TRUE TERNARY FISSION, COLLINEAR CLUSTER TRI (CCT) PARTITION OF ^{252}Cf*

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"Ternary fission" has been studied previously as a process, where a lighter fragment with $A < 30$ amu is emitted perpendicular to the fission axis. We use the term "true ternary fission" for a fission process with three masses of comparable size, $A > 35$. Theoretical considerations repeatedly predicted that true ternary fission must occur in a collinear (prolate) geometry. Over the last decade (see Pyatkov *et al.* in *Eur. Phys. J.* **A45** 29 (2010) and refs. therein), the true ternary fission decay in neutron induced fission ^{235}U (n,fff) and in spontaneous fission of ^{252}Cf (sf) has been studied. The name used for this process is (CCT), with three cluster-fragments of similar size in a collinear decay. The measurements are based on binary coincidences containing TOF and energy determinations, in two detector telescopes placed at 180°. The masses and energies of the registered two fragments give complete kinematic solutions. The missing mass events in binary coincidences can be determined, these events are obtained by blocking one of the lighter fragments (after multiple scattering in the backing) on a structure in front of the detectors. The relatively high total yield of CCT (more than 10^{-3} per binary fission) is explained. It is due to the favorable Q-values (more positive than for binary) and the large phase space of the CCT-decay, dominated by three (magic) clusters: *e.g.* isotopes of Sn, Ca and Ni. The kinetic energies of the fragments have been calculated. The fragments placed in between the other two have very low kinetic energies (below 20 MeV) and have thus escaped their detection in previous work on "ternary fission", where in addition an oblate shape and a triangle for the momentum vectors had been assumed.

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1. Introduction

Nuclear (binary) fission is a process, where a heavy nucleus decays into two fragments of intermediate mass (*e.g.* Ba+Kr), it has been identified by Hahn and Strassmann in 1938. It was discovered by chemical analysis, while irradiating natural uranium with neutrons [1]. The energy release is very large, typically 200–205 MeV (*e.g.* Meitner and Frisch [2]). This has been deduced from the liquid drop model and the nuclear mass formula of Bethe and Weizsäcker [3]. However, the large collective motion through a large prolate deformation, today called super- and hyper-deformation, was considered at that time to be unlikely.

Fission of heavy low-excited nuclei into *three* fragments of comparable masses, so-called "true ternary fission", has been predicted soon after the discovery of fission. Swiatecki [4] has shown within the framework of the liquid drop model, that fission into three heavy fragments is energetically more favorable than binary fission. For all nuclei with fission parameters $30.5 < Z^2\ /\ A < 43.3$ ternary fission can be expected. In 1963 Strutinsky [5] has calculated the equilibrium shapes of the fissioning nuclei and has shown, that along with the ordinary deformed configurations with one neck, there are a variety of more complicated elongated configurations, with two and even three necks. Later in 1974 Diehl and Greiner [6] have shown, that for ternary fission prolate collinear configurations are preferred. The same result has been obtained by Royer in 1992, see Ref. [7] and Manimaran *et al.*, see Ref. [8]

The Coulomb interaction in the potential energy is the smallest [8] for *linear arrangements* of the three charged fragments. Furthermore, results demonstrating the decisive role of shell effects in the formation of the multi-body chain-like nuclear molecules were obtained by Poenaru *et al.* [9, 10]. From a more recent calculation of the fragmentation potentials [8], we can compare the yields of oblate (triangular) and of prolate (collinear) pre-fission shapes. This is shown in Fig. 1, where we can see the preference (note the vertical scale with steps of 5 orders of the magnitude) for collinear ternary splits, which occur with masses of the third (middle) fragments heavier than *ca.* $A = 30$ and charges larger than $Z = 16$.

Extensive and comprehensive searches for ternary fission has been done at the ILL with the Lohengrin mass separator [11], which gives complete mass separation of the registered fission fragments. As an example, we cite the case for the the neutron induced fission, ^{249}Cf$\,$(n$_{nt}$,f) (see Ref. [12]). In this work, the absolute yields of ternary fragments down to the levels of 1×10^{-8} for neutron-rich isotopes have been obtained. The range of masses of ternary fragments with $A = 10$ (^{10}Be) up to $A = 33$–37, like ^{37}S, has been covered. The name "ternary" fission has been used for decays, when a third light particle is emitted perpendicular to the binary fission axis [11]. For this type of decay, an experiment to detect three coincident masses with the possible heaviest third particle, has been performed by Schall *et al.* [13] with four segments (covering a range of 80°) of gridded ionisation chambers. In this work, an oblate pre-fission shape is assumed and ternary coincidences were searched for. The ionisation chambers have been previously used in a system called "Diogenes", they imply a lower energy threshold of 25 MeV, which was set in order to reduce background. This experiment gave a lower limit of 1×10^{-8}, relative to binary decays, for true ternary decays. The yield with this limit is connected to masses of the third particle (and relative angles of *ca.* 120°) at around $A = 35$. This experimental result once more emphasizes as discussed above, that *true ternary fission of heavy nuclei is only possible in a collinear geometry.*

