Eur. Phys. J. A **24**, 379–388 (2005)
DOI 10.1140/epja/i2005-10026-2

THE EUROPEAN PHYSICAL JOURNAL A

New experimental studies on the quaternary fission of ^{233,235}U(n_{th}, f) and ^{252}Cf(sf)

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Received: 6 January 2005 / Revised version: 17 March 2005 /
Published online: 7 June 2005 –
Communicated by D. Schwalm

Abstract. Experiments have been performed for studying quaternary fission (QF) in spontaneous fission of ^{252}Cf, on the one hand, and for the neutron-induced fission reactions ^{233,235}U(n_{th}, f), on the other hand. In this higher-multiplicity fission mode, by definition, four charged products appear in the final state. In other words, as a generalization of the ternary-fission process, not only one but two light charged particles (LCPs) are accompanying the splitting of an actinide nucleus into the customary pair of fission fragments. In the two sets of measurements, which have used quite different approaches, the yields of several QF reactions with α-particles and tritons as the LCPs have been determined and the corresponding kinetic-energy distributions of the α-particles measured. The QF process can appear in two basically different ways: i) the simultaneous creation of two LCPs in the act of fission ("true" QF) and ii) via a fast sequential decay of a single but particle-unstable LCP in common ternary fission ("pseudo" QF). Experimentally the two varieties of QF have been distinguished by exploiting the different patterns of angular correlations between the two outgoing LCPs. The experiments described in the present paper are the first to demonstrate that both types of reactions, true and pseudo QF, occur with quite comparable probabilities. As a new result also, the kinetic-energy distributions related to the two processes have been shown to be significantly different. For all QF reactions which could be explored, the yields for ^{252}Cf(sf) were found to be roughly by an order of magnitude larger than the yields found in the ^{233}U(n_{th}, f) and ^{235}U(n_{th}, f) reactions. 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.

PACS. 25.85.Ec Neutron-induced fission – 25.85.Ca Spontaneous fission – 24.75.+i General properties of fission – 29.30.Ep Charged-particle spectroscopy

1 Introduction

Usually fission of actinide nuclei proceeds by decay into two fragments of comparable size. With probabilities at the level of 10^{-3}/fission, a third, in most cases light charged particle (LCP) accompanies the customary fission fragments. The process is called ternary fission (TF). In about 87% of TF events an energetic α-particle with a mean energy of about 16 MeV shows up. Helium isotopes (^{4,6,8}He) and hydrogen isotopes (^{1,2,3}H) contribute nearly 97% of the total TF yield, heavier LCP species being emitted very rarely [1,2]. There is ample evidence from experiment, and also from theory, that by far the majority of LCPs are born right at scission in the neck region of the nascent fission fragments. Thus, although being a rather rare process, the study of TF provides the experimentalist with one of the few means to explore the behaviour of the fissioning system near scission.

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.

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Probably due to the low yields, QF has been barely studied in the past. Only a few experiments are known from the literature both for thermal-neutron–induced fission of ^{235}U [3,4], and spontaneous fission of ^{252}Cf and ^{248}Cm [5, 6]. 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 [7]. 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 [8].

Quaternary fission is observed in experiment as a coincidence between two light charged particles (and two fission fragments) in separate detectors. Up to now only α-particles and H-isotopes could be identified in QF, *i.e.* precisely those light nuclei which are also the most abundant LCPs in TF. However, the timing capabilities of detectors do not allow to distinguish between the simultaneous creation of two LCPs in the act of fission ("true" QF) and the fast sequential decay of a single but particle-unstable LCP in common TF. Very similar to the neutron-unstable LCPs ^{5}He, ^{7}He and ^{8}Li*(2nd excited state) which have been traced recently [9] and which disintegrate into a charged particle and a neutron before reaching the detectors, there exist also particle-unstable LCPs decaying with short lifetimes into charged-particle pairs [10]. In the latter case a basically ternary event turns quaternary in a sequential process and is registered as such. This process has been called "pseudo" QF [11]. The most prominent example of pseudo QF is the formation of a primary ^{8}Be, either in its ground state or in excited states, and its subsequent decay into two secondary α's. Similarly, pseudo (α, t) QF has been detected which is attributed to the decay of ^{7}Li* when it is born in its 2nd excited state, as already conjectured by N. Feather in 1974 [12]. For ^{7}Li the ground and 1st excited state are particle-stable and cannot mimic QF.

The experiments to be reported in the following have made use of quite different approaches for studying QF in the spontaneous fission of ^{252}Cf, on the one hand, and in the neutron-induced fission reactions ^{233,235}U(n_{th}, f), on the other hand. In both sets of measurements the yields of several QF reactions and the kinetic-energy distributions of the corresponding LCPs have been obtained. Experimentally the two varieties of QF have been distinguished by exploiting the different patterns of angular correlations between the two outgoing LCPs.

2 Quaternary fission in ^{252}Cf(sf)

2.1 Experiment

Quaternary fission in the spontaneous decay of ^{252}Cf(sf) was studied with a detector assembly which is sketched in fig. 1. A ^{252}Cf source with a diameter of 3 mm and having a strength of about 5000 fissions/s was viewed by a set of eight ΔE-$E_{\rm rest}$ Si diode telescopes for detection and identification of LCPs. The active area per telescope was 1 cm^2 and all telescopes together subtended a solid angle of about 20%. The 12 μm thick ΔE detectors were made from CHICSI-type chips from SINTEF, Trondheim, Norway, and the 380 μm thick $E_{\rm rest}$ detectors from SFH 871 chips from SIEMENS, München, Germany. The ^{252}Cf source was covered on both sides with a 1 mg/cm^2 Ni foil. An additional 20 μm thick kapton foil of spherical shape was placed around the source for protecting the telescopes from being hit by fission fragments and the 30 times more frequent 6.1 MeV α-particles from the radioactive decay of ^{252}Cf. Fission fragments were hence not recorded. The spherical shape of the kapton foil was chosen in order to minimize the variation of the energy cut-off with detection angle. The whole setup was placed in a closed chamber filled with nitrogen gas of regulated pressure. The gas served as a further "variable thickness" absorber for fine adjustment of the discriminator thresholds for the detection of LCPs. The tuning of the threshold levels is crucial to set thresholds as low as possible but avoid interference between the LCPs and the α-particles from α-radioactivity in the ΔE detectors.

