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PARTICULAR ASPECTS OF PARTICLE-ACCOMPANIED FISSION

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Abstract. A brief survey is presented of particular recent experiments on particle-accompanied fission. Very recently, a new experiment was performed to re-measure the ternary α-particle energy spectrum in ^{252}Cf spontaneous fission by time-of-flight techniques. Compared to many previous experiments, which entailed detection thresholds of 6 to 9 MeV α-particle energy due to the $\Delta E - E$ method applied and use of protection foils on detectors, the energy distribution of ternary α-particles could, for the first time, be measured down to 1 MeV. We, furthermore, report on experimental studies on quaternary fission in spontaneous fission of ^{252}Cf, on the one hand, and the neutron-induced fission reactions ^{233,235}U(n$_{th}$,f), on the other hand. Finally, in a third experiment, ternary fission in the reaction ^{235}U(n,f) with cold polarised neutrons was under investigation. From peculiar but unmistakable shifts in the α-particle angular distribution when flipping the neutron spin the fissioning nucleus was deduced to undergo rotation at scission. This startling new phenomenon was dubbed the "ROT" effect.

Key words: ternary fission; ^{252}Cf(sf); E distributions of ternary ^{4}He and ^{6}He; quaternary fission in ^{252}Cf(sf) and ^{233,235}U(n$_{th}$,f), ROT effect in ^{235}U(n$_{th}$,f).

1. INTRODUCTION

Whether by spontaneous or neutron-induced nuclear fission, the mother nucleus disintegrates in the overwhelming fraction of cases just into two fragments. Fragmentation into three or more daughter nuclei of about equal mass has up to the present not been detected unambiguously [1]. Sometimes, however, instead of the standard "binary fission" a higher-multiplicity process with three or more charged particles in the outgoing channel is observed, but with the accompanying particles being very light compared to the fission fragments proper. With probabilities at the 10^{-3} level, one light charged particle (LCP), mainly an α particle, accompanies the customary fission fragments (FF), the process being known as ternary fission (TF) (see reviews [2,3]). An even rarer particle-accompanied fission mode, with probabilities in the range of 10^{-7} to 10^{-6}, is quaternary fission (QF), where a pair of LCPs is simultaneously emitted in one fission event [4]. Intriguingly, the two LCPs (mainly either two α particles, or an α and a triton) measured in one fission event can originate not only from their simultaneous creation in the fissioning nucleus ("true" quaternary fission) but also from the sequential decay of short-lived particle-unstable states in heavier ternary LCPs, also called "pseudo" quaternary fission. Extrapolating the series binary-ternary-quaternary one may ask whether there is a chance to discover in low energy fission also quinary fission where, by definition, 5 charged particles appear in the final state. The yield estimated for quinary fission is not larger than 10^{-10} / f and hence at the limit of feasibility for experiments.

As known from ternary fission, there is ample evidence from experiment, and also from theory, that by far the majority of light particles are born right at scission in the neck region of the nascent fission fragments [5]. Thus, although being a rather rare process, particle accompanied fission provides one of the few means to the experimentalist to explore the behaviour of fissioning systems near the point of rupture (scission). In the following, we are going to present a brief survey of particular recent experiments on the issue and the progress achieved.

2. **ENERGY DISTRIBUTION OF TERNARY α-PARTICLES FROM ^{252}Cf(SF)**

Since the discovery of ternary fission in the 1940s, there have been numerous experiments to measure energy spectra of ternary particles [2,3]. Nevertheless, surprisingly little is known about the low-energy part of these distributions. The precise shape of the low-energy tailing of the spectrum has been at issue for decades [6]. The yield at low-energy is particularly significant. Low-energy α particles presumably arise from low α-particle initial energies or more stretched scission configurations of the main fragments. They thus may provide important insight into both, the emission mechanism of ternary particles and the scission stage of the fission process. However, particle-unstable ternary particles (e.g., ^{5}He and ^{8}Be [7]) may also give rise to low-energy α's in sequential processes, and thus mask the true ternary particle emission. Without any doubt, precise experimental data are highly needed to address the problem further.