In our experiments described below, with two heavier fragments in coincidences at 180° we observe, due to a particular experimental set up, the missing mass in a ternary collinear decay.

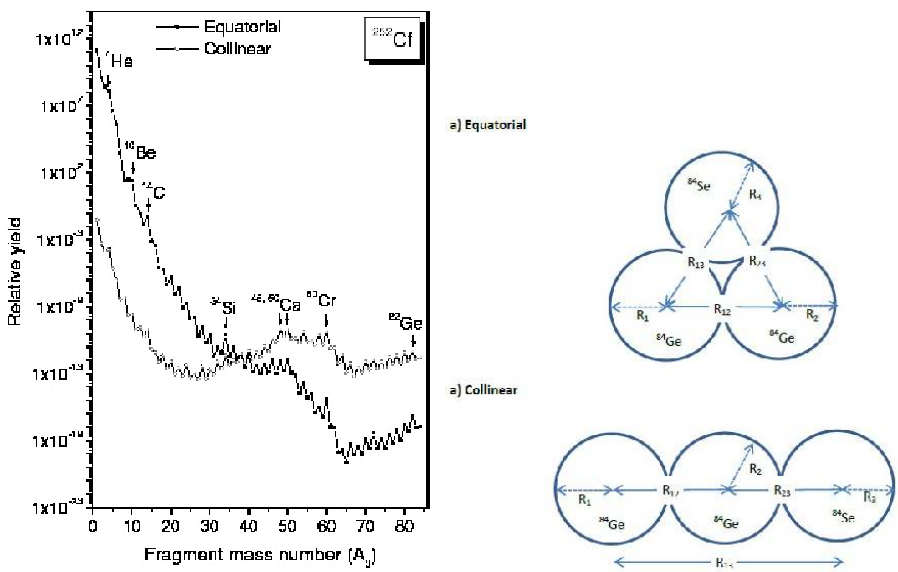

Fig. 1. From the fragmentation potentials for the decay of ^{252}Cf into three fragments of similar size the yields in a statistical decay are obtained. The collinear (prolate deformation, a chain), and the oblate fission shapes are compared. The corresponding relative yields (note the steps of 10^{-5}) of the ternary decay process of ^{252}Cf are shown as a function of the ternary mass ($A3$), of the third (middle) fragment, as in Fig. 2. The shapes are shown on the right-hand side. With the crossover of the two curves at $A3 = 35$ the yield for ternary decays with prolate deformations, the collinear decay, will prevail. (Courtesy of Balasubramanian [8]).

2. Dynamics and phase space of the CCT-decay

We have seen in Section 1 (see, in particular, also Ref. [4]), that the fission decay for heavy nuclei into a number of fragments n larger then 2, will be collinear, due to the dominant Coulomb interaction. Further, we analyze the phase space of the CCT decay [14]. We assume that the ternary decay occurs in two steps from a hyper-deformed prolate shape, as illustrated in Fig. 2. We use energy and momentum conservation in each step of the sequential decay — in this way we are able to calculate the energies of the fragments. The Q-values for ternary decays are larger than for the binary mass split, see Fig. 2, where we define the sequential decay products $A1$, $A2$ and $A3$. Variations of the neutron number in $A1$ and $A3$ give a region for $A1 \times A3 = (128\text{–}134) \times (54\text{–}46)$ with high Q-values, defining a large phase space for ternary decays (46 isotope combinations with high Q-value): Due to high Q-values, fragments can be created in excited states, with levels with spins up to $J = 6$. The spin multiplicity of such fragments will allow for an additional factor for a larger phase space in the ternary decays. In total, an additional factor of 1000–3000 to be multiplied to the probability of the single isotope emission, can be expected. The CCT decay, observed [15] as a bump in the $M1$–$M2$ correlations (see Fig. 5), can thus have a total probability of 3×10^{-3}.

