Experimental Investigations of Ternary Fission of Heavy Nuclei

Yuri Kopatch

Frank Laboratory of Neutron Physics Joint Institute for Nuclear Research Dubna, Russia

Contents

- Introduction: What is Ternary Fission?
- Experimental Access by Advanced Correlation Experiments:

Crystal Ball (Nal) experiment

²⁵²Cf(sf) – MPI Heidelberg

Super-Clover Ge (VEGA) experiment

²⁵²Cf(sf) – GSI Darmstadt

Neutron-induced fission at ILL (Grenoble)

²³⁵U(n,f) – ILL Grenoble

Full energy spectrum of aparticles

²⁵²Cf(sf) – JYFL Jyvaskyla

Quaternary Fission experiments

²⁵²Cf(sf) – GSI Darmstadt ^{233,235}U(n,f) – ILL Grenoble

• Summary and Outlook

Selected Results:

- Neutron emission in TF D Formation of neutron-unstable LCPs (5He, 7He, 8Li*)
- Fragment spin alignment
- Isotope yields for ²⁵²Cf
- Isotope yields for ²³⁵U(n,f)
- Manifestation of shell effects in ternary fission
- Deviation of a-particle energy distribution from Gaussian shape
- Formation of two LCPs (a-a, a-t): "pseudo-" and "true" quaternary fission

Ternary Fission – nuclear fission, accompanied by emission of a Light Charged Particle (LCP accompanied Fission)

Quaternary Fission – simultaneous emission of 2 LCP's

Quinary Fission – 3 LCP's

Discovery of ternary fission



Tsien San-Tsiang et al., Phys. Rev. 71, 382 (1947)

Some Parameters of Ternary Fission



• Yield (TF) »

1/300 for ²⁵²Cf (sf)

1/500 for ²³⁵U (n,f)

• LCP's are emitted nearly perpendicular to the fission axis

• > 90% of all ternary particles are aparticles

• Yields of heavier particles drastically decreases with the increase of LCP mass

• The energy distribution of LCP has nearly Gaussian shape with $\langle E_a \rangle \gg 15$ MeV

• The emission of LCP slightly changes energy and mass distributions of fission fragments as well as other parameters, e.g. n, M_g etc.

• Ternary fission is a unique tool to study the energetics and dynamics of the fission process at scission.

LCP yields from n_{th}- induced reactions



F. Gönnenwein et al., Pont d'Oye (1999)



Angle-energy correlation for a-accompanied fission in polar coordinates.

Polar a's

Equatorial a's mean angle $q_{aL} = 83^{\circ}$



Crystal Ball Experiment for Measuring Spontaneous Ternary Fission of 252Cf

- Crystal Ball spectrometer as high efficiency neutron and gamma-ray detector
- New type of Ionization Chamber for measuring Fission Fragments
- A ring of ∆E-E telescopes for measuring Light Charged Particles

View of the experimental setup



CODIS - COmpact Dlogenes Setup



Ring of 12 DE-E telescopes

Double ionization chamber with sectored cathode



Double ionization chamber with sectored cathode



S2 Phi A \$3 S **S**4 1 V13 0.5 $R = V (V_{13} - 0.5)^2 + (V_{24} - 0.5)^2$ V₁₃ = S1+S3 **S2** Phi = arctan($\frac{V_{24}}{V_{13}}$) V24 = -S2+S4 Cosinus 6 <- 1.0 0.8-Cosinus 0 ~ 0.6 0.3->

Allows to measure energies and emission angles of the fission fragments

Ring of 12 DE-E Telescopes

Aim:

Measurement of energy, type and angle of emission of ternary particles

Construction: 1. Frisch grid ionization chamber for ΔE, using the same gas as the double ionization chamber
 2. PIN-diode for residual energy E



Separation of different ternary particles in a ΔE -E Plot:



DE-E method of identification of charge and mass of a particle



Crystal Ball Spectrometer



LCP energy and angular distributions



LCP	Events	Ethres MeV	<elcp> MeV</elcp>	FWHM MeV	Yield per 10 ⁴ a	v,
ЗН	5.6×10^{4}	3	8.2(6)	7.2(6)	950(90)	2.9
⁴ He	1.3×10 ⁶	8	15.7(2)	10.9(2)	104	3.1
⁶ He	2.6×10^{4}	10	12.3(5)	9.0(5)	270(30)	2.8
Li	2.5×10^{3}	17	14.3(10)	14.3(10)	60(10)	2.5
binary	7× 107					3.8

Fission Fragment kinetic energy and mass distributions for different ternary fission modes



Simplifies random neck rapture model







Average neutron multiplicity from fission fragments vs Total Excitation Energy



Possible additional sources of neutrons in ternary fission

 ${}^{5}He(3/2^{-}) \rightarrow {}^{4}He + n \quad \tau = 1.1 \times 10^{-21} \text{s} \quad Q = 0.85 \text{ MeV}$

 $^{7}He(3/2^{-}) \rightarrow ^{6}He + n \quad \tau = 4.1 \times 10^{-21} \text{s} \quad Q = 0.44 \text{ MeV}$

 $^{8}Li^{*}(3^{+}) \rightarrow ^{7}Li + n \quad \tau = 2 \times 10^{-20} \text{s} \quad Q = 0.25 \text{ MeV}$

 ${}^{6}He^{*}(2^{+}, E^{*} = 1.8 \text{ MeV}) \rightarrow {}^{4}He + 2n \quad \tau = 6 \times 10^{-21} \text{s}$ Q = 0.90 MeV

 ${}^{8}He^{*}(2^{+}, E^{*} = 2.8 \text{ MeV}) \rightarrow {}^{6}He + 2n \qquad \tau = ? \qquad Q = 0.69 \text{ MeV}$

Trajectory calculations of ternary fission



Neutron- and gamma-ray angular correlations: *"in situ"* coordinate system



Correlation of neutrons and LCPs





Correlation of neutrons and LCPs



Intermediate LCP	Resonance width, Γ	Lifetime, T	Q-value
$^{5}\text{He}(3/2^{\circ}) \rightarrow ^{4}\text{He} + n$	600 (20) keV	1.1 × 10 ⁻²¹ s	0.89 MeV
$^{7}\text{He}(3/2^{-}) \rightarrow ^{6}\text{He} + n$	160 (30) keV	4.1×10^{-21} s	0.44 MeV
${}^{8}\text{Li}^{*} \rightarrow {}^{7}\text{Li} + n$	33 (6) keV	2×10^{-20} s	0.25 MeV



Ternary α-particle spectrum of ²⁵²Cf(sf)

True ternary α 's and residues from ⁵He decay:

Two-Gaussian Fit:

One-Gaussian Fit:



_		-	
-		_	4
	a		****
	12.52		
	-		

		Experiment		Calculation	
LCP	< E > MeV	FWHM MeV	χ^2/N	< E > Me∀	FWHM MeV
⁴ He	15.7 (2)	10.9 (2)	5.9		
true ⁴ He	16.4 (3)	10.3 (3)	6.6	14.4	18.2
⁵ He residues	12.4 (3)	8.9 (5)		11.1	11.0
⁶ He	12.3 (5)	9.0 (5)	2.2		
true ⁶ He	12.6 (5)	8.9 (5)	2.2	9.9	10.5
⁷ He residues	11.0 (15)	8 (2)		8.6	8.6

E_a is 0.7 MeV higher than usually measured!

