RECENT MULTI-PARAMETER STUDIES ON PARTICLE-ACCOMPANIED FISSION OF ^{252}Cf(sf) AND ^{235}U(n_{th},f)

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Abstract. Recent multi-parameter experiments were performed on particle-accompanied fission in ^{252}Cf(sf) and ^{235}U(n_{th},f), respectively. With a dedicated detection system CODIS2, angular and energy correlations between fission fragments (FF) and the light charged particles (LCP) were measured . The FFs were detected by an energy and angle sensitive 4π twin ionization chamber while the LCPs were intercepted by a number of 24 ΔE-E_{rest} telescopes. LCP isotopes could be disentangled up to Be. In addition, in the ^{252}Cf(sf) experiment, γ-rays emitted in the reaction were detected by a pair of large-volume segmented clover Ge detectors. Here, the main interest is to study, on the one hand, the γ-decay from FFs in ternary fission including multiplicity and anisotropy of the γ-rays, and, on the other hand, the population of excited states in the LCPs by their characteristic decay. The study of FF-LCP correlations was also the main purpose of the ^{235}U(n_{th},f) experiment performed at the high-flux reactor of the ILL, Grenoble, using an intense cold neutron beam of $3 \cdot 10^9$ neutrons/(cm^2 $\cdot$ s). Here, a counting rate as high as $2.5 \cdot 10^5$ fissions/s could be registered with CODIS2 without compromising its high-resolution capability. For the first time, FF-LCP correlations were measured also for very rare quaternary fission events which occur with probabilities down to 10^{-7} / fission. At the current status of data evaluation, yields and energy distributions of light particles have been evaluated. For the present contribution, in particular, our still preliminary results on the yields of ternary Li isotopes for the two reactions studied are compared and discussed. For ^{252}Cf(sf) the isotopic yields of the elements heavier than He were hitherto rarely known. For ^{235}U(n_{th},f) the present results and earlier data measured at the recoil mass spectrometer LOHENGRIN at the ILL nicely complement one another with respect to the energy coverage.

Keywords: Ternary Fission, Quaternary Fission, Yields of Light Charged Particles in Fission
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Introduction

Studies on particle accompanied fission have already a long history [1,2]. Spontaneous and thermal-neutron induced fission of actinides has been extensively investigated searching for the decay of a nucleus into two fission fragments (FF) and, additionally, one light charged particle (LCP), the so-called ternary fission (TF) process, or two light particles, so-called quaternary fission (QF). There is strong evidence from experiment, and also from theory, that the LCPs are born together with the main fragments when the nucleus breaks apart at scission. Being evidently formed in the neck region between the nascent fragments, the LCP energy and emission angles develop from acceleration in the strong time-dependent fragment Coulomb field at and shortly after scission. The study of FF–LCP correlations in particle accompanied fission, though being a quite rare process, may thus provide a unique tool for exploring nuclear fission dynamics near the instant of rupture. The TF probabilities never exceed a few tenths of a percent in low-energy fission, and QF yields are even down to the order of 10^{-6} to 10^{-7} /fission.

In the present paper, we are going to describe two recent experimental studies in a series of FF–LCP correlation experiments that either include registration of neutrons and/or γ-rays with LCPs and FFs [3-5], or the coincident registration of two LCPs [6]. In a first experiment on ^{252}Cf(sf), besides the pair of FFs and a further LCP, the gammas emitted in ternary fission have been intercepted with high-efficiency Ge detectors [7]. Of interest is here the angular anisotropy of γ-rays relative to the fission axis for both, binary and ternary fission. The comparison of anisotropy in the two processes should give some insight whether the distributions of fragment angular momentum are disturbed by the emission of LCPs. But also the LCPs themselves may be produced in an excited state and emit γ-rays. The relative probability for LCPs to be born either in the ground or excited state may give a clue to the excitation mechanism or nuclear temperature at scission. The study of FF-LCP correlations was also the main purpose of the ^{235}U(n_{th},f) experiment performed at the high-flux reactor of the ILL, Grenoble, using an intense cold neutron beam of $3 \cdot 10^9$ neutrons/(cm^2 · s) [9]. Here, the high rate of $2.5 \cdot 10^5$ fissions/s permitted to register, for the first time, FF-LCP correlations also for very rare quaternary fission events.