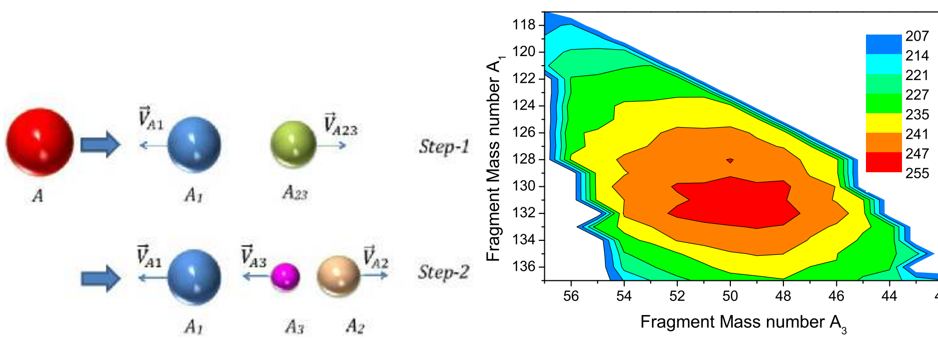

Fig. 2. The sequential ternary fission decay of ^{252}Cf(sf) leading to three fragments of medium size [14]). We use the label A_3 for the central fragment. Right-hand side: the Q-values, defining the phase space for the ternary decay (momentum range, the number of mass partitions, *etc.*).

3. The experiments, missing mass method

The results of the first two experiments with the FOBOS-detectors built in the FLNR in Dubna [16], have been reported in Refs. [15, 17]. New experiments (Ex3 and Ex4) have been performed with pin diodes (system called

COMETA, see Fig. 3) replacing the Bragg ionization chambers and with neutron coincidences [17]. The blocking medium for the scattered fragments in this case are the frames of the individual pin-detectors, see Fig. 3 for some details. Actually, the efficiency for the registration of a ternary decay process with blocking in this case is much lower than with the FOBOS-detectors, see Ref. [17].

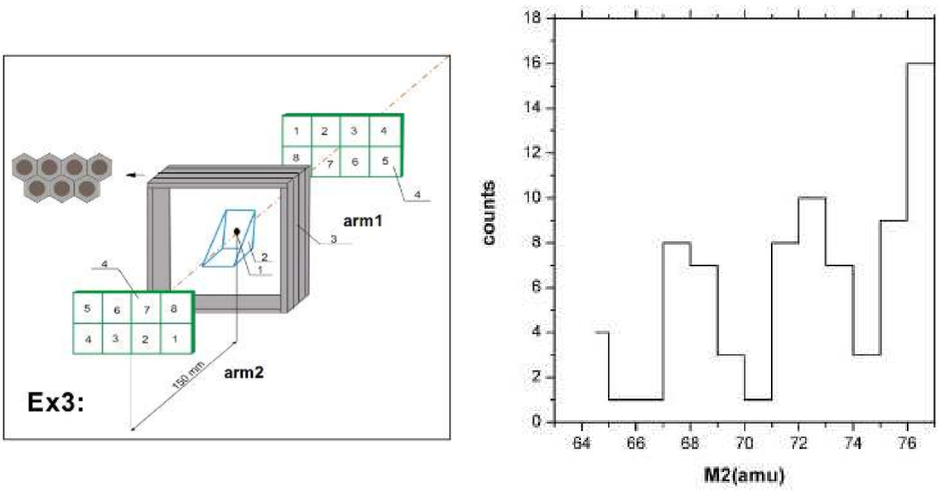

Fig. 3. Left: The COMETA set-up for the registration of two fission fragments in coincidence in two pin-diodes and a neutron detector (belt) perpendicular to the fission axis. The binary fission mass peaks have no background in this case. Right: it shows the projection of the mass yields for masses, shown in Fig. 5, (right side) and below the binary mass peak. Two lines in the mass range of 68/72 become visible, they are absent in arm2. The mass resolution in this case is $\Delta m = 1.8$ amu.

The experiments [15, 17] are based on binary coincidences between two fragments with a system containing two detectors (FOBOS-modules) placed at a relative angle of 180°, see Figs. 4. The masses of two reaction products are obtained independently with the TOF-E (time-of-flight *vs.* energy) method. The measurement is kinematically complete for events with a relative angle of 180° and allows to determine the sum of the masses uniquely. If the sum of the masses is smaller than the initial mass, it is called “the missing mass method”, this has been applied in all our experiments. For the case of a missing mass, ternary and other multi-body decays can be detected. The method is often used in high energy physics, when a new particle is not observed in coincidences with a particular detector, the reconstruction of the event may create events with missing energy (or mass) of the new particle.