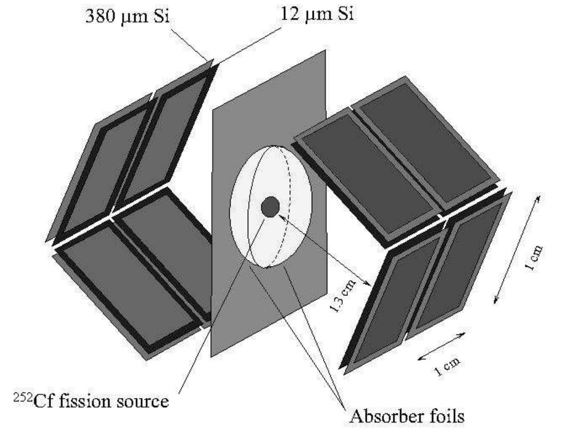

Fig. 1. Experimental setup for measuring (α-α) and (α-t) coincidences in ^{252}Cf(sf).

A sample ΔE-$E_{\rm rest}$ spectrum of LCPs measured with one of the eight telescopes is shown in fig. 2. The telescopes permit a clean separation of LCP nuclear charges. As regards the energy spectra, the lowest energy of α-particles to reach the $E_{\rm rest}$ detectors is 8 MeV. The ternary α spectrum used in the present work for normalisation includes the small contributions from ^{6}He and ^{8}He LCPs which were not fully separated from the α-particles ($\approx$ 3% and $\approx$ 0.2%, respectively), and also the contribution of 17% due to residual α-particles from the neutron decay of ternary ^{5}He, as recently confirmed in ref. [9]. As for the H-isotopes, triton events extracted from the ΔE-$E_{\rm rest}$ scatter plots (see fig. 2) are well separated from protons but contain a minor contamination due to deuterons. However, in previous work it was reported that in QF tritons, deuterons and protons come in the ratios 87% : 3% : 10% [5]. Compared to the abundances of H-isotopes from the ternary decay in ^{252}Cf(sf) with ratios t : d : p = 76% : 6% : 18% [13,14], it appears that the relative yields are quite similar in TF and QF. The contamination is, hence,

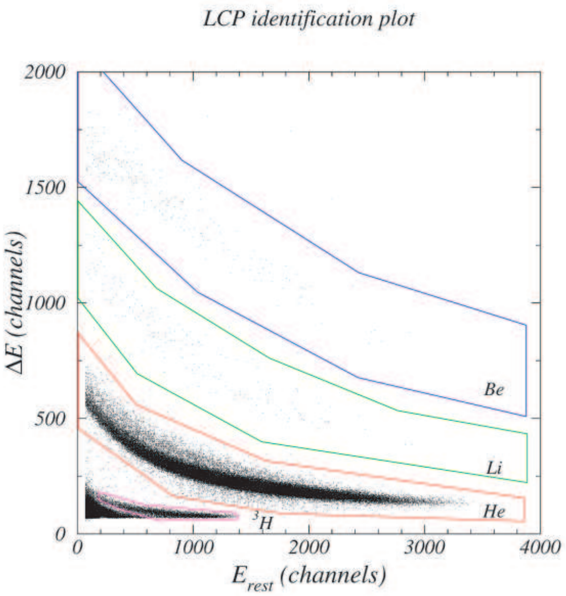

Fig. 2. ΔE-E_{rest} patterns from ternary LCPs in ^{252}Cf. The contour lines define the identification windows used for the analysis. The weak ($\approx$ 3%) patterns of ternary ^{6,8}He LCPs visible above the dominant α-particle component have not been treated separately (see text).

indeed negligible. In the triton spectra there is an upper limit of 11.5 MeV for full energy registration which is due to the thickness of the silicon detectors.

2.2 Evaluation

In four weeks of measuring time a total of 255 (α, α) and 37 (α, t) coincidences were registered within a narrow time window of 10 ns, random coincidences being negligibly rare.

The detector geometry chosen permits to cover a wide angular range between 16° and 180° for the mutual opening angles δ between two measured LCPs. The lower value of 16° comes from the detector frames. The 8 telescopes allow to assign 7 values for the mean opening angle between the LCPs from a pair. Angular distributions measured for (α, α) and (α, t) coincidences are on display in fig. 3. For convenience the double LCP events have been collected in the figure in a histogram, the histogram bars being placed at average angles corresponding to the opening angle between the centres of telescope surfaces as seen from the target, while the height of the bars give the number of counts observed. It should be noted that, first, the smallest average opening angle as just defined is about 50° and, second, the angular spread covered by any combination of detectors is considerably wider than the width of the histogram bars in the figure. The present data on angular distributions are in close agreement with the early work of Kataria *et al.* [5].

The distributions of relative angles between two α-particles for ^{252}Cf(sf) shown in fig. 3 point to an angular correlation which is independent of the angle for opening angles δ larger than some 60°. This feature is taken as the fingerprint for the independent emission of two α-particles and is, hence, identified as true quaternary fission. At smaller mutual angles there is a clear enhancement in the probability of emission of two α-particles which indicates the presence of a second component of QF. The pronounced correlation in angle is traced to the decay of ^{8}Be, which is particle unstable even in its ground state. An enhancement in the coincidence rate at smaller angles δ is observed in fig. 3 also for (α, t) pairs. Here it is the decay of ^{7}Li* from an excited state which introduces the close correlation in angles.

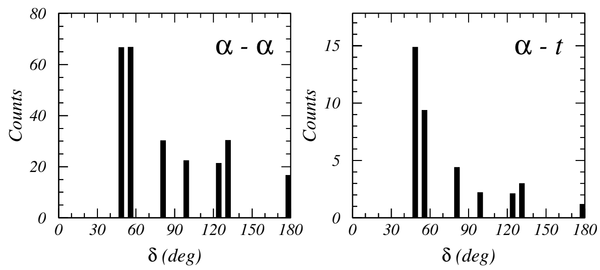

Fig. 3. Distribution of opening angles between two LCPs from quaternary fission of ^{252}Cf. Left: (α, α) coincidences, right: (α, t) coincidences. The angle δ denotes the angles as seen from the target centre to the centres of a pair of detectors registering two LCPs.

