Systematic measurements of prompt fission γ-rays and what they tell us about fission fragment de-excitation

Andreas Oberstedt

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WONDER-2018, Aix-en-Provence (France), October 8 – 12, 2018
The fission process

- Scission
- 90% $E_k$
- Prompt n emission
- Prompt γ emission
- $\beta^-$ decay delayed n and γ

Time (s): $10^{-15}$, $10^{-13}$, $10^{-10}$, $10^{-7}$

Distance (m): $10^{-6}$
The fission process

For reactor applications:
- nuclear energy
- chain reaction
- heat
- reactor control

scission 90% $E_k$ prompt n emission prompt $\gamma$ emission $\beta^-$ decay delayed n and $\gamma$
The fission process

Many nuclear data needs, but: our main focus of research (HPRL by OECD/NEA)

For reactor applications: nuclear energy chain reaction heat reactor control

scission $90\% E_k$ prompt n emission prompt $\gamma$ emission $\beta^-$ decay delayed n and $\gamma$

10$^{-9}$ $10^{-6}$ $10^{-15}$ $10^{-13}$ $10^{-10}$ $10^{-7}$ t (s)
d (m)
Outline

• Introduction
• Experimental setup
• Data treatment
• Results: PFGS
  • characteristics
  • dependence of compound system
  • impact of excitation energy
  • angular distribution & multipolarities
• Summary
• Outlook
Introduction

• For the past years: precise measurement of prompt fission $\gamma$-ray spectra (PFGS)
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• Determination of characteristics:
  • $< M_{\gamma}>$, $< \varepsilon_{\gamma}>$, and $<E_{\gamma,\text{tot}}>$
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• Study of the dependence of $A$ and $Z$
Introduction

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- Study of energy dependence
Introduction

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- Determination of characteristics:
  - $< M_\gamma >$, $< \varepsilon_\gamma >$, and $< E_{\gamma,\text{tot}} >$
- Study of the dependence of $A$ and $Z$
- Study of energy dependence
- Details about the de-excitation process of fission fragments
Experimental setup

Fission fragment – γ-ray coincidences

- (Frisch-grid) ionization chamber
  (fission trigger)

- LaBr₃:Ce and CeBr₃ scintillation detectors
  (plus BaF₂ and/or NaI:Tl/LaBr₃:Ce phoswich detectors
  (gamma rays))

- Coincidences

- Time-of-flight measurement
  (n/γ discrimination)
Experimental setup
n/γ discrimination by time-of-flight

Due to good resolving power + excellent timing resolution of LaBr₃:Ce detectors

Prompt fission γ-rays
γ-decay after inelastic neutron scattering
Intrinsic and external background
Experimental setup

\( n/\gamma \) discrimination by time-of-flight

**Experimental Setup**

Due to good resolving power + excellent timing resolution of LaBr\(_3\):Ce detectors

**Graph:**
- **Time-of-flight (ns)** vs. **Energy (keV)**
  - **n, n’**
  - **E \approx 847 \text{ keV}**
  - **E \approx 1436 \text{ keV}**
  - **E \approx 276 \text{ keV}**

**Prominent Features:**
- **Prompt fission \( \gamma \)-rays**
- **\( \gamma \)-decay after inelastic neutron scattering**
- **Intrinsic and external background**

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Data treatment
Unfolding the detector response

measured spectrum

\[
\begin{pmatrix}
y_1 \\
y_2 \\
\vdots \\
y_n
\end{pmatrix}
\]

measured spectrum

\[
\begin{pmatrix}
y_1' \\
y_2' \\
\vdots \\
y_n'
\end{pmatrix}
\]

\[
\begin{pmatrix}
x_1 \\
x_2 \\
\vdots \\
x_n
\end{pmatrix}
\]

\[
\begin{pmatrix}
x_1' \\
x_2' \\
\vdots \\
x_n'
\end{pmatrix}
\]
Data treatment
Unfolding the detector response

\[ \begin{pmatrix}
  y_1 \\
  y_2 \\
  \vdots \\
  y_n
\end{pmatrix} =
\begin{pmatrix}
  r_{11} & r_{12} & \cdots & r_{1n} \\
  r_{21} & \vdots & \ddots & \vdots \\
  \vdots & \ddots & \ddots & \vdots \\
  r_{n1} & \cdots & r_{nn}
\end{pmatrix}
\begin{pmatrix}
  x_1 \\
  x_2 \\
  \vdots \\
  x_n
\end{pmatrix} \]
Data treatment
Unfolding the detector response

\[ \begin{pmatrix} y_1 \\ y_2 \\ \vdots \\ y_n \end{pmatrix} = \begin{pmatrix} r_{11} & r_{12} & \cdots & r_{1n} \\ r_{21} & \vdots & & \vdots \\ \vdots & & \ddots & \vdots \\ r_{n1} & \cdots & & r_{nn} \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ \vdots \\ x_n \end{pmatrix} \]

measured spectrum

emission spectrum

response matrix

usually simulated \((\text{GEANT4}, \text{PENELOPE})\)