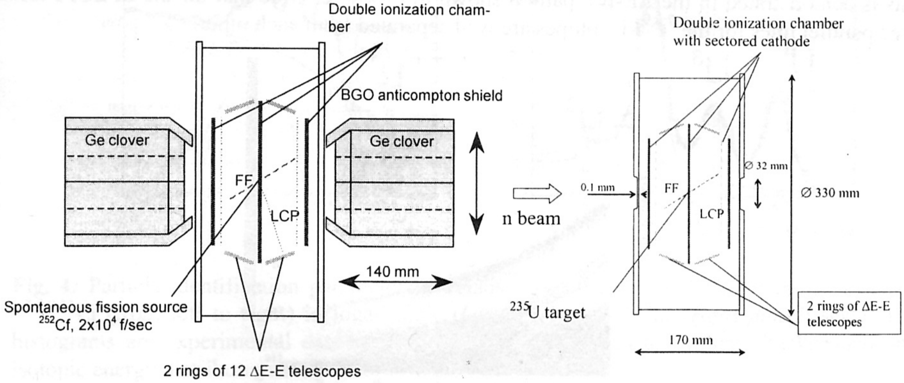

Fig. 1: ***Left panel:*** Detector arrangement for the study of ternary fission in the spontaneous decay of ^{252}Cf(sf). The central part is the FF and LCP detector system CODIS2. The two segmented Super Glover Ge detectors on both sides of CODIS2 are equipped with BGO anticompton shields. ***Right panel:*** Detector system CODIS2 adapted for the measurement of ^{236}U* ternary fission following capture of cold neutrons.

Layout of Experiments

A schematic drawing of the detector setup for the ^{252}Cf(sf) experiment is given in the left part of Fig. 1. A californium source with $2.5 \cdot 10^4$ fissions/s on a thin backing is placed at the center of the cathode of a twin back-to-back ionization chamber, operated with pure CH_4 at a pressure of 760 mb. As a specific feature of this chamber called CODIS2 the cathode is segmented into 8 sectors which permits to not only measure the energies of both FFs but also their polar and azimuthal angles of emission relative to the chamber axis. For identifying the LCPs two rings with 12 ΔE-E_{res} telescopes each are installed in the chamber. They are designed from ΔE ionization chambers and E_{res} silicon detectors for identifying the LCPs emitted in coincidence with the FFs and determining their energies and emission angles. Further, for detecting the γ-rays emitted by FFs and LCPs two segmented large-volume Super-Clover Ge-detectors were aligned on the chamber axis in close geometry. Each detector has 4 large Ge crystals, 14 cm in length and 6 cm in diameter. More details are to be found in [7].

The detector assembly for the experiment on ternary fission of $^{236}U^*$ following capture of cold neutrons is shown on the right in Fig. 1. A thin (50 μg/cm²) and highly enriched ^{235}U target was placed in a cold neutron flux of $3 \cdot 10^9$ neutrons /(cm²·s). The same double ionization chamber CODIS2 as in the experiment with ^{252}Cf was used but modifications had to be made to adapt it to the high neutron flux. The gas was CF_4 at 300 mb pressure, the anodes plates were replaced by grids, and the neutron entrance and exit windows were made of 0.1 mm thick Al foils. In actual practice a count rate capability of $2.5 \cdot 10^5$ / s was achieved for binary fission without compromising the overall performance. The count rate for ternary fission was 70 / s and for quaternary fission $7 \cdot 10^{-4}$ / s. More details are given in [8].

Data Analysis

At this time, final results on the main goals of the two complex experiments, i.e. the LCP-FF correlations and related topics, are not readily evaluated because of the quantity and complexity of the data registered. Partial results are however available on LCP yields and energy distributions. In the ^{252}Cf experiment data for LCPs ranging from helium up to carbon could be taken, and for helium up to beryllium even the individual isotopes were resolved. This is demonstrated in the ΔE-E_{res} pattern shown in of Fig. 2. Note that for the Li LCPs the three parallel lines for the $^{7,8,9}Li$ isotopes are well separated from each other.