In the decay of ^{8}Be from the ground state, the comparatively long half-life ($T_{1/2} = 0.07$ fs) together with the low Q-value of the reaction ($Q = 0.092$ MeV) lead to a very narrow angular correlation between the two α's. Calculations simulating the kinematics of the decay have yielded a maximum opening angle δ_{max} of 8°, at the average kinetic energy of ^{8}Be. Since the smallest angle for detection of an α pair in neighbouring detectors is $\delta = 16°$, it is evident that in the Cf experiment only the decay from excited states of ^{8}Be can contribute to the observed enhancement at angles $\delta \leq 60°$. Thus, the measured pseudo (α, α) QF has to be attributed to the sequential decay of the short-lived 2^+ first excited state in ^{8}Be at $E^* = 3.04$ MeV ($T_{1/2} = 3 \cdot 10^{-22}$ s; $Q = 3.13$ MeV). In that case, simulation calculations have yielded a significantly broader distribution of opening angles which matches with high probability the angular range covered by the setup. A similar observation holds for the decay of ^{7}Li* from the 2nd excited state at $E^* = 4.63$ MeV ($T_{1/2} = 4.9 \cdot 10^{-21}$ s; $Q =$ 2.16 MeV). Contributions from higher-lying states than those quoted can be safely ruled out for ^{8}Be, while for ^{7}Li the 3rd excited state at $E^* = 6.68$ MeV may be responsible for an estimated 10% of the total (α, t) decay observed.

The kinetic energies of all LCPs were deduced from the measured E_{rest} pulse heights. They had to be corrected for energy losses in the absorbers in front of the telescopes and in the ΔE detectors. The α energy spectra from (α, α) coincidences for instantaneous and sequential QF are shown in fig. 4. The two variants of QF were disentangled by first gating on those events in fig. 3 (left pattern) with opening

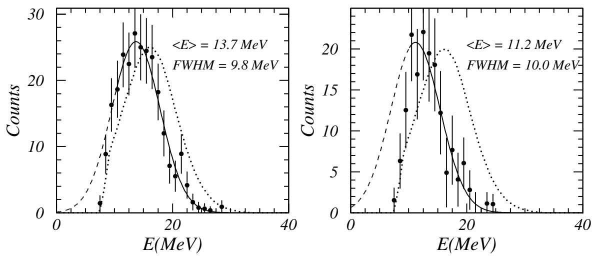

Fig. 4. Energy spectra of α-particles (corrected for energy losses) from true (α, α) (left panel) and pseudo (α, α) (right panel) quaternary fission of ^{252}Cf. Solid and dashed lines are Gaussian fits to the data. The ternary α spectrum for ^{252}Cf is given as the dotted line. Experimental spectra are truncated by the low-energy cut-off of the detectors at 8 MeV.

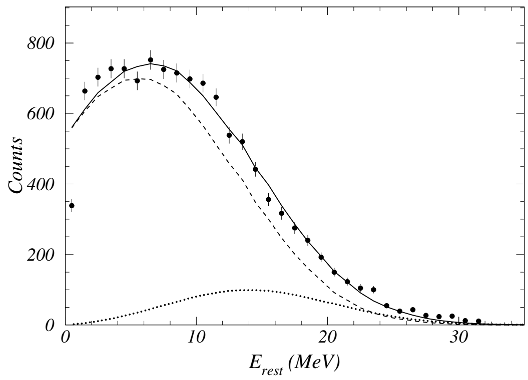

Fig. 5. Decomposition of the $E_{\rm rest}$ spectrum gated on the ΔE-E patterns for Li LCPs (data points). Dashed line: ternary Li spectrum. Dotted line: sum spectrum from the two α-particles from the decay of the ^{8}Be(g.s.) decay. Solid line: sum of both contributions. The cut-off energies are 17 MeV for Li, and 8 MeV for α-particles.

angles $\delta \geq 60°$ between the two α's to find the spectrum of both α-particles related with true QF. The spectrum is shown in the left pattern of fig. 4. The α spectrum from pseudo QF was deduced from coincidence events with opening angles $\delta \leq 60°$. Here, α-particles from true QF, taken to be isotropic and hence contributing equally to all angles, were subtracted after solid-angle normalisation. The resulting spectrum which corresponds to sequential QF with an intermediate ^{8}Be* is plotted in the right pattern of fig. 4. Both spectra have been fitted by Gaussians (solid and dashed curves). The spectrum of ternary α-particles from ^{252}Cf(sf) is given for comparison (dotted curve). As seen from the figure and the numerical values given in table 1, α-particles from (α,α) coincidences in QF have lower energies than those from ternary decays. This finding is in agreement with former data from refs. [4,5]. The α-particles assigned to pseudo (α, α) QF via the decay of ^{8}Be* (right figure) have even significantly lower energies than those from true (α, α) QF.

The yield of true (α, α) QF was determined for the full energy distribution, shown as a Gaussian in fig. 4, by extrapolating the angular distribution from data at opening angles $\delta \geq 60°$ to the full angular range. Furthermore, corrections from a Monte Carlo calculation were applied for the probability of double hits in the same telescope. Similar calculations were undertaken for the yield of producing ^{8}Be* in its 1st excited state. In the yield calculation it was assumed that the LCP pairs from both true QF and ^{8}Be* pseudo QF, are born right at scission in between the nascent fission fragments and are, as the LCPs in TF, subject to strong focussing in the fragment Coulomb field. Emission of the LCP pairs thus occurs at polar angles around 90° with respect to the fission axis. An analysis of the (α, t) coincidences (right panel of fig. 3) was made following the same lines. The resulting yields for both the true (α, t) QF and the sequential decay of ^{7}Li* in the 2nd excited state are included in table 1.

As explained, the yield of ^{8}Be formed in its ground state could not be observed in the angular correlation data and had, therefore, to be determined indirectly in the following way. The two α-particles from the ground-state decay of ^{8}Be hit with a probability close to 80% the same telescope. In 20% of all cases only one α-particle impinges on a detector and the second α is lost. As brought to evidence in the ΔE-$E_{\rm rest}$ scatter plot of fig. 2, the double α hits in a detector telescope should add up as an admixture to the pattern for the Li LCPs. In the evaluation an attempt was, hence, made to disentangle these double α events from Li LCPs. As on display in fig. 5, the measured $E_{\rm rest}$ spectrum in the evaluation window for Li was fitted with a superposition of the sum spectrum of two α-particles from ^{8}Be decay and the energy spectrum of Li-LCPs known from other experiments [14,15]. Both the two–α-particle sum spectrum and the Li spectrum were corrected for energy losses in absorbers and ΔE detectors. The mean energy of fully accelerated ^{8}Be was assumed to be 20 MeV before decay and, accordingly, a mean energy of 10 MeV was adopted for each of the two α-particles. The mean energy of 20 MeV for ^{8}Be is deduced by extrapolating known average energies for the heavier stable ternary Be-isotopes, the experimental data being corroborated by trajectory calculations [16]. The fractional yield for ^{8}Be(g.s.) found in this way is liable to large systematic errors which are estimated to be at least 50%. The main uncertainty is introduced by the energy loss corrections and the not precisely known ternary ^{8}Be and Li energies and spectral widths.