Super-Glover (VEGA) Experiment on ²⁵²Cf(sf) at GSI

Experimental Setup / Detectors

- New compact and high-efficiency fission-fragment and light-particle detector system "CODIS2"
- Two segmented large-volume super-clover Ge spectrometers (VEGA) as high-resolution γ-ray detectors

Experimental parameters

- high efficiency for ffs (\geq 90%) and LCPs (\approx 15%)
- good fragment energy resolution at high count-rate (2.5 x 10⁴ s-1)
- good isotopic LCP separation by dE-E (gas IC Si)
- ternary fission count-rate: 7 s-1 (4 x 10⁶ events total)
- \bullet large-volume high-resolution $\gamma\text{-ray}$ detectors in close geometry

Aim of the experiment

- γ-ray spectroscopy of fission fragments in <u>binary</u> and <u>ternary</u> fission (Doppler-shift correction for emission from fission fragments in flight).
- Angular anisotropy of individual fragment γ -rays in binary and ternary fission.
- Excited states in LCPs decaying by γ-rays. (Doppler-shift correction for emission from LCPs in flight). Li and Be isotopes including ¹⁰Be.
- Isotopic LCP yields (Li and Be) due to improved dE-E resolution of CODIS2.
- Improved data on the energy correlation between LCPs and FFs due to better LCP separation and statistics.

Experimental setup



Segmented Clover Ge detector



Experimental Set-up

Chamber and one of the two segmented clover Ge detectors





Measured parameters

Fragments:E, J, jLPCs:E, DE,Gammas:E, J, j

 E, J, j
 Þ
 M, v

 E, DE, J, j
 Þ
 Z, M, v

Examples of measured spectra



Fission fragments observed with DIC



Doppler-shift corrected g-ray spectra



No Doppler correction

LF-corrected spectrum

HF-corrected spectrum

Fragment-gamma angular correlations





DE-E separation of LCPs



Experimental and simulated ΔE -E distributions for identification of LCPs

Particle discrimination with the CODIS2 ΔE-E telescopes

The various steps in the analysis of LCP yields:



Identification of the He and Li Isotopes







PI = 5Z + (A-2Z)/2





Isotopic yields of He and Li LCPS in ²⁵²Cf(sf)

(relative to $10^4 \alpha$ -particles)

Isotope	⁶ He	⁸ He	⁶ Li	⁷ Li	⁸ Li	⁹ Li	¹¹ Li	
	315(20)	19(2)	1.2(3)	11.1(4)	5.4(2)	10.9(4)	≤ 0,3	This work (preliminary analysis)
	386(12)			∑ Li =	55(4)			G T Grachev et al.,1988. experiment
	403(26)	25(4)		17(4)	10(5)	25(11)		Z. Dlouhy et al., 1992, experiment
	315(60)	25(5)		∑ Li =	52(5)			P Singer, PHD, Darmst.,1997, exp
	369(15)	31(3)	0.3(1)	16 (2)	13(2)	32(3)		G.V. Valskii, 2004, model

²⁵²Cf(sf) experiment at GSI

First measurement of isotopic Be yields in ²⁵²Cf(sf)





•	GSI	Experiment

Isotope	⁷ Be	⁸ Be ^{*)}	⁹ Be	¹⁰ Be	¹¹ Be	¹² Be	
	?	10(6) [*]	17(3)	123(10)	10(4)	4(2)	This work (preliminary analysis)
	1.3x10 ⁻⁴	15(3)	28(5)	193(16)	[67(6)]	[52(6)]	G V Valskii, 2004, model
		23(2)	10(1)	160(20)	20(2)	25(2)	W Baum, PHD,Darmst., 1992, model
			∑Be =	164(9)			G T Grachev et al.,1988. experiment
			∑Be =	185(9)			Z. Dlouhy et al., 1992, experiment
			∑Be =	126(9)			P Singer, PHD, Darmst.,1997, exp.