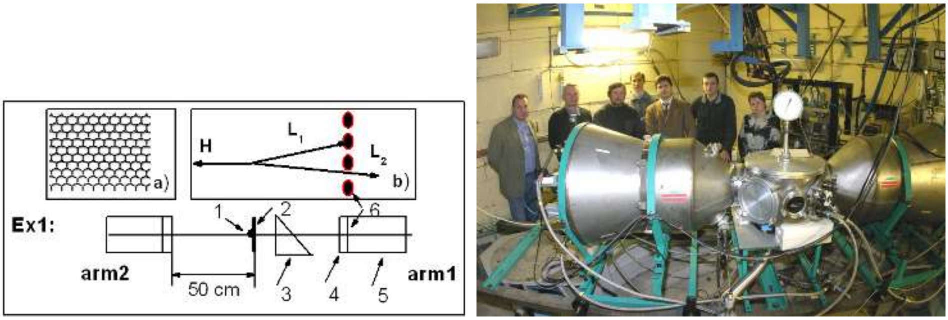

Fig. 4. Scheme of (Ex1) for coincidence arrangement for the measurements of two fragments of the fission decay of ^{252}Cf, with two arms (arm1, and arm2). Here: 1 — Cf source, 2 — source backing, 3 — micro-channel plate (MCP) based timing "start" detector, 4–6 FOBOS detectors, Bragg-ionisation chambers, 4 — position sensitive avalanche counter (PSAC) as "stop" detector, 5 — ionization chamber (BIC) with the supporting mesh, 6 — the mesh of the entrance window. The front view of the mesh is shown in the insert (a), an enlarged mesh section is presented in the insert (b). After passage of the two fragments through the source backing, two light fragments L_1 and L_2, are obtained with a small angle divergence due to multiple scattering. In (b) we show that one of the fragments (L_1) can be lost hitting the metal structure of the mesh, while the fragment L_2 reaches the detectors. The source backing (2) causes the mentioned angular dispersion in the direction towards the right arm1. On the right-hand side, a photograph of the FOBOS-modules is shown.

The measurements are based on a specific feature (see scheme shown in the left-hand side in Fig. 4), which allows the blocking of one of the three fragments, by multiple scattering in only one (see Fig. 4) of the arms. The essential experimental circumstance for the missing mass method is the multiple scattering in the source backing (pointing to arm1), of two fragments (of the three fragments) traveling to arm1. This causes an angular spread in such a way, that only one of the two fragments emitted towards arm1, reaches the ionization chamber, whereas the other gets blocked by the specific support structure in front of the ionization chambers. The results are presented as correlation plots of the two registered masses, as shown in Fig. 5. In this case, the yields are concentrated in the two binary fission yields, with very strong scattering effects due to the grids at the entrance of the FOBOS detectors. There is a distinct difference in the structure observed in arm1 and arm2, only in the first the ternary bump (covering $M2$: 138–144 with $M1$: 60–70) for the missing masses (in the region of $M3$: 38–54) can be observed. These are most likely S and Ca isotopes. To obtain the CCT yields we use the difference (arm1–arm2) to obtain the signal for the ternary fission events [15]. In the first experiment in total 13×10^6

events have been collected (3000–4000 for the ternary fragment). Quite similar results are obtained for the neutron induced fission in the ^{235}U (n,ff) reaction, the results are shown in Fig. 6. With projections on the M_1-axis or the M_s-axis, we get a strong signal for the CCT-effect. The measurements with neutron coincidences and pin-diodes (COMETA) for ^{252}Cf (sf) gave similar results, but with smaller statistics, see Ref. [17]. There is literally no background because the pin-detectors have no frames in front of them, which has produced background. The plot shown in Fig. 5 illustrates these facts. It is important to have some statements on the mass resolution of the experiment. This is obtained by using alpha-particles to determine the time resolution. For the PSACs a resolution of 400 ps is obtained, for the pin-diodes, also with the alpha particles, a time resolution of 2.7% is obtained, which depends on the flight-path. The structures observed in Fig. 5 point to the fact, that the ternary decay modes are dominated by nuclei with "magic" numbers for protons and neutrons.