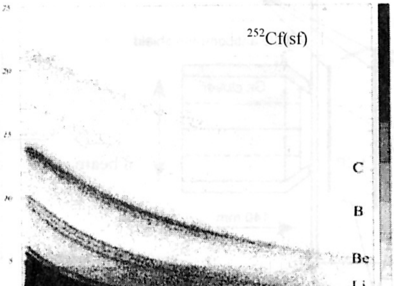

Fig. 2: Sample plot of ΔE-E_{res} patterns for LCPs from He to C (from bottom to top), measured with the LCP telescopes in CODIS2.

For deducing the isotopic energy distributions and yields the measured ΔE-E_{res} patterns were converted to the parameter $PI = 5Z + (A - 2Z)/2$ in an analogous manner as described in [9], Particle identification parameters PI versus residual energies E_{res}, for LCPs from hydrogen to beryllium are shown in Fig. 3, together with energy integrated PI distributions. Here, for He and Li the deconvolution into isotopes is indicated. The background seen between the curves for He and Li isotopes is caused by pile-up due to some interference between the small telescope ΔE signals and the more than four orders of magnitude more frequent 6.1 MeV α particles from ^{252}Cf radioactivity stopped just in front of the LCP telescopes.. Finally, Fig. 4 depicts the group of Li isotopes, with the deconvolution into isotopic contributions indicated.

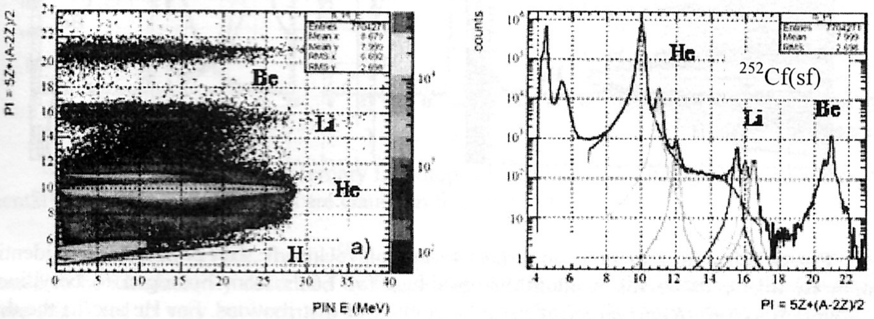

Fig. 3: Separation of LCPs in ternary fission of ^{252}Cf(sf): ***Left panel:*** Particle identification parameter PI versus residual energies E_{res}, for LCPs from hydrogen to beryllium (from bottom to top). ***Right panel:***. Energy integrated PI distributions. For He and Li the deconvolution into isotopes is indicated.

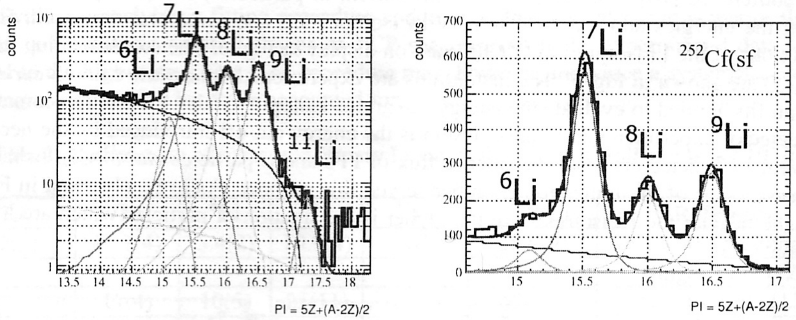

Fig. 4: Particle identification parameter PI versus residual energies E_{res}, for the Li isotopes ^{6,7,8,9,11}Li (from left to right) in logarithmic (***left panel***) and linear (***right panel***) scale. The histograms are experimental data while lines are fitted curves to evaluate background and isotopic energy distributions.