Experimental studies at low energy are still scarce, and the data are not consistent. This is true also for ternary α-particles from the spontaneous fission (sf) of ^{252}Cf – one of the most often studied decay processes [8,9]. The main reasons for this situation are twofold. First the intense background from the alpha decay of ^{252}Cf that has forced many researchers to use protection foils on the detectors cutting α-particle spectra at and above 6 MeV. The second is the preference given to the $\Delta E - E$ method to identify ternary particles. The method does not allow to explore energy distributions at energies below ΔE. Such rather high threshold values compared to the 16 MeV mean energy do not only cut away the interesting low-energy part of the spectrum but leave also substantial ambiguity in the energy assignment of the above threshold events due to uncertainty in absorber thicknesses and related energy losses.

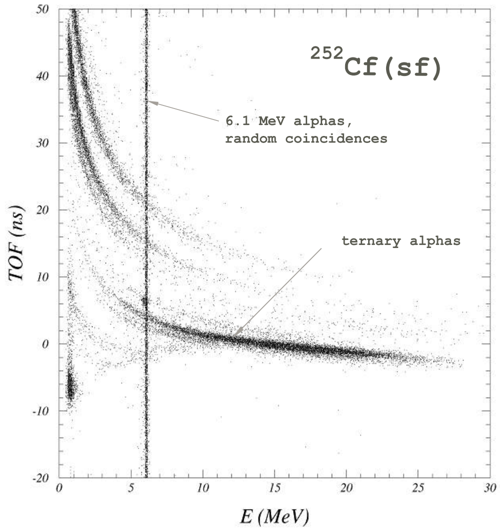

Fig. 1 – Scatter plot ToF *vs.* E of ternary particles in ^{252}Cf(sf), as measured with 10 silicon p-i-n diodes of 380 μm thickness and 30×30 mm^2 in size, located at 20 cm distance from the source. Time is in ns with respect to the time of 16 MeV α particles; energy E is in MeV.

At the Physics Department of Jyväskylä University, Finland, in a new experiment the ternary α-particle energy spectrum from ^{252}Cf(sf) was measured using an array of silicon detectors and discriminating ternary α particles from neighbouring isotopes by time-of-flight (ToF) techniques using fission fragments as the start [10]. No protection foils were used in front of the semiconductor detectors. There is thus no material between the open side of the

^{252}Cf source and the surface of the energy detectors that could slow down the ternary particles, even at low energy. To ensure that the energy spectrum measured corresponds to all equatorial α particles, for the present counting geometry a correction was applied (with the aid of the GEANT4 code from CERN) for the energy dependence of the angular distribution of equatorial ternary α-particles with respect to fission fragments as measured by Heeg *et al.* [11].

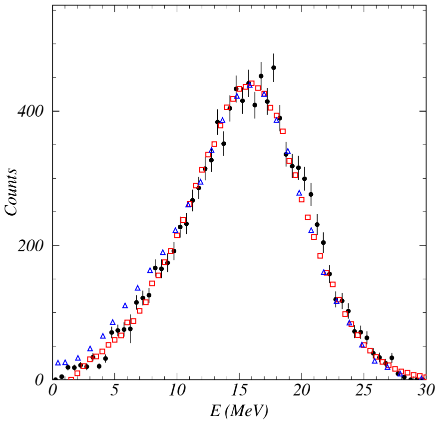

Fig. 2 – Energy distribution of ternary α particles from ^{252}Cf fission: Comparison of the present data (points with error bars) with results from Tishchenko *at al.* (squares) [9] and Loveland (triangles) [8].

With the novel experimental approach the energy distribution of ternary α-particles in ^{252}Cf(sf) was, for the first time, measured reliably down to 1 MeV. In the obtained spectrum at energies below 9 MeV there is an excess in the yield as compared to the Gaussian shape that was frequently used to fit the experimental data. The spectrum agrees with the recent measurement by Tishchenko et al. [9] but contradicts the earlier data by Loveland [8] where the reported excess at the low-energy yield of ternary α particles was too high (Fig. 2). For the first time, the full energy spectrum of ternary ^{6}He-particles was also deduced, although still with rather poor statistics [10].