A summary of results on yields is given in table 1. The yield for true (α,α) QF, when calculated relative to binary fission, is $(1.0 \pm 0.3) \cdot 10^{-6}$, in fair agreement with $(1.5 \pm 0.5) \cdot 10^{-6}$ from the work of Kataria *et al.* [5], where an angular distribution similar to that shown in fig. 3 was measured but an admixture from ^{8}Be* was disregarded in the analysis. The present yields for true (α,α) QF and sequential ^{8}Be* decay sum up to $(1.7 \pm 0.6) \cdot 10^{-6}$, still in agreement with ref. [5]. On the other hand, the present result is larger by a factor of (3 ± 1) compared to the former work of Fomichev *et al.* [6].

The present analysis might be upgraded by performing trajectory calculations of the four-body kinematics. Hopefully these calculations should also give insight into

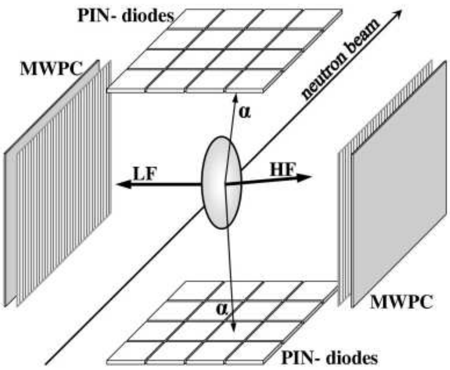

Fig. 6. Experimental setup for measuring of (α-α) and (α-t) coincidences in ^{233,235}U(n_{th}, f) installed at the high-intensity neutron beam PF1 at the ILL Grenoble, France.

the different starting conditions at scission which lead to the differences in the kinetic energies of α-particles in the ternary- and quaternary-fission modes. As regards a more direct and more accurate value for the fractional yield of ^{8}Be in its ground state a new experiment is planned. Besides a stronger Cf source, the method of pulse-shape discrimination in E_{rest} Si detectors made from raw material with resistivity profiles of better homogeneity than available for the present study will be applied [17]. The aim is to distinguish between signals from two α-particles hitting in coincidence the same telescope and signals from Li particles by exploiting their different rise times. This technique was successfully applied in the search of QF in U(n_{th}, f) reactions as demonstrated in the following sections.

3 Quaternary fission in ^{233,235}U(n_{th}, f)

3.1 Experiment

Studies of the fission reactions ^{233,235}U(n_{th}, f) were performed in a cold polarised neutron beam of the High Flux Reactor of the Institut Laue-Langevin in Grenoble. In parallel to the measurement of asymmetries and correlations of fission fragment and ternary-particle angular distributions relative to neutron spin [18,19], also quaternary fission was observed. While the neutron spin of the polarised neutron beam was flipped every second for these studies, quaternary data were taken irrespective of neutron spin orientation. The layout of the experiment is sketched in fig. 6. Under neutron bombardment the fissile U targets, either highly enriched ^{233}U or ^{235}U, emit fission fragments which are intercepted by multi-wire proportional counters labelled MWPC in the figure. For the detection of LCPs from TF and QF two arrays of silicon PIN diodes were placed at right angles to both the neutron beam and the fission axis. Since for the majority of LCPs the emission angles θ between fragments and LCPs are known to be very nearly perpendicular, this arrangement optimises the chance to observe TF and/or QF in coincidence between fragments and LCPs. Each of the two arrays consisted of 16 diodes, made of SFH 872 chips from SIEMENS, München, Germany. The 380 μm thick fully depleted detectors, $3 \times 3\,\text{cm}^2$ in size, were mounted in a quadratic frame with the n^+ rear side facing the source. Particle identification in the semiconductor detectors was based on the analysis of rise times of the current pulses. With injection of the ions from the rear side of the diodes the separation of α-particles from the hydrogen isotopes was perfect. The electronics, in particular the design of specially developed preamplifiers and timing filter amplifiers is described in ref. [17]. Due to the ranges of H-isotopes exceeding in many cases the thickness of the detectors, it was not possible to distinguish in all cases between tritons and deuterons. As already outlined in sect. 2.1, in the reaction ^{252}Cf(sf) it is known that the tritons carry almost 80% of the yield of H-isotopes, both in TF and QF. A similar predominance of tritons is also known from TF in the U reactions under study here. Alike for the Cf experiment, the detectors had to be shielded from fission fragments and α-particles from radioactive decay in order to avoid damage from too high ion doses. An aluminium foil used for this purpose necessarily entails a low-energy cut-off in the kinetic-energy spectrum of LCPs.

With a beam intensity of $6 \cdot 10^8$ neutrons/(s $\cdot$ cm^2) a binary fission rate of more than 10^6 fission events/s was achieved, while the diode arrays registered almost 10^3 ternary particles/s. Quaternary events are spotted as coincident signals in two PIN diodes. The installation of arrays on either side of the target together with their high granularity allows to study quaternary-fission events with small and large opening angles δ between the two LCPs. Small opening angles are obtained for two LCPs hitting the same array, while large opening angles correspond to hits in opposite arrays. It could be shown that a safe recognition of quaternary fission is feasible by inspecting merely the coincidences in the PIN diodes from the two arrays. Therefore, to further improve the statistics, in the evaluation of quaternary data all events with a 2-fold coincidence in the diode arrays were taken into account without insisting on a coincidence with fission fragments.

3.2 Evaluation

For U targets with some 5 mg of fissile material about $7.5 \cdot 10^3$ and $1.5 \cdot 10^4$ quaternary events from fission of ^{234}U* and ^{236}U*, respectively, were registered in 6 weeks of measuring time. Particle identification enabled to sort the data into 3 categories: (α, α), (α, H-isotope) and (H-isotope, H-isotope). By far, in most cases, two α-particles are observed in QF. On the other hand, the count rates in the third category were too low to warrant further evaluation. In the following only results from the first two categories are reported.