Counts/fission

\[ E_Y \text{ (keV)} \]

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Data treatment

Unfolding the detector response

\[
\begin{pmatrix}
  y_1 \\
  y_2 \\
  \vdots \\
  y_n \\
\end{pmatrix}
= 
\begin{pmatrix}
  r_{11} & r_{12} & \cdots & r_{1n} \\
  r_{21} & r_{22} & \cdots & r_{2n} \\
  \vdots & \vdots & \ddots & \vdots \\
  r_{n1} & r_{n2} & \cdots & r_{nn} \\
\end{pmatrix}
\begin{pmatrix}
  x_1 \\
  x_2 \\
  \vdots \\
  x_n \\
\end{pmatrix}
\]

usually simulated (\textit{Geant4, Penelope})
Data treatment
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emission spectrum

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\vdots \\
y_n \\
\end{pmatrix} =
\begin{pmatrix}
r_{11} & r_{12} & \cdots & r_{1n} \\
r_{21} \\
\vdots \\
r_{n1} & \cdots & r_{nn} \\
\end{pmatrix}
\begin{pmatrix}
x_1 \\
x_2 \\
\vdots \\
x_n \\
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\]

response matrix
emission spectrum

measured spectrum
Results: PFGS

Overview: studied systems so far

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Compound systems
Results: PFGS
Overview: studied systems so far

Previous work: (sf)
Results: PFGS
Overview: studied systems so far

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<td>Np 242</td>
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<td>1.85 a</td>
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Previous work: (sf), (n_{th}, f)
Results: PFGS

Overview: studied systems so far

Previous work: (sf), (n_{th}, f), (n, f)

compound systems
Results: PFGS
Overview: studied systems so far

Previous work: (sf), (n\text{th}, f), (n, f), (d, pf)
Results: PFGS
Overview: studied systems so far

Previous work: (sf), (n,th, f), (n, f), (d, pf)
### Results: PFGS

#### Overview: studied systems so far

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<th>Half-Life</th>
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<td>Cf 240</td>
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#### Previous work:
- $(sf)$
- $(n_{th}, f)$
- $(n, f)$
- $(d, pf)$

#### Recent experiments:
- $(sf)$

*compound systems*
Results: PFGS

Overview: studied systems so far

<table>
<thead>
<tr>
<th>Compound Systems</th>
<th>Previous work:</th>
<th>Recent experiments:</th>
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<tbody>
<tr>
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<td>(sf), (n,&lt;sub&gt;th&lt;/sub&gt;, f)</td>
<td>(sf), (n,&lt;sub&gt;th&lt;/sub&gt;, f)</td>
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<td>(n, f), (d, pf)</td>
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</table>
Results: PFGS

Overview: studied systems so far

Previous work: (sf), (n, f), (n, th, f), (d, pf)

Recent experiments: (sf), (n, th, f)
Results: PFGS

Overview: studied systems so far

Previous work: (sf), (n_{th}, f), (n, f), (d, pf)

Recent experiments: (n_{th}, f)

Approved proposals: (n, f)
Results: PFGS
High precision $\gamma$-ray measurements

Excellent agreement between our experimental results and those from advanced model calculations *)

*) full Hauser-Feshbach Monte Carlo simulations by
• D. Regnier et al. (code: FIFRELIN, CEA Cadarache)
• P. Talou et al. (code: CGMF, LANL)
Results: PFGS
High precision $\gamma$-ray measurements

Examples for different compound systems
Results: PFGS
High precision $\gamma$-ray measurements

Examples for different compound systems

Similar low energy peak structures!
Results: PFGS
High precision $\gamma$-ray measurements

Similar low energy peak structures!

Due to de-excitation of the (same) heavy fragments?