The present study clearly demonstrates the low-energy tailing and settles the issue experimentally. The physical explanation, however, is still an open question. Shown in Fig. 3 are also 2 Gaussians which result from a fit to the present data above 9 MeV, and consider besides true ternary α particles (the dominant peak) also the about 17% contribution of residual α particles from the decay of ternary ^{5}He, as was measured recently by Kopatch *et al.* [7]. It is obvious from the data presented in Fig. 3, that the ternary α spectrum

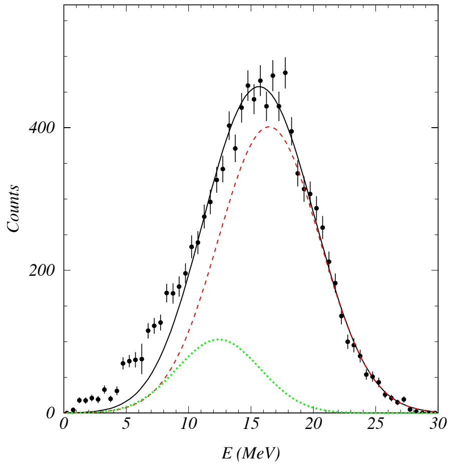

Fig. 3 – Energy distribution of ternary α particles from ^{252}Cf fission [10]. The two Gaussians fitted to the data above 9 MeV take true ternary α's (the dominant peak) and residual α's from ^{5}He decay into account [7].

shows more low-energy α particles than would be predicted by the sum of Gaussians. Apparently, the spectral shape measured previously at energies $E > 9$ MeV [6,7] can not be extrapolated meaningfully to lower energies.

New even more sophisticated measurements are needed to decide whether low-energy tailing is a common issue to most ternary particle species, and is thus possibly decoding particularities of the nuclear configuration at scission, or rather a special feature of α-particle accompanied fission. Application of the present technique for neutron induced fission reactions where interference with α's from radioactive decay is generally less, e.g. in ^{235}U(n$_{th}$,f), is also to be envisaged.

3. QUATERNARY FISSION IN ^{252}Cf(sf) AND ^{233,235}U(n$_{th}$,f)

An even rarer particle-accompanied fission mode with probabilities down to the level of 10^{-6}/fission and below is quaternary fission (QF). Here a pair of LCPs is emitted apparently simultaneously in one single fission event. Probably due to the low yields, QF has been barely studied in the past. Only a few experiments are known from literature both, for thermal-neutron induced fission of ^{235}U [12, 13], and spontaneous fission of ^{252}Cf and ^{248}Cm [16, 17]. As regards the theory of QF as a generalization of TF, it has been conjectured that the Rayleigh instability of cylinder-like necks may lead to QF for sufficiently heavy nuclei like the actinides [14]. In a different approach the probability of multi-cluster accompanied fission has been postulated from inspecting the energetics of scission configurations with several clusters in the neck region at a time [15].

Quaternary fission in ^{252}Cf(sf) and ^{233,235}U(n$_{th}$,f) was studied with two different experimental set-ups [4]. Figure 4 sketches the set-up for the ^{252}Cf(sf) measurement. The measurements on ^{233,235}U(n$_{th}$,f) were performed at the cold neutron beam PF1 at the high-flux reactor of the ILL Grenoble, France. Very high binary fission rates of more than 10^6 fissions/s could be achieved, and, hence, ternary particle rates on two large arrays of semiconductor detectors were almost 10^3/s. Thus, besides α-α and α-t coincidences events with t-t coincidences could be registered (Fig. 5). In both experiments, angular distributions and correlations of two light charged particles accompanying the two main fission fragments were measured. Likewise the energy spectra of the LCPs could be taken.