Evidence for QF is observed in the time spectrum of coincident events in two different LCP detectors. In fig. 7 the time distribution between successive hits in two PIN diodes of either the same array (left panel) or opposite arrays (right panel) is given for the reaction ^{235}U(n, f). The

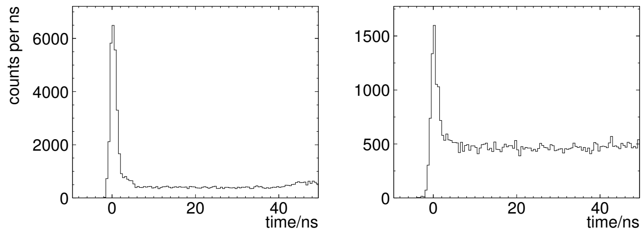

Fig. 7. Coincidence spectra of double hits. Left: same array. Right: opposite arrays.

peaks with a half-width of a few ns are the signature for QF. The half-width is larger than the electronic timing resolution due to both the different flight paths from the target to the individual diodes, and the different velocities of the LCPs. While the rates of background coincidences outside the peak region in the figure are very much the same for double hits in the same array or in opposite arrays, the number of true coincidences in the peaks is evidently much larger for events in one and the same array (left panel). This directly points to a strong component in QF where two LCPs are correlated with narrow opening angles δ between the two flight paths. These events are attributed to sequential QF with a ternary short-lived LCP in the intermediate state decaying well before reaching the detectors. As already outlined in the introduction, the LCPs in question are ^{8}Be and ^{8}Be* in the ground and 1st excited state contributing to (α, α) events, and ^{7}Li* in the 2nd state in the (α, H-isotope) category.

When analysing the distribution of opening angles δ between two coincident α-particles it is indeed found that at small angles δ the yields are enhanced. For the two U(n, f) reactions studied the distributions are very similar. The δ distribution for QF of ^{234}U* is displayed in fig. 8. The histogram bars indicate the opening angles δ between the midpoints of the diodes as seen from the centre of the target, and the heights of the bars correspond to the count rates observed per solid angle. Due to the setup of the detectors chosen (fig. 6) not all δ angles were accessible to experiment. The group at smaller angles $\delta \leq 90°$ corresponds to coincident events within the same detector array, while, for $\delta \geq 90°$, detectors in opposite arrays have registered such an event. The angular acceptance of each diode is around 8°, on the average, *i.e.* wider than the bars as shown in the figure.

The distributions of relative angles δ between the two α-particles shown in fig. 8 for ^{233}U(n, f) and those in fig. 3 for ^{252}Cf(sf) complement each other in pointing to enhanced yields for pseudo QF at small relative angles δ and an isotropic distribution for true QF at the larger angles. But, in contrast to the angular distribution measured for ^{252}Cf, the higher granularity of the detectors for U(n, f) permits to capture, with some probability, also pseudo QF via the decay of the ^{8}Be ground state. The yields for true QF enter as a correction to the yields of double events from the same diode array where both versions of QF contribute. Subtracting from the measured yields for $\delta \leq 90°$ the extrapolated yields from $\delta \geq 90°$ due to true QF, finally the yields for pseudo QF are obtained.

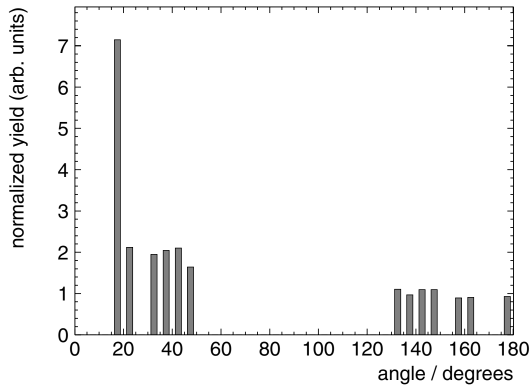

Fig. 8. Distribution of opening angles δ between α-particles in quaternary fission of ^{233}U(n_{th}, f).

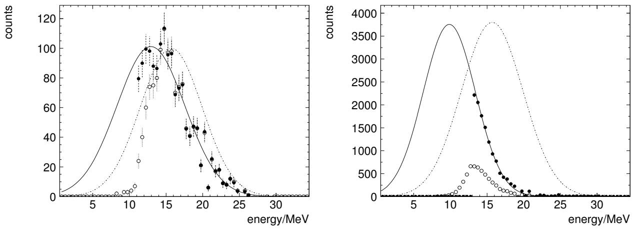

Fig. 9. Energy distributions of α-particles for the reaction ^{233}U(n_{th}, f) from true (left panel) and pseudo (right panel) quaternary (α, α) fission. Open circles: raw data. Full circles: data corrected for counting losses (see text). Continuous curve: Gaussian fit to the data. Dash-dotted line: energy spectrum of ternary α-particles.

Kinetic-energy distributions of α-particles from true and pseudo QF with two α-particles in the exit channel from ^{233}U(n_{th}, f) are on display in fig. 9 in the left and right panel, respectively. All data were corrected for energy losses in the target backings and the protecting Al foils in front of the diodes. The open points represent measured data, while for the full points corrections for counting losses were taken into account. In true QF the major losses arise due to the protecting Al foil in front of the PIN diodes which introduce a cut-off threshold in the energy spectrum. Since with the experimental setup of fig. 6 the LCPs enter the detectors under various angles, the energy threshold is not a simple step function. Instead, a correction factor depending on energy had to be calculated by a Monte Carlo simulation. For pseudo QF the corrections are more delicate. Additional substantial losses are here due to events from the decay of intermediate ternary LCPs with very narrow opening angles δ and where, therefore, an important fraction enters the same diode. Although this effect is less pronounced in the uranium data than for ^{252}Cf (see sect. 2.2), more than half of the events from pseudo (α, α) QF are lost that way and a correction for these "hidden" events had to be performed. The corrections were based on a Monte Carlo simulation of the patterns of distributions for the angle δ which traced the decay and its detection. For pseudo quaternary (α, α) fission, simulation calculations were pursued with summing up the decays from both the ^{8}Be ground state and ^{8}Be* 1st excited state. Thus, no effort has been made to separate the QF modes related to the decays of the two ^{8}Be states.