Yields per 10⁴ α-particles

*) Kopatch et al., Phys.Rev.C65,044614 (2002)

Identification of the B and C Isotopes



no or weak evidence for 12C ? 35

The LCP-FF correlation experiment on TF and QF of $^{235}\text{U}(n_{th}^{}\,\text{,f})$ at the ILL-PF1

Experimental Setup / Detectors

- The compact and high-efficiency fission fragment and light particle detector system "CODIS2"
- Intense neutron beam PF1 (3 x 10⁹ n cm⁻² s⁻¹) from the ILL high-flux reactor

Experimental parameters

- high efficiency for ffs and LCPs
- good fragment energy resolution at very high count-rate (2.5 x 10⁵ s⁻¹)
- (good) isotopic LCP separation by dE-E
- ternary fission count-rate: 70 s⁻¹ (4 x 10⁷ events total)
- quaternary fission count-rate: 7x10⁻⁴ s⁻¹ (4 x 10² events total)

Aim of the experiment

- Energy spectra and yields of light LCPs due to improved dE-E resolution of CODIS2.
- Novel data on the energy correlation between ternary a's and FF's due to good LCP separation and high statistics.
- Data on the fragment energy and mass distributions correlated with LCPs other than ternary d's.
- First data on the fragment energy and mass distributions in quaternary fission.

Correlation experiment for the study of ternary and quaternary fission of 235U(nth,f), performed at the high-flux reactor of the ILL Grenoble (March/April 2004)

Detector system CODIS2





At the casemate of PF1 at the ILL

Schematic view

Ternary He and Li yields in ²³⁵U(n_{th},f)





Yields per $10^4 \alpha$ - particles

Isotope	⁶ He	⁸ He	⁷ Li	⁸ Li	⁹ Li	
	154(10)	4.5(3)	3.8(4)	1.6(2)	2.2(3)	Present work, preliminary
	191(8)	8.2(6)	4.1(3)	1.8(1)	3.0(4)	A.A. Vorobyov, 1972, exp.
	169(2)	7.4(8)	4.0(2)	1.7(1)	3.5(1)	W. Baum, PHD, Darmstadt, 1992, exp.
	140(70)	4.6(5)	3.6(2)	1.9(1)	3.4(3)	G.V. Valskii, 2004, model
			6.0(5)	1.3(3)	1.6(3)	W. Baum, PHD, Darmst., 1992, model

²³⁵U(n_{th},f) experiment at the ILL Spectra of ternary Li isotopes from ²³⁵U(n_{th},f)



LOHENGRIN data

ILL Report 3-01-380, S Oberstedt et al. 2000

²³⁵U(n_{th} ,f) experiment at the ILL

²³⁵U(n_{th},f) Mass Distribution from CODIS2



Measurement of Full a-particle Energy Spectrum Jyvaskyla (September-December 2005)



Measurement of Full a-particle Energy Spectrum Jyvaskyla (September-December 2005)



Schematic view

Measurement of Full a-particle Energy Spectrum Jyvaskyla (September-December 2005)



Measurement of Full a-particle Energy Spectrum Jyvaskyla (September-December 2005)





Measurement of Full a-particle Energy Spectrum Jyvaskyla (September-December 2005)



Low energy component which is not explained by ⁵He decay

Experiment for the study of quaternary fission of ²⁵²Cf(sf) at GSI (Darmstadt)



 E_{rest} (channels)

Experiment for the study of quaternary fission of ²³³U(n,f) and ²³⁵U(n,f) at ILL (Grenoble)



Angular distributions for a-a and a-t coincidences $^{252}\text{Cf}(\text{sf})$

α - α coincidences;

²³³U(n,f)



α-t coincidences:



a-a coincidences



Angular distributions from trajectory calculations



⁸Be ground state yield determination for ²⁵²Cf(sf)

LCP identification plot



Fit of the Li energy distribution with two componenets: true Li and $\alpha\text{-}\alpha$

Energy distributions of a-particles from "true" and "pseudo" quaternary fission of ²⁵²Cf(sf) and ²³³U(n,f)



Quaternary fission yields



Yields of LCPs in excited states P nuclear temperature in ternary fission

 $Y_{g.s.}\mu exp[E/T]$

Υ_{exc.s.}μ exp[(E+E*)/T]