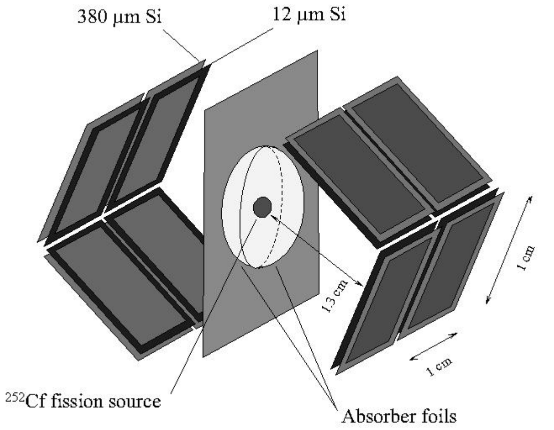

Fig. 4 – Experimental set-up for measuring α-α and α-t coincidences in ^{252}Cf. LCP identification is performed by a ΔE–$E_{\rm rest}$ measurement. The source was covered with 20 μm kapton for protecting the 8 detector telescopes from ^{252}Cf α's [4].

Similar to the neutron-unstable LCPs like ^{5}He, there exist other short-lived particle unstable LCP species that decay close to the fissioning nucleus and, thus, escape from direct observation. The most prominent example is the ^{8}Be decay into two α-particles, from its ground state and the 3.13 MeV excited state. Besides this basically ternary decays which in a secondary process become quaternary (so-called “pseudo” QF), the question arises whether “true” QF with the independent emission of two charged particles right at scission exists. In the present experiments, the two varieties of QF have been differentiated from one another by exploiting the different patterns of angular correlations between the two charged LCPs. Yields and energy distributions of LCPs for each of the two processes were obtained here for the first time in one and the same experiment [4].

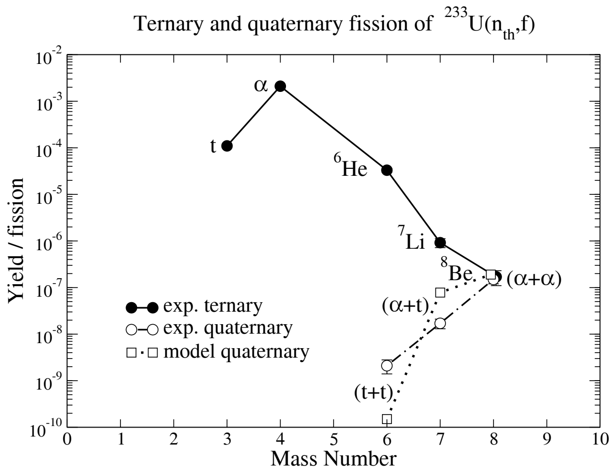

Fig. 5 – Quaternary fission yields in ^{233}U(n$_{th}$,f), relative to the corresponding yields of the composite ternary particles. In the model calculation the law of mass action was assumed to be valid for the number of nucleons forming the LCPs. The temperature being requested for applying the law was deduced from the measures yield ratio ^{7}Li*/^{7}Li [21].

As to the energy distribution of α-particles it is remarkable that for the three reactions under study both, the mean values $\langle E_\alpha \rangle$ and the widths FWHM, are very similar to each other for any of the decay modes in TF or QF. However, compared to the α-energies from ternary decay, it is apparent that for simultaneous (α,α) QF and for the residual α's from sequential ^{8}Be decay the average energies are down by about 2 - 3 MeV and about 6 - 7 MeV, respectively. The smaller energies in the case of true QF may indicate that, on average, the deformation of the scission configuration is larger and the main fragments are farther apart in QF than in TF. Indeed, for fragments farther apart, the Coulomb forces accelerating the α-particles will be smaller. Since in QF two α-particles have to be accommodated in the neck region between the two main fission fragments, larger overall deformations and, hence, lower kinetic energies are quite understandable.