As seen in the right panel of fig. 9 the corrections for pseudo QF were pursued only for energies above 13 MeV, where threshold corrections no longer apply. The thin full lines in fig. 9 are Gaussians fitted to the corrected data. In the fitting procedure it was assumed that the energy distributions of the light particles extend to zero energy both in simultaneous and sequential quaternary decay. For ternary-fission spectra this is well known to be the case. For the present fits a yield of 2% of the maximum yield was imposed at zero LCP energy. Especially for sequential QF it is evident from the right panel in the figure that for the determination of yields a sizable uncertainty is introduced when the energy distribution has to be extrapolated to zero energy for taking into account all QF events. The resulting averages and variances of the distributions were calculated from the Gaussians. The results are summarised for all reactions measured in table 1. It is worthwhile to note that the data in fig. 9 exhibit a clear shift in the energies with α-particles from pseudo QF having lower energies than from true QF. Thus, the energy distributions of α-particles shown in fig. 9 for $^{233}U(n, f)$ and those in fig. 4 for $^{252}Cf(sf)$ closely complement each other.

Alike for ^{252}Cf, the yield of true (α, α) QF was determined for the full energy distribution, shown as a Gaussian in fig. 9, and by extrapolating the angular distribution shown in fig. 8 from the data at opening angles $\delta \geq 60°$ to the full angular range. Subtracting from the measured yields for $\delta \leq 90°$ the extrapolated yields from $\delta \geq 90°$ due to true QF, finally the yields for pseudo QF are obtained. A summary of results on yields is given in table 1.

For thermal-neutron–induced reactions only two experiments have become known in the literature which were restricted to the reaction $^{235}U(n_{th}, f)$. In the first experiment the yield of ^{8}Be was measured [3]. From 33 ^{8}Be events observed, the total yield for sequential quaternary fission was calculated to be 10^{-7} per fission. This figure is in agreement with $(1.4 \pm 0.4) \times 10^{-7}$ from the present work when normalising the measured QF yield to binary fission. Also the average energy of 20 MeV reported for ^{8}Be is in line with the average α energies of 9.9(10) MeV for each of the two decay α-particles as found here (see table 1). Unfortunately, however, the experimental setup used in ref. [3] did not allow to separately detect the simultaneous emission of two α-particles from true quaternary fission and, thus, a mixture of the two QF modes was observed. In this respect the second experiment was complementary since there only true quaternary fission could be spotted [4]. The yield for the sum of true $(\alpha + \alpha)$ and (α + H-isotope) quaternary decay was evaluated to be $1.0(1) \times 10^{-6}$ per fission, while from the present study a sum of yields of $0.6(2) \times 10^{-7}$ per fission emerges. These results are, hence, in conflict.

4 Results and discussion

The main parameters of the energy distributions of α-particles (mean value $\langle E_\alpha \rangle$ and FWHM) along with the yields for the different types of decay modes studied in the present work are summarised in table 1 for the three reactions, $^{233}U(n_{th}, f)$, $^{235}U(n_{th}, f)$ and $^{252}Cf(sf)$. The data for ternary fission accompanied by α-particles, ^{7}Li and ^{10}Be are included in the table for comparison. Attention is drawn to the fact that the yields are given relative to 10^4 ternary α-particles in each fissioning system. It should be stressed that for each of the reactions, the energy and yield data in table 1 have been obtained for the first time in one and the same experiment for the two competing modes of simultaneous and sequential decay.

As to the energy distribution of α-particles in table 1 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 α energies from ^{8}Be and $^{8}Be^*$ decay, their still lower energies are nevertheless in line with the average energy of about 20 MeV to be expected for a ^{8}Be(g.s.) particle prior to decay, as estimated from the systematics for the heavier $^{9,10,11,12}Be$ LCPs known for the reaction $^{235}U(n_{th}, f)$ [16, 1]. On average, therefore, each of the two α-particles should carry some 10 MeV of average kinetic energy.

Before starting the discussion of yields a particularity in the interpretation of the ^{8}Be yields has to be noted. Among the Be-isotopes also ^{9}Be is generated as a ternary particle. In its ground state ^{9}Be is stable. But ^{9}Be has several low-lying and short-lived excited states which may either decay by γ emission or neutron emission. In case of neutron decay of ^{9}Be the isotope ^{8}Be is formed. If now in TF ^{9}Be is not only produced in its ground state but also in excited states, there will be a certain side-feeding of ^{8}Be which in experiment cannot be disentangled from the "genuinely" ternary ^{8}Be. The amount of side-feeding is not readily assessed but could be quite sizable. For instance, assuming that at scission the nucleus has a temperature of 1 MeV, roughly half of the measured (^{8}Be + $^{8}Be^*$) yield could be due to side-feeding. This would lower the original ^{8}Be yield to about half of the measured ones. This source of systematic error is not considered in the yields given in table 1 which, in the following, will be taken at face value for discussion.

A salient feature in table 1 is the observation that, 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

Table 1. Spectral parameters and fractional yields of the different QF modes, in $^{233,235}U(n_{th}, f)$ and $^{252}Cf(sf)$. The yields for ^{10}Be and the ground state of ^{7}Li are average values from literature data. All yields are normalised to 10^4 ternary α-particles in each reaction.

		$^{233}U(n_{th}, f)$			$^{235}U(n_{th}, f)$			$^{252}Cf(sf)$		
Decay mode		$\langle E_\alpha \rangle$ (MeV)	FWHM (MeV)	Yield per 10^4 TF α's	$\langle E_\alpha \rangle$ (MeV)	FWHM (MeV)	Yield per 10^4 TF α's	$\langle E_\alpha \rangle$ (MeV)	FWHM (MeV)	Yield per 10^4 TF α's
α	TF	15.7(6)	9.8(6)	10^4	15.5(6)	9.8(6)	10^4	15.9(6)	11.3(6)	10^4
(α, α)	QF	12.9(8)	10.9(8)	0.41(13)	13.0(8)	10.9(8)	0.32(10)	13.7(8)	9.8(6)	3(1)
^{8}Be(g.s.)	QF							$10^{(a)}$	$10^{(a)}$	10(6)
$^{8}Be^*$	QF							11.2(8)	10.0(8)	2(1)
$\Sigma^{8}Be, ^{8}Be^*$	QF	9.0(10)	8.9(10)	0.94(30)	9.9(10)	8.3(10)	0.83(30)			
^{10}Be	TF			43(3)			30(2)			$140(15)^{(b)}$
(α, t)	QF	12(2)	9(1)	0.03(1)	12(2)	9(1)	0.03(1)			0.4(1)
$^{7}Li^*$	QF	11(2)	9(1)	0.03(1)	11(2)	9(1)	0.04(1)			0.3(1)
^{7}Li	TF			4.4(7)			4.1(3)			17(4)

(a) Fixed in the fitting routine (see text).
(b) Calculated as 80% of the measured total Be yield.

