table 1. Parameters of the linear structures shown in figure 6(a).

| Str № | Missing fragment | Heavy magic core | Number of neutrons | Experiments |
|-------|------------------|------------------|-------------------|-------------|
| 1     | $^{62}\text{Cr}$ | $^{198}\text{W}$ | 116               | Ex1         |
| 2     | $^{48,50,54}\text{Ca}$ | $^{198}\text{Pt}$, $^{202}\text{Hg}$, $^{204}\text{Pt}$ | 120, 122, 124 | Ex1 & Ex2 |
| 3     | $^{44}\text{S}$ | $^{212}\text{Po}$ | 128               | Ex1 & Ex3 |
| 4     | $^{40}\text{Si}$ | $^{216}\text{Po}$ | 132               |             |
| 5     | $^{36}\text{S}$ | $^{218}\text{Po}$ | 134               | Ex1         |

As can be inferred from figure 9 demonstration the shell corrections map [17], the numbers of neutrons in the heavy magic cores (the forth column in table 1) manifest themselves through the discussed structures correspond to the valley of negative shell corrections. In the recent publication [18] on the study of pear-shaped nuclei, it was noted that the octupole correlations enhanced at the magic neutron numbers 34, 56, 88, 134. Thus, it is reasonable to expect that the nuclei having 116-134 neutrons would be pear-shaped and show magic properties. According to the calculations, for instance [19], the fissioning system becomes pear-shaped at the beginning of the descent from the fission barrier, at least in the most populated fission valley of the potential energy surface.
4th International Conference on Particle Physics and Astrophysics (ICPPA-2018)  
Journal of Physics: Conference Series 1390 (2019) 012011  
doi:10.1088/1742-6596/1390/1/012011

Figure 9. Shell energy diagram [18] depending on neutron number and deformation. Areas corresponding to negative shell energy are shaded, and the contour separation is 1 MeV.

We propose the following scenario of the process leading to the formation of the structures under discussion. At the initial stage of the descent from the fission barrier, the fissioning nucleus undergoes transformation from a prolate to a more complicated shape. The nucleus of such shape consists of a heavy magic octupole deformed core and the light cluster in contact with the thin edge of the “pear”. At further elongation, the core takes dumbbell-like shape with more and more long neck between the heavy and light parts of the dumbbell. The mass of the light cluster stays unchanged. After the first rupture in the neck, a heavy fragment, and a di-nuclear system consisting of the light cluster and some part of the neck fly apart. Later, due to the second rupture, the light cluster and the light FF become free. In general, the cinematics of the process seems to be similar to that discussed in Ref. [16].

4. Conclusions
Based on the new data, we clarify a mechanism of the CCT mode similar to heavy ion radioactivity [9]: octupole deformed magic core plays the same role as magic Pb cluster in the “Lead radioactivity”.

Clustering of heavy cold nuclei seems to be a promising field for further research with no less interesting and bright physics.

Acknowledgments
This work was supported, in part, by the Russian Science Foundation and fulfilled in the framework of MEPhI Academic Excellence Project (Contract No. 02.a03.21.0005, 27.08.2013) and by the Department of Science and Technology of the Republic of South Africa (RSA).

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