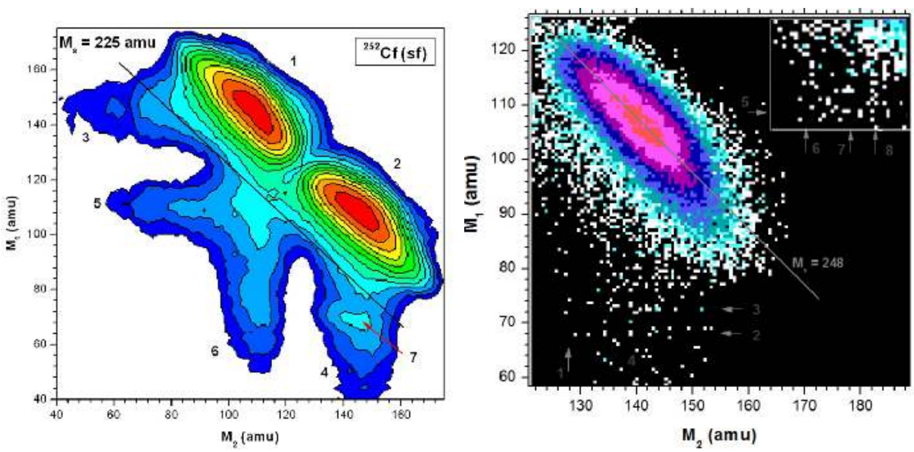

Fig. 5. Left part: The $M1$–$M2$ mass correlation for two fission fragments in coincidence in two FOBOS detectors, see Fig. 4. The two binary fission mass peaks have their maximum at a sum $M_s = 250$, well above the line showing the total mass of $M_s = 232$, which separates the main binary yield. The missing mass bump in arm1 is marked by an arrow (masses $M1 = 68$ and $M2 = 144$), it is absent in arm2. Right-hand side: measurements with the COMETA detectors (Fig. 3) in Ex4. Below the fission bump (in arm1), of the binary fission of Cf in the correlation ($M1$, $M2$), we observe two horizontal lines (marked by arrows), which we attribute to Ni (or Cr) isotopes, see Fig. 3.

4. Neutron induced fission for ^{235}U (n,ff)

For the systematics of the experimental feature with the blocking method to observe CCT events with missing mass, it is quite important to have independent experiments. We have performed such an independent experiment with the neutron induced fission in ^{235}U (n,ff). The FOBOS detectors are smaller (Mini FOBOS) than in experiment Ex1. In the uran-experiment, due to another geometry (see [15]), we have some slightly different background from fission fragments scattered from the metal support structure as illustrated in the scheme of Fig. 4. The latter separates together with a thin foil the region of higher pressure in the Gas–Bragg-ionization chamber from the region with low gas pressure in the parallel plate counters (PPAC), see also Ref. [15]. For the blocking mechanism we rely on this metal structure in front of the PPACs, and on the multiple scattering in the target support. In total, 4.7×10^{-3}/(binary fission) CCT events as the difference between arm1 and arm2, have been recorded.

Also in this experiment with the target of 100 μg/cm^2 uranium, the support has been an Al_2O_3 backing of 50 μg/cm^2 pointing to arm1. The target was integrated in a special start detector to obtain a very compact device. In total 1.6×10^6 events have been recorded.

The lower part of Fig. 6 shows the projections, on the M_s-axis (sum of the registered binary fragments) in the missing mass regions for the Ex1(Cf), for the Ex2(U) and for the Cf (sf,fff) reaction in coincidence with neutrons, marked here as Ex2, and projections on M_1, the axis of fragments registered in arm1. For comparison, the missing mass spectrum of Ex1 Cf (fff) is also shown. We find for the ^{235}U (n,ff) reaction a missing mass bump of similar strength as in Ex1. There is a clear shift in the total mass between the fission of ^{236}U and the Cf-spontaneous fission. The masses projected in the M_1-scale, show the same position of the maximum in the neutron-rich Ni isotopes, around $m = 68$–70. However, in the total mass projection on M_s, there is a distinct shift of the maximum to lower values of M_s for the case of U. This points to the fact, that now the total mass is smaller (compared to the Cf case) by 16 mass-(and 6 charge)-units. The difference between, Cf and U emphasizes the persistence of the Sn- and Ni-clusters. Thus the decay process dominantly produces a smaller ternary missing mass, the isotopes of S and Ca. In “Ex2” described in Ref. [17] the spontaneous fission has been studied in coincidence with neutrons. The registration of neutrons reduces the background, the final yield is obtained by subtraction of the yields in the two arms, arm1–arm2. Also in this case an extremely reduced background appears for the measured neutron multiplicity $n = 2$. With this value, a higher primary rate of neutrons $n = 4$ or 5, must be assumed. These are mostly prescission neutrons, due to the geometry of the neutron counters pointing to the center of mass of the fissioning system. This reduces the probability of a coincidence rather drastically.