As to the yields, for all different QF reactions which could be explored, the yield for the ^{252}Cf(sf) reaction is roughly by an order of magnitude larger than the yields found in the two ^{233}U(n$_{th}$,f) and ^{235}U(n$_{th}$,f) reactions. The difference in the QF yields for the heavier nucleus ^{252}Cf compared to the two lighter ^{234,236}U*-isotopes becomes even more pronounced when the yields are normalised to binary fission. The ternary α-particle yields for the reactions under study have been reported as $3.3 \cdot 10^{-3}$, $2.1 \cdot 10^{-3}$, and $1.7 \cdot 10^{-3}$, for ^{252}Cf, ^{234}U*, and ^{236}U*, respectively. When normalised to binary fission, the enhancement of QF in ^{252}Cf(sf) is found to be in excess of an order of magnitude. Recalling the systematics of ternary fission yields (e.g., ref. [18]) for fissioning compounds of increasing mass (or charge or fissility), the higher QF yields for ^{252}Cf are not surprising. Indeed, the heavier the compound nucleus is, the larger become the yields for ternary particles of increasing mass. A lesson to be learned from this observation is that a search for quaternary fission with ejectiles heavier than α-particles or for "quinary" fission with three lighter particles accompanying the main fragments should be more successful for heavy compound nuclei.

For pseudo QF mediated by ^{8}Be it has been found that the ratio of ternary yields $Y(^{10}\mathrm{Be})/Y(^{8}\mathrm{Be})$ varies smoothly for the three reactions under study. For ^{252}Cf(sf), ^{235}U(n$_{th}$,f) and ^{233}U(n$_{th}$,f) one finds values for the ratio $Y(^{10}\mathrm{Be})$ / $Y(^{8}\mathrm{Be})$ of about 12, 36 and 46, respectively, while the absolute yields are largely different. Conspicuously the ^{8}Be yields are for all reactions studied at least one order of magnitude lower than the ^{10}Be yields. The low ^{8}Be yields are most remarkable since Q-values having been calculated for the three reactions at hand are definitely larger for ternary fission with ^{8}Be than with ^{10}Be as the ternary light particle [19]. The difference in emission probability tells that it may be ambiguous to rely exclusively on Q-values for predicting LCP yields. The experimental values for the ^{8}Be/^{10}Be yield ratio could, hence, become a cornerstone for testing any theory of ternary fission. Furthermore, as has been pointed out in [4], an interesting by-product has been the measurement of yields of excited LCPs which allows to deduce nuclear temperatures at scission by comparison to the respective yields in the ground state.

4. TERNARY FISSION AS A TOOL TO STUDY FISSION OF ROTATING NUCLEI

Fission reactions induced by polarised cold neutrons have so far mainly been used to study symmetry laws in fission like e.g. parity conservation [20]. Searching, furthermore, for a violation of Time-Reversal-Invariance in ternary fission, a hitherto unknown asymmetry in ternary α-particle emission was observed these last years. The asymmetry could be attributed to the influence of the Coriolis force present in a rotating nucleus which is about to undergo fission and which, besides the two main fragments, is ejecting an α-particle. The effect was found in the reactions ^{233}U(n,f) and ^{239}Pu(n,f) induced by polarised cold neutrons from the PF1 beam line installed at the high-flux reactor of the ILL, Genoble, France. It was called the "TRI" effect [22].

In continuation of these experiments for the ^{235}U(n,f) reaction, surprisingly a startling new phenomenon was observed. As deduced from peculiar but unmistakable shifts in the angular distributions of the α-particles the nucleus is rotating. It was concluded that the fission axis, i.e. the axis along which the fission fragments are flying apart in opposite directions, is rotating in a plane perpendicular to the spin of the polarised neutron. Since following capture of a polarised neutron also the compound nucleus will be polarised, a collective rotation of the fissioning system appears to be quite feasible. However, following scission the fragments are accelerated within times of some 10^{-21} s up to their eventual velocity of about 1 cm/ns. The moment of the rotational inertia will increase with a similar time constant. For fragments at infinity also the moment of inertia will be infinite. Hence, for a given angular momentum the angular velocity will very quickly come to a virtual stop. For a typical angular momentum of 1 $\hbar$ simple estimates yield integrated total angles of rotation of much less than 1°. The surprising fact is possibly less that nuclei undergoing fission induced by polarised neutrons are rotating than the power of the spin flip technique allowing to measure angles of rotation well below 1° in a violent nuclear reaction like fission. Trajectory calculations for α-particles moving in a rotating Coulomb field provided by the two main fragments confirmed the interpretation. The phenomenon was dubbed the "ROT" effect. Furthermore, in a newly developed quantum theory of ternary fission the main characteristics of both, the TRI and the ROT effect could be reproduced. For theory, however, the challenge will be to understand in detail which properties of the fissile nuclei govern the outcome of the ROT effect [23, 24].