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. [20]) 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. Let us consider the ratio of yields for ternary ^{10}Be and α-particle as an example. From table 1 it is read that these ratios relative to 10^4 α's are 140 : 43 : 30, for ^{252}Cf : $^{234}U^*$: $^{236}U^*$. Scaled to yields per fission one finds ^{10}Be yields of 460 : 90 : 51. Since for ^{252}Cf no isotopic Be yields are available from experiment, the yield for ^{10}Be was estimated as $\approx 80\%$ of the measured yield for all beryllium LCPs [15,21]. An enhanced probability of heavier actinides for emitting a larger number of nucleons as light particles is, hence, a feature which is shared by ternary and quaternary fission. 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 true QF an interesting point is to confront the instantaneous (α, α) yield to the square of the α yield from TF. The idea behind is that possibly the two α's from the (α, α) decay in QF are simply due to the independent emission of α-particles from both the light and the heavy fragment. For $^{252}Cf(sf)$ the ratio of the (α, α) QF yield to the square of the α yield in TF is about 1 : 10, while for the two $U(n_{th}, f)$ reactions this ratio is about 1 : 100. Especially the result for the U reactions clearly demonstrates that quaternary fission is a new and independent phenomenon not linked to a multiple ternary process.

For pseudo QF mediated by ^{8}Be it is apparent from table 1 that the ratio of ternary yields $Y(^{10}Be)/Y(^{8}Be)$ varies smoothly for the three reactions under study. For $^{252}Cf(sf)$, $^{233}U(n_{th}, f)$ and $^{235}U(n_{th}, f)$ one finds values for the ratio $Y(^{10}Be)/Y(^{8}Be)$ of about 12, 46 and 36, 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 [22]. 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.

It should further be tempting to exploit from the data for $^{252}Cf(sf)$ in table 1 the ratios of yields for ^{8}Be in its ground state and its first excited state at 3.04 MeV. From the Boltzmann factor for this ratio a temperature could be deduced [23]. However, as already explained, the possibly large but not well-known amount of side-feeding by $^{9}Be^*$ in excited states contributing to the production of ^{8}Be is unfortunately spoiling this thermometer of the nucleus at scission.

Addressing quaternary fission with two main fragments, an α-particle and a triton, the first remark to be made when inspecting table 1 is that the yields for true (α, t) QF are down by roughly an order of magnitude compared to the (α, α) case. Stated otherwise, the ratio of QF yields is $Y(\alpha, t)/Y(\alpha, \alpha) \approx 10\%$. In ternary fission the ratio of triton to α yields, *e.g.* for $^{233}U(n_{th}, f)$, is $Y(t)/Y(\alpha) \approx 5\%$ and, hence, the two ratios from QF and TF are very close to each other. But in contrast to the (α, α) case, where true (α, α) QF and ^{8}Be-accompanied TF have probabilities quite close to each other, in the (α, t) case the probabilities for (α, t) QF and ^{7}Li-accompanied TF are largely different. The yield ratios $Y(\alpha, t)/Y(^{7}Li)$ are found in table 1 to be 2.3% and 0.7% for the reactions $^{252}Cf(sf)$ and $^{233,235}U(n_{th}, f)$, respectively. If it is allowed to apply an energy argument in case the nucleons involved are identical to estimate the ratio of reaction probabilities, then the differences in energy for $(\alpha,\ \alpha) \leftrightarrow {}^8Be$ on the one hand, and $(\alpha,\ t) \leftrightarrow {}^7Li$ on the other hand, should play a decisive role. In the $(\alpha,\ \alpha)$ case one has to point to the near degeneracy of the (α, α) pair and ^8Be, where the energy gain in the decay of $^8Be \rightarrow (\alpha,\ \alpha)$ is only 92 keV. By contrast, in the $(\alpha,\ t)$ case the rest energy of ^7Li is by 2.47 MeV lower than for the $(\alpha,\ t)$ pair. Hence, energy considerations could explain both, the near equality of $(\alpha,\ \alpha)$ and ^8Be yields in QF and TF, respectively, and the markedly favoured yield of ^7Li in TF compared to the $(\alpha,\ t)$ yield in QF.

Finally, very useful quantities carrying interesting information prove to be the yields measured for $^7Li^*$. In its second excited state at an excitation energy $E^* = 4.63$ MeV, the nucleus decays with a half-life of $4.9 \cdot 10^{-21}$ s to $(\alpha + t)$ and becomes accessible to observation as sequential QF. Contrary to ^8Be (see discussion above) there is no reaction in ternary fission which by side-feeding could contribute substantially to the yields of both ^7Li in its ground state or excited state. Also contributions from decays into $(\alpha,\ p)$ or $(\alpha,\ d)$ which could not be perfectly distinguished from $(\alpha,\ t)$ should be negligible. Therefore, the yields of ^7Li and $^7Li^*$ are "clean" and, hence, ideally suited for applying the Boltzmann law to find nuclear temperatures at scission [24]. The only drawback are the small yields for $^7Li^*$ making measurements difficult and introducing large error bars. Anyhow, taking into account the proper spin factors for the two ^7Li states in question, one calculates from the yields given in table 1 the temperatures $T = 0.98(12)$ MeV for $^{252}Cf(sf)$, $T = 0.80(9)$ MeV for $^{233}U(n_{th}, f)$ and $T = 0.87(6)$ MeV for $^{235}U(n_{th}, f)$. In these calculations the contribution by the third excited state of ^7Li at $E^* = 6.68$ MeV has been neglected because at the temperatures indicated its contribution is less than 10%. Temperatures around 1 MeV for $^{252}Cf(sf)$ have also been found from the yields of $^{10}Be^*$ in its first excited state at $E^* = 3.37$ MeV and the ground-state yield of ^{10}Be [14,21]. The excited state of ^{10}Be decays to the ground state by γ emission.