In the experiment on ^{235}U(n_{th},f) data for LCPs ranging from helium up to beryllium could be taken. Cut-off energies are somewhat higher compared to ^{252}Cf(sf) since the energy loss in the chamber gas in CODIS2 had to be slightly increased to safely stop fission fragments from the light mass group in the FF ionization chamber in front of the Frisch grid. The ΔE-resolution for LCP identification (see Fig. 5) was inferior compared to the ^{252}Cf(sf) experiment (Fig. 3) due to not fully solved problems with the interference between ground loop noise and the signals in the very sensitive ΔE electronic channels. This leaves some limitation on the quality of deconvolution into isotopes, as is seen from the plots of the particle identification parameter PI versus residual energies E_{res} shown in Fig. 5.

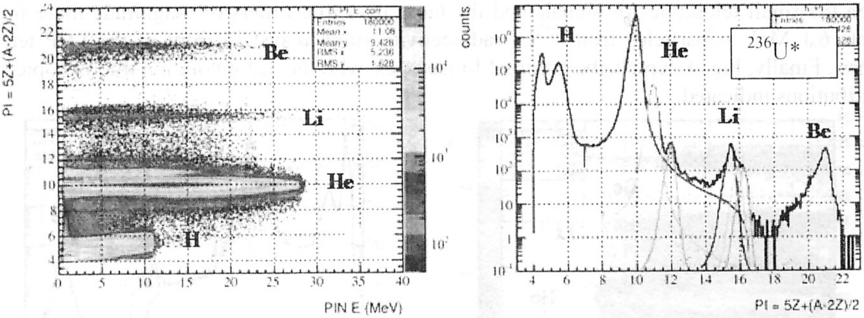

Fig. 5: Separation of LCPs in ternary fission of $^{235}U(n_{th},f)$: ***Left panel:*** Particle identification parameter PI versus residual energies E_{res}, for LCPs from hydrogen to beryllium (from bottom to top). ***Right panel:***. Energy integrated PI distributions. For He and Li the deconvolution into isotopes is indicated.

First Results

As a partial result for the two reactions studied the LCP yields and energy distributions have been evaluated. The yields of Be isotopes have already been presented at another recent conference [10]. For the present contribution, in particular, our (still preliminary) results on the energies and yields of ternary Li isotopes are compared and discussed. Energy distributions of the Li isotopes in $^{252}Cf(sf)$ are on display in Fig. 6. The corresponding data on $^{235}U(n_{th},f)$ are shown in Fig. 7. The histograms are experimental data while smooth curves are Gaussian fits applied to evaluate the energy-integrated isotopic yields. A difficulty common to many detector experiments in ternary fission is the energy cut-off introduced by the necessity to prevent LCP detectors from the intense flux of FFs and α-particles (for ^{252}Cf). In the present case the gas of the ionization chamber served this purpose. As to be observed in Figs. 6 and 7, the heavier the Li isotopes are the higher is the fraction of particles which are lost for detection.

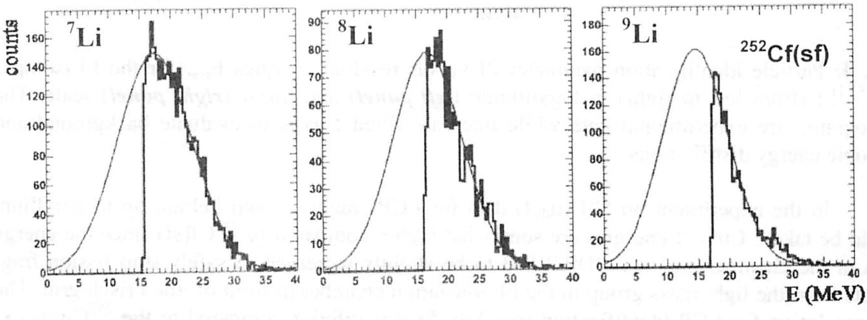

Fig. 6: Energy spectra of ternary Li isotopes $^{7,8,9}Li$ in $^{252}Cf(sf)$. Histograms are experimental data while smooth curves are Gaussian fits.