(J. Wagemans)
Results: PFGS
High precision $\gamma$-ray measurements

Examples for different compound systems
Results: PFGS
High precision $\gamma$-ray measurements

Examples for different compound systems

PFGS characteristics:

$$
\overline{M}_\gamma = \int N_{\gamma}(E_\gamma)dE_\gamma
$$

$$
E_{\gamma,\text{tot}} = \int E_\gamma \times N_{\gamma}(E_\gamma)dE_\gamma
$$

$$
\epsilon_\gamma = \frac{E_{\gamma,\text{tot}}}{\overline{M}_\gamma}
$$
Results: PFGS
High precision $\gamma$-ray measurements

Examples for different compound systems

PFGS characteristics:

- Systematics!

$$\overline{M}_\gamma = \int N_\gamma(E_\gamma) dE_\gamma$$

$$E_{\gamma,\text{tot}} = \int E_\gamma \times N_\gamma(E_\gamma) dE_\gamma$$

$$\epsilon_\gamma = \frac{E_{\gamma,\text{tot}}}{\overline{M}_\gamma}$$
Results: A and Z dependence
Systematics of PFGS average total energy per fission

According to Nifenecker (1972) and Valentine (2001), revised 2017: A. Oberstedt et al., PRC 96, 034612
According to Nifenecker (1972) and Valentine (2001), revised 2017: A. Oberstedt et al., PRC 96, 034612

Results: A and Z dependence
Systematics of PFGS average total energy per fission

$[E_{y,\text{tot}} (\text{MeV}) - 4.0] / n_f$ only!

$10^{-5} Z^2 A^{1/2}$

$234, 236\text{U}$, $240, 242\text{Pu}$, $252\text{Cf}$

Our work
New evaluation
Experiments ≤ 1973
Valentine 2001

Andreas Oberstedt
WONDER-2018, Aix-en-Provence (France), October 8 – 12, 2018
According to Nifenecker (1972) and Valentine (2001), revised 2017: A. Oberstedt et al., PRC 96, 034612

Results: A and Z dependence
Systematics of PFGS average total energy per fission

\[ \frac{[E_{y,\text{tot}} (\text{MeV}) - 4.0]}{n} \]

\[ 10^{-5} \cdot Z^2 \cdot A^{1/2} \]

\( (n_{\text{th},f}) \) and \( (sf) \) only!
Results: A and Z dependence
Systematics of PFGS average total energy per fission

Allows interpolation to unmeasured fissioning systems, here $^{238}\text{U}(n_{\text{th}},f)$: A. Oberstedt et al., PRC 96, 034612

$E_{y,\text{tot}}$ (MeV) - 4.0 vs $\nu_n$ vs $10^{-5}Z^2A^{1/2}$

(n$_{\text{th}},f$) and (sf) only!
Results: energy dependence
Average total energy per fission
From thermal to fast neutron-induced fission

- Tudora: Point-by-Point model
- CEA DAM/DIF & LICORNE: preliminary experimental results
Results: energy dependence
Average $\gamma$-ray multiplicity
From thermal to fast neutron-induced fission

- CEA DAM/DIF & LICORNE: preliminary experimental results

A. Oberstedt et al., PRC 96, 034612 (2017)
Results: energy dependence
Mean energy per photon

From thermal to fast neutron-induced fission

- CEA DAM/DIF & LICORNE: preliminary experimental results

A. Oberstedt et al., PRC 96, 034612 (2017)
Experimental setup
Frisch grid ionization chamber + LaBr$_3$ detector

Correlations between fission fragments and $\gamma$-rays
Results: angular distribution
Prompt fission $\gamma$-ray multipolarities

$W(\theta) = \frac{I_{\gamma}(\theta)}{A_0} = A_0 \left[ 1 + \left\{ \frac{A_2}{A_0} \right\} P_2(\cos\theta) + \left\{ \frac{A_4}{A_0} \right\} P_4(\cos\theta) \right]$
Results: angular distribution

Prompt fission $\gamma$-ray multipolarities

Fit result: $\{A_2/A_0\} = 0.13 \pm 0.03$
Results: angular distribution

Angular Distributions in $^{109}$Te

- Typically $A_4/A_0$ is close to zero
- And $A_2/A_0 \sim +0.3$ for a pure quadrupole ($\Delta I = 2$) transition
- Or $A_2/A_0 \sim -0.3$ for a pure dipole ($\Delta I = 1$) transition
Results: angular distribution
Prompt fission $\gamma$-ray multipolarities

Theory: $\{A_2/A_0\} = +0.3$ for quadrupole radiation
$\{A_2/A_0\} = -0.3$ for dipole radiation

Statistical + systematic uncertainties!
Results: angular distribution
Comparison with previous measurements *)

\[ W(\theta) = \frac{I(\theta)}{A_0} \]

\[ E_\gamma = 0.1 - 7.2 \text{ MeV} \]

*) Hoffman, Phys. Rev. 133 (1964)
Results: angular distribution
Comparison with previous measurements *)

Good agreement in dominating E2 character!