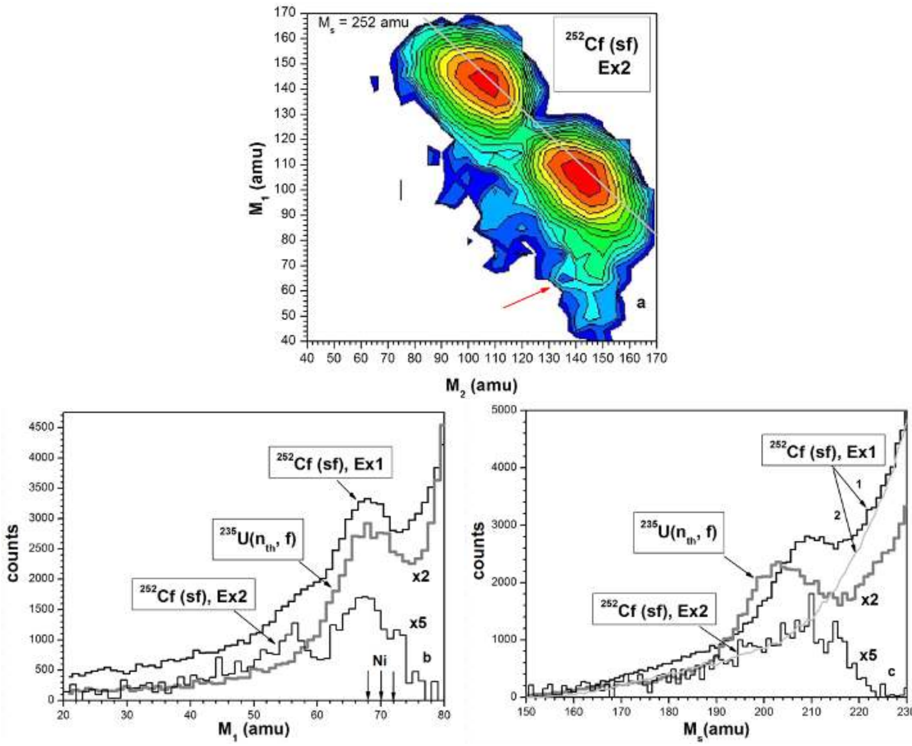

Fig. 6. Spontaneous fission in coincidence with neutrons of ^{252}Cf (ff) in comparison with the fission reaction ^{235}U (n,ff) measured with the FOBOS (mini FOBOS) set-up. For technical details we refer to the paper on CCT by Pyatkov *et al.* [15]. The projection on M_1 shows the dominance of the Sn and Ni-clusters, the projection on M_s shows for ^{235}U(n,ff) the shift towards smaller mass, implying a smaller mass for the ternary fragments (isotopes of S and Ar).

5. Conclusions

We have seen that the theoretical predictions about true ternary fission [4–8, 10] are quite numerous, and they all come to the same conclusion: for systems with large total charge (or fissibility parameter Z^2/A) the emission of the ternary (additional) fragments and further multiple decays will be collinear along the primary (binary) fission axis. In particular, the survey of Royer *et al.* [7] emphasizes the conditions for the collinearity of the ternary decay.

The experimental results on true ternary collinear fission presented here rely on the missing mass method. This is only marginally influenced by the very low kinetic energy of the "third" fragment. Experiments with a registration of all three fragments are highly desirable. The best way to study the collinear ternary (or multiple) decay in a unique way will be by fission in flight, in experiments at the fragment separator FRS (at GSI (see Refs. [18, 19]) and with the FRSs at MSU, RIKEN, FAIR and other future facilities. These experiments can be performed at energies of 100 MeV/u or even higher at around 1.0 GeV/u. In this case, the large momentum of the center of mass of the decaying nucleus will boost all decay vectors to energies allowing the registration in a multiple coincidence of all three (or more) fragments.

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