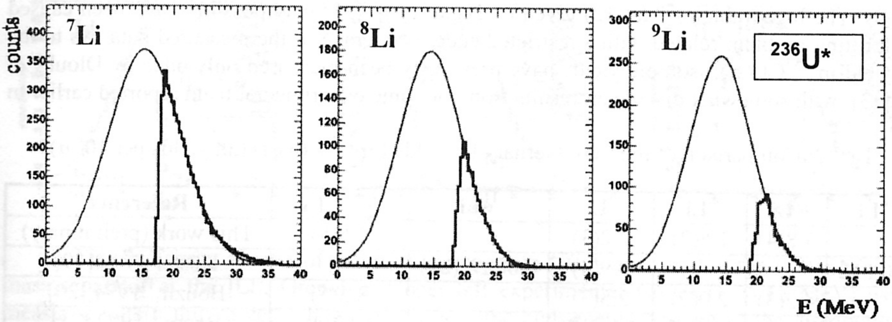

Fig. 7: Energy spectra of ternary Li isotopes ^{7,8,9}Li in ^{236}U*. Histograms are experimental data while smooth curves are Gaussian fits.

As discussed in [10], for the evaluation of LCP yields all energy distributions were assumed to be satisfactorily described by Gaussians. Additionally, the widths of the Gaussians were stipulated to vary between the limits $0.6 \leq \Delta E$ (fwhm) ≤ 1.0, in units of the respective mean energy <E>. This presumption has been shown by Gönnenwein et al. [11] to be well met by experimental LCP energies in many thermal neutron induced reactions, for LCPs up to mass number $A \approx 25$. The condition applied leaves some small yield close to zero energy, as is seen from Figs. 6 and 7. In practice, it is compatible with artificially requiring the fitted Gaussians to carry (1-2) % of the maximum yield in the energy bin including zero energy [10]. Evidently applying a fitting procedure conditioned that way at the low-energy side results in quite realistic spectral shapes of LCP spectra even if the experimental data are restricted to the high-energy side. The procedure may, however, introduce a systematic error on the integral yield which has to be accounted for.

Tab. 1: Comparison of measured ternary Li yields for ^{252}Cf (sf) (all yields per 10^4 α)

^{6}Li	^{7}Li	^{8}Li	^{9}Li	^{11}Li	$\sum^{6-11}$ Li	**Reference**
1.2(3)	11.1(4)	5.4(2)	10.9(4)	≤0.3	28.6(9)	This work (preliminary)
					52(5)	Singer 1997 [12]
	17(4)	10(5)	25(11)		52(13)	Dlouhý 1992 [13]
					55(4)	Grachëv 1988 [14]
	12.7(21)	8.5(14)				Bayer 1981 [15]
					29(5)	Gazit 1970 [16]
					13.2(16)	Cosper 1967 [17]
					12(6)	Whetstone 1967 [18]

In Table 1 the Li yields for ^{252}Cf(sf) are compared with literature data, while Table 2 contains measured yields for ^{236}U* from different experiments All yields are normalized to 10^4 ternary α-particles from the respective reaction. Missing entries in the table indicate that the corresponding yields were not measured or, as in the case for ^{6}Li and ^{11}Li from the present experiment on ^{235}U(n,f), are not yet evaluated.

The agreement between the results reported by different groups for the ^{7,8,9}Li yields from fission of ^{236}U* has to be considered acceptable in view of the difficulties of the experiments. The high value of 3.54(12) given by Baum et al. [21] may possibly have been caused by a fitting problem related with a restricted energy coverage of the measured data. As to the Li yield in ^{252}Cf (sf), isotopic yields have previously been measured only once by Dlouhý et al. [13], with somewhat discrepant results from the same experimental team reported earlier in [15].

Tab. 2: Comparison of measured ternary Li yields for ^{235}U(n_{th},f) (all yields per 10^4 α)

^{6}Li	^{7}Li	^{8}Li	^{9}Li	^{11}Li	$\sum^{6\text{-}11}$ Li	Reference
	3.8(4)	1.6(2)	2.2(3)		7.6(8)	This work (preliminary)
≤0.012			2.9(3)	0.00090(22)		Köster 1999 [19]
	4.2(3)	2.0(2)	2.3(3)		8.5(5)	Bouzit [1)]1994 [20]
1.0(3)	4.0(2)	1.68(6)	3.54(12)		10.2(4)	Baum [1)]1992 [21]
					10	D'Hondt 1980 [22]
0.05(2)	4.1(3)	1.8(3)	3.0(4)		8.9(6)	Vorobyov 1972 [23]
					90	Andreev 1969 [24]
					12	Blocki 1969 [25]

[1)] both values from partly the same experimental data .