*) Hoffman, Phys. Rev. 133 (1964)

\[ E_\gamma = 0.1 - 7.2 \text{ MeV} \]
Results: PFGS
Angular distributions for 500 keV energy bins

$\gamma(E, \theta)$

$E_\gamma$ (MeV)

$\cos(\theta)$

$^{252}\text{Cf}(sf)$
Results: PFGS

Angular distributions for 500 keV energy bins

$\gamma$-ray angular distribution

$M_{\gamma} (E, \theta)$

$E_\gamma$ (MeV)

$\cos(\theta)$

$^{252}$Cf(sf)

$\rightarrow$ consider energy range $E_\gamma = 0.1 - 1.5$ MeV
Results: angular distribution
Comparison with previous measurements

For $E_\gamma = 0.1 - 1.5$ MeV:
Results: angular distribution
Comparison with previous measurements

\[ W(\theta) = \frac{I_\gamma(\theta)}{A_0} \]

\[ ^{252}\text{Cf}(\text{sf}) \]

For \( E_\gamma = 0.1 - 1.5 \text{ MeV} \):
\( \rightarrow \) differences!

\( \text{*) Kopach et al., Phys. Rev. Lett. 82 (1999) } \)
Results: PFGS
Angular distributions for 500 keV energy bins

$252\text{Cf}(sf)$

$\rightarrow$ again: fit of Legendre polynomials

$\rightarrow$ decomposition of multipolarities $L = 1$ and 2
Results: PFGS
Decomposition of multipolarities

\[ 252 \text{Cf(sf)} \]

\[ \text{Photons} / (\text{MeV fission}) \]

\[ \text{E}_\gamma (\text{MeV}) \]

\[ \cdot \text{This work} \]
\[ \text{binned} \]
\[ L = 1 \]
\[ L = 2 \]

\[ \rightarrow \text{multipolarity-dependent spectra} \]
\[ \rightarrow \text{multipolarity-dependent PFGS characteristics} \]
Results: PFGS
Comparison with FIFRELIN calculations *)

Good agreement between integral spectra!

*) A. Chebboubi, priv. comm.
Results: PFGS
Comparison with FIFRELIN calculations *)

Good agreement between integral spectra!
But FIFRELIN also provides multipolarity-dependent PFGS.

*) A. Chebboubi, priv. comm.
Results: PFGS
Comparison with FIFRELIN calculations *)

From our observations: unassigned transitions → L = 2,
L = 2 + unassigned → L = 2'.

*) A. Chebboubi, priv. comm.


Results: angular distribution
Comparison with FIFRELIN calculations *)

<table>
<thead>
<tr>
<th>$^{252}$Cf(sf)</th>
<th>Experiment (this work)</th>
<th>Calculations (FIFRELIN)</th>
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<tr>
<td>$\overline{M}_\gamma$</td>
<td>8.28 ± 0.51</td>
<td>8.28 (adjusted)</td>
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<td>$\overline{M}_\gamma$ (L = 1)</td>
<td>2.40 (29 %)</td>
<td>3.20 (39 %)</td>
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<td>$\overline{M}_\gamma$ (L = 2')</td>
<td>5.88 (71 %)</td>
<td>5.08 (61 %)</td>
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<td>$\overline{M}_\gamma$ (unassign.)</td>
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<tr>
<td>$\overline{E}_\gamma$</td>
<td>0.81 ± 0.10 (MeV)</td>
<td>0.76 (MeV)</td>
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<td>$\overline{E}_\gamma$ (L = 1)</td>
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<td>0.94 (MeV)</td>
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<td>0.65 (MeV)</td>
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<tr>
<td>$\overline{E}_\gamma$</td>
<td>6.75 ± 0.76 (MeV)</td>
<td>6.30 (MeV)</td>
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*) A. Chebboubi, priv. comm.
Summary I

- High precision PFGS measurements → reference for model calculations – e.g.: $^{252}\text{Cf}(sf)$
Summary I
Sequential emission of neutrons and $\gamma$-rays

Entry region for primary fragments

Entry region for secondary fragments

$^{252}\text{Cf}(sf)$

$\gamma$-rays

(dipole transitions)

(discrete levels)

(quadrupole transitions)

Statistical $\gamma$-rays
Summary I

Sequential emission of neutrons and γ-rays

$^{252}\text{Cf}(sf)$

Entry region for primary fragments

Entry region for secondary fragments

$M_\gamma \approx 8.3$

$\Delta E_\gamma \approx 6.7$ MeV

$S_n$

$E^*$

$E(\text{Yrast})$

$E^*_{\text{lim}}$

Statistical γ-rays (dipole transitions)