 $Y_{exc.s.}/Y_{g.s.} = exp(E^*/T)$

Decay mode		Yield per 10 ⁴ TF α's				
		²⁵² Cf(sf)	²³³ U(n,f)	²³³ U(n,f)		
α – per binary fission	TF	3.3x10 ⁻³	2.1x10 ⁻³	1.7x10 ⁻³		
(α, α)	QF	3(1)	0,41(13)	0,32(10)		
8Be(g.s.)	QF	10(6)				
8Be*	QF	2(1)				
Σ8Be,8Be*	QF		0,94(30)	0,83(30)		
10Be	TF	140(15)	43(3)	30(2)		
(α,t)	QF	0,4(1)	0,03(1)	0,03(1)		
7Li*	QF	0,3(1)	0,03(1)	0,04(1)		
7Li	TF	17(4)	4,4(7)	4,1(3)		

Summary and Outlook

- Ternary fission is a unique tool for studying the behavior of the nuclear system at the moment close to scission.
- It is only possible to get access to the nuclear fission dynamics by multiparameter measurements of all fission components.
- High resolution and high efficiency gamma-ray and neutron spectroscopy of ternary fission is a step forward in understanding of this rare process.
- It would be interesting to observe a "quinary" fission splitting of a nucleus into 5 fragments (2FF and 3 LCP)

Authors List

Super-Clover (VEGA) experiment with ²⁵²Cf(sf) at GSI:

Crystal-Ball experiment with ²⁵²Cf(sf) at MPI-K:

P. Singer	IKP TU Darmstadt
Yu.N. Kopatch	JINR Dubna
M. Mutterer	IKP TU Darmstadt
M. Klemens	IKP TU Darmstadt
J. von Kalben	IKP TU Darmstadt
A. Hotzel	MPI-K Heidelberg
D. Schwalm	MPI-K Heidelberg
P. Thirolf	MPI-K Heidelberg
F. Gönnenwein	Univ. Tübingen
M. Hesse	Univ. Tübingen

Yu.N. Kopatch P. Jesinger M. Mutterer E. Lubkiewicz H.-J. Wollersheim I. Kojouharov J. von Kalben W.H. Trzaska Z. Mezentseva F. Gönnenwein

JINR Dubna, GSI Darmstadt IKP TU Darmstadt IKP TU Darmstadt Jag. Univ. Cracow GSI Darmstadt GSI Darmstadt IKP TU Darmstadt Univ. Jyväskylä FLNR-JINR Dubna Univ. Tübingen

LCP-LCP correlation experiment with ²⁵²Cf(sf) at GSI:

Yu.N. Kopatch	JINR Dubna, GSI Darmstadt
HJ. Wollersheim	GSI Darmstadt
M. Mutterer	IKP TU Darmstadt
J. von Kalben	IKP TU Darmstadt

QF experiment with ²³³U(n,f) and ²³⁵U(m,f) at ILL:

P.Jesinger
M.Mutterer
F.Gönnenwein
A.M.Gagarski
J.v. Kalben
V.Nesvizhevsky
G.A.Petrov
W.H.Trzaska

Univ. Tübingen IKP TU Darmstadt Univ. Tübingen **PNPI** Gatchina IKP TU Darmstadt **ILL Grenoble** PNPI Gatchina Univ. Jyväskylä

Ternary Fission experiment with ²³⁵U(n,f) at ILL:

Yu N Kopatch V Tishchenko M Mutterer M Speransky F Gönnenwein A M Gagarski V Nesvizhevsky W H Trzaska

JINR Dubna **JINR Dubna** IKP TU Darmstadt JINR Dubna Univ. Tübingen PNPI Gatchina ILL Grenoble Univ. Jyväskylä

Ternary Fission experiment with ²³⁵U(n,f) at ILL:

M Mutterer
Yu N Kopatch
W H Trzaska
S.Yamaletdinov
V.Lyapin
S.Khlebnikov
G.Tyurin

IKP TU Darmstadt JINR Dubna Univ. Jyväskylä Univ. Jyväskylä Univ. Jyväskylä **RI St.Petersburg RI St.Petersburg**