Summary and Conclusions

In summary, already the first steps in the evaluation of the two complex experiments having been performed have brought new and more precise results on the yields and energy distributions of light charged particles. For the present contribution, in particular, the energies and yields of ternary Li isotopes for the two reactions studied were presented and discussed. As to the ^{236}U* data, it is worthwhile to note that the present results obtained with modern counting techniques nicely compare to previous data obtained exclusively with electromagnetic devices such as the LOHENGRIN installed at the high-flux reactor of the ILL, Grenoble. With this instrument, energy distributions have to be brought together point by point from measurements at various spectrometer settings. Main uncertainties concern the ionic charge states q, burn-up of the sample in the high neutron flux of more than 10^{14} neutrons/(cm^{2}s), and normalisation to ternary α-particles or ^{10}Be nuclei. Furthermore, the accessible energy range is limited by the electric fields to $E \leq 5$ MeV/q , resulting in $E_{Li} \leq 15$ MeV as demonstrated in Fig. 8 taken from a recent work by Oberstedt et al. [26]. In that work it was attempted to reach the higher energy values also, in an indirect way by slowing down the LCPs with a thick Ni degrader into the sensitive energy region of LOHENGRIN. This procedure naturally entails further sources of systematic errors. In a sense the present data on ^{235}U(n_{th},f) at the high-energy part of the spectra (Fig. 7) and data from LOHENGRIN (Fig. 8) at the lower part favorably complement one another with respect to the energy coverage. The combination of both sets of data may prove how accurate LCP spectra can indeed be described by single Gaussians.

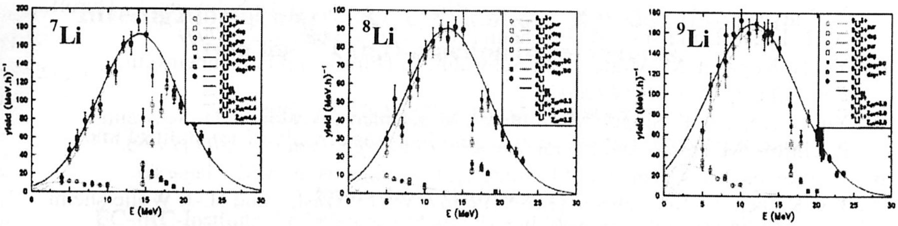

Fig. 8: Energy spectra of ternary Li isotopes in ^{236}U* measured at the LOHENGRIN mass separator at the ILL, Grenoble. Upper left experimental values ($E \leq 15$ MeV) were measured with a thin target, while the values at the high-energy descent of the distributions were recalculated from data measured with the sample covered by a thick absorber. Data are from S. Oberstedt et al. [26]

As to the Li yields in ^{252}Cf(sf), which cannot be measured with a separator such as LOHENGRIN, the situation is still quite confusing. The previous data measured by Dlouhý at al. [13] using a double-ΔE ionization chamber setup exceed the present results by a factor of up to 2.5. Published cumulative Li yields are even different by up to a factor of 4. Since all these data were deduced from the registration of residual energies after passage through some absorber foils or gas gaps, supplementation of the spectra towards energies below detection threshold has possibly introduced unexpectedly large systematic uncertainty.

Since particle separation and detection thresholds in the present experiment on ^{252}Cf(sf) (Fig. 6) were superior to the case of ^{236}U* (Fig. 7), and the latter data on extrapolated yields compare well with LOHENGRIN measurements, our Li yields on ^{252}Cf(sf) appear to be quite safe already at the current status of the analysis. Experimental errors could be dropped by more than an order of magnitude compared to Ref. [13]. Some fine tuning of the evaluation procedures is still under study and, therefore, the yields presented above, and the errors stated, should be considered as preliminary. Though, we are confident that no major changes will be incurred in the final results. With the yields and energy distributions of light charged particles already analysed the evaluation of the more involved LCP-FF correlations and the emission of γ-rays in ^{252}Cf ternary fission may be tackled.

Acknowledgements

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