In the present experiment ternary fission in the reaction ^{235}U(n,f) with cold polarised neutrons was under investigation. A characteristic feature of this process is the angular distribution of the ternary particles which is centred roughly perpendicular to the fission axis. The experimental setup takes these angular correlations into account. In the experiment a thin ^{235}U target is irradiated by a neutron beam with spin polarisation pointing either parallel or anti-parallel to the beam direction. Fission fragments and ternary particles are intercepted near a plane perpendicular to neutron beam and polarisation. If now the nucleus undergoing fission is rotating, the α-particles will experience a rotating Coulomb field and at least partly will follow the rotation. Their trajectories will hence be slightly bent which means that their angular distributions will be shifted as compared to a non-rotating nucleus. In experiment the fact is exploited that, upon flipping the neutron spin, also the sense of rotation of the fissioning compound is inverted. Flipping periodically the neutron spin the angular distributions of the α-particles will wobble back

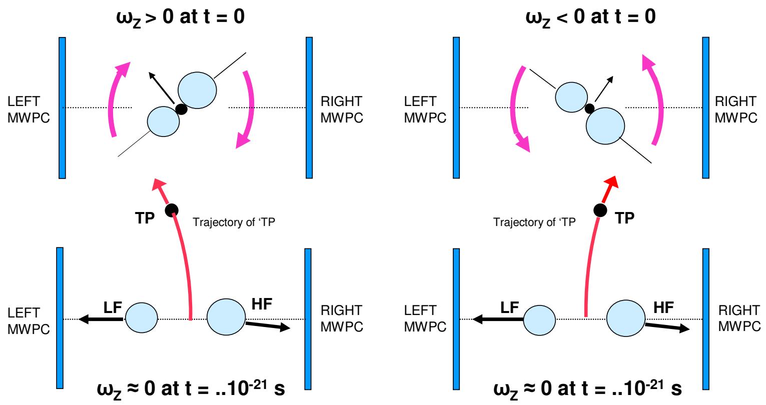

Fig. 6 – Model to explain how the rotation of the fission axis is measured. Left (right) panels are for rotations clockwise (anti-clockwise). Top panels sketch the configuration at scission ($t = 0$). Bottom panels represent the final situation some 10^{-21}s later with light (LF) and heavy (HF) fragments prone to be detected. Ternary particles (TP) escape with their angular distributions being shifted in opposite directions for the two senses of rotation. The Figure is not to scale.

and forth. This is illustrated in Fig. 6 for a neutron beam perpendicular to the drawing plane. Depending on the sense of rotation $\omega_z > 0$ or $\omega_z < 0$ at the time of scission $t = 0$ the trajectories are bent in opposite directions. Intercepting the α-particles to the right and left of the most probable angle, i.e. on the slopes of the angular distributions, the count rates measured will be sensitive to very small shifts in the distribution. When evaluating the asymmetry $A = (N^+ – N^-)/(N^+ + N^-)$, with N^+ and N^- the count rates for opposite senses of rotation, a characteristic pattern of signs for A is observed. On average the modulus of A equals $\mid A \mid = 3.30(13) \times 10^{-3}$. Albeit small, the effect having been dubbed the “ROT” effect is seen to be clearly manifested for the reaction investigated.

The properties of LCPs ejected in ternary fission from the neck between the separating fragments have since their discovery been thought to carry information on the dynamics of the scission configuration. However, due to the difficulties inherent in the analysis of three-body systems the results obtained have not always been unambiguous. In the ROT effect the LCPs serve to explore the collective rotation of the fissioning nucleus. The ROT effect appears to clearly demonstrate that ternary fission indeed is giving insight into the dynamics of the fission process.

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