This work was supported in parts by INTAS (call 99-229) and the German Minister for Education and Research (BMBF) under contracts 06DA913 and 06TU669.

References

1. M. Mutterer, J.P. Theobald, in *Nuclear Decay Modes*, edited by D.N. Poenaru (IOP Publ., Bristol, England, 1996) Chapt. 12.
2. C. Wagemans, in *The Nuclear Fission Process*, edited by C. Wagemans (CRC Press, Boca Raton, FL, USA, 1991) Chapt. 12.
3. V.N. Andreev, V.G. Nedopekin, V.I. Rogov, Yad. Fiz. **8**, 38 (1969)), (Sov. J. Nucl. Phys. **8**, 22 (1969)).
4. S.S. Kapoor, S.K. Choudhury, S.K. Kataria, S.R.S. Murthy, V.S. Ramamurthy, *Proceedings of the Seventeenth Nuclear Physics and Solid State Physics Symposium, Chandigarh, India, 1972-73*, Vol. **15B** (Department of Atomic Energy, Bombay, 1973) p. 107.
5. S.K. Kataria, E. Nardi, S.G. Thompson, *Proceedings of the Nuclear Physics and Chemistry of Fission, Rochester 1973*, Vol. **II** (IAEA, Vienna, 1973) p. 389.
6. A.S. Fomichev, I. David, M.P. Ivanov, Yu.G. Sobolev, Nucl. Instrum. Methods Phys. Res. A **384**, 519 (1997).
7. N. Carjan, A.J. Sierk, J.R. Nix, Nucl. Phys. A **452**, 381 (1986).
8. D.N. Poenaru, W. Greiner, J.H. Hamilton, A.V. Ramayya, E. Hourany, R.A. Gherghescu, Phys. Rev. C **59**, 3457 (1999).
9. Yu.N. Kopatch, M. Mutterer, D. Schwalm, P. Thirolf, F. Gönnenwein, Phys. Rev. C **65**, 044614 (2002).
10. M. Mutterer, Yu.N. Kopatch, P. Jesinger, F. Gönnenwein, *Proceedings of the International Conference on Dynamical Aspects of Nuclear Fission, DANF01, Častá Papiernička, Slovakia* (World Scientific, Singapore, 2002) p. 326.
11. F. Gönnenwein, P. Jesinger, M. Mutterer, A.M. Gagarski, G.A. Petrov, W.H. Trzaska, V. Nesvishevsky, O. Zimmer, *Proceedings of the International Workshop on Fission Dynamics of Atomic Clusters and Nuclei, Luso, Portugal, May 15-20, 2000*, edited by J. de Providencia, (World Scientific, Singapore, 2001) p. 232.
12. N. Feather, Proc. R. Soc. Edinburgh, **71**, 21 (1974).
13. C. Wagemans, in *Particle Emission from Nuclei*, edited by D.N. Poenaru, M.S. Ivascu, Vol. **III** (CRC Press, Boca Raton, FL, USA, 1988) Chapt. 3.
14. P. Singer, Yu.N. Kopatch, M. Mutterer, M. Klemens, A. Hotzel, D. Schwalm, P. Thirolf, M. Hesse, *Proceedings of the International Conference on Dynamical Aspects of Nuclear Fission, DANF96, Častá Papiernička, Slovakia*, edited by J. Kliman, B.I. Pustylnik (JINR, Dubna, 1996) p. 262; P. Singer, PhD Thesis, Technische Universität Darmstadt (1996).
15. Z. Dlouhy, J. Svanda, R. Bayer, I. Wilhelm, *Proceedings of the International Conference on Nuclei Far from Stability and 9th International Conference on Atomic Masses and Fundamental Constants, Bernkastel-Kues, Germany, 1992*, edited by R. Neugard, A. Wöhr, Inst. Phys. Conf. Ser. **132**, 481 (1993).
16. W. Baum, PhD Thesis, Technische Universität Darmstadt (1992).
17. M. Mutterer, W.H. Trzaska, G.P. Tyurin, A.V. Evsenin, J. von Kalben, J. Kemmer, M. Kapusta, V.G. Lyapin, S.V. Khlebnikov, IEEE Trans. Nucl. Sci. **47**, 756 (2000).
18. P. Jesinger, A. Kötzle, A.M. Gagarski, F. Gönnenwein, G. Danylian, V.S. Pavlov, V.B. Chvatchkin, M. Mutterer, S.R. Neumaier, G.A. Petrov, V.I. Petrova, V. Nesvizhevsky, O. Zimmer, P. Geltenbort, K. Schmidt, K. Korobkina, Nucl. Instrum. Methods Phys. Res. A **440**, 618 (2000).
19. P. Jesinger, PhD Thesis, Universität Tübingen (2001).
20. U. Köster, PhD Thesis, Technische Universität, München (2000).
21. A.V. Daniel, G.M. Ter-Akopian, J.H. Hamilton, A.V. Ramayya, J. Kormicki, G.S. Popeko, A.S. Fomichev, A.M. Rodin, Yu.Ts. Oganessian, J.D. Cole, J.K. Hwang, Y.X. Luo, D. Fong, P. Gore, M. Jandel, J. Kliman, L. Krupa, J.O. Rasmussen, S.C. Wu, I.Y. Lee, M.A. Stoyer, R. Donangelo, W. Greiner, Phys. Rev. C **69**, 041305(R) (2004).

22. D.N. Poenaru, W. Greiner, R.A. Gherghescu, At. Data Nucl. Data Tables **68**, 91 (1998).
23. G.V. Val'skiĭ, Yad. Fiz. **24**, 270 (1976), (Sov. J. Nucl. Phys. **24**, 140 (1976)).
24. F. Gönnenwein, P. Jesinger, M. Mutterer, W.H. Trzaska, G.A. Petrov, A.M. Gagarski, V. Nesvizhevsky, P. Geltenbort, *Proceedings of the Symposium on Nuclear Clusters: from Light Exotic to Superheavy Nuclei, Rauischholzhausen, Germany, 2002*, edited by R. Jolos, W. Scheid (EP Systema Bt., Debrecen, Hungary, 2002) p. 293; Acta Phys. Hung. New Ser. Heavy Ion Phys. **18**, 419 (2003).