Discrete levels (quadrupole transitions)
Summary I

- High precision PFGS measurements → reference for model calculations – e.g.: $^{252}$Cf(sf)
- Revised **systematics** for spontaneous and thermal neutron-induced fission
- Predictions of PFGS characteristics for fast neutron-induced fission → rather good agreement
- Measured γ-ray **angular distribution** from $^{252}$Cf(sf) → dominant E2 character, in good agreement with previous observations + FIFRELIN calculations
- Preliminary results: \(<M_{\gamma,L=1}> \approx 2.4\) and \(<M_{\gamma,L=2}> \approx 5.9\), as well as \(<E_{\gamma,\text{tot}(L=1)}> \approx 2.1\ \text{MeV}\) and \(<E_{\gamma,\text{tot}(L=2)}> \approx 4.6\ \text{MeV}\)
Summary I
Sequential emission of neutrons and $\gamma$-rays

252\text{Cf}(sf)

Entry region for primary fragments

Entry region for secondary fragments

$\Delta E_\gamma \approx 2.1$ MeV
$\Delta E_\gamma \approx 4.6$ MeV

preliminary!

Sequential emission of neutrons and $\gamma$-rays

$\gamma$-rays

$dipole transitions$

$quadrupole transitions$

$M_\gamma \approx 2.4$

$M_\gamma \approx 5.9$

statistical $\gamma$-rays

discrete levels

$prior$
Summary I
Sequential emission of neutrons and γ-rays

$^{252}\text{Cf}(\text{sf})$

Sequential emission of neutrons and γ-rays

$S_n/2$

$E^*$

Entry region for primary fragments

Entry region for secondary fragments

$\gamma$

$\gamma$

$n$

$n$

$n$

$\Delta E_\gamma \approx 2.1 \text{ MeV}$

$\Delta E_\gamma \approx 4.6 \text{ MeV}$

$M_\gamma \approx 2.4$

$M_\gamma \approx 5.9$

Statistical γ-rays (dipole transitions)

Discrete levels (quadrupole transitions)

Preliminary!
Summary I

✚ High precision PFGS measurements → reference for model calculations – e.g.: $^{252}$Cf(sf)

✚ Revised systematics for spontaneous and thermal neutron-induced fission

✚ Predictions of PFGS characteristics for fast neutron-induced fission → rather good agreement

✚ Measured γ-ray angular distribution from $^{252}$Cf(sf) → dominant E2 character, in good agreement with previous observations + FIFRELIN calculations

✚ Preliminary results: $\langle M_{\gamma, L=1} \rangle \approx 2.4$ and $\langle M_{\gamma, L=2} \rangle \approx 5.9$, as well as $\langle E_{\gamma, \text{tot}(L=1)} \rangle \approx 2.1$ MeV and $\langle E_{\gamma, \text{tot}(L=2)} \rangle \approx 4.6$ MeV

✚ Average spin of fission fragments: $\Delta J \approx 14 \hbar$
Summary I

Sequential emission of neutrons and γ-rays

252\text{Cf}(sf)

Entry region for primary fragments

Entry region for secondary fragments

\Delta J \approx 14 \hbar

S_{n}/2

M_{\gamma} \approx 2.4

\Delta E_{\gamma} \approx 2.1 \text{ MeV}

statistical γ-rays (dipole transitions)

discrete levels (quadrupole transitions)

\Delta J \approx 14 \hbar

M_{\gamma} \approx 5.9

\Delta E_{\gamma} \approx 4.6 \text{ MeV}

preliminary!
Summary II

But:

- Discrepancies compared to Kopach et al., PRL 88 (1999)
- From angular distribution:
  \[ <S_n> = 2 \times <E_{\gamma,\text{tot}(L=1)}> \approx 4.2 \text{ MeV}, \text{ while weighted with fission fragment distribution: } <S_n> \approx 5.9 \text{ MeV}! \]
  (fission fragment distributions from GEF, \( S_n(Z,A) \) according to Vogt et al., PLB 517 (2001))
- High energy quadrupole \( \gamma \) rays (of several MeV) observed, whose origin cannot be explained with rotational states

• To be continued ...
Outlook I

• New results from recent measurements
• New experiments are approved and scheduled
• Study of PFGS characteristics depending on fission fragment mass
• Study of entrance channel effects
  • \((n,f)\) vs. \((d,pf)\)
  • \((p,p’f)\) vs. \((\gamma,f)\)
  • etc.
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• New instruments (talk by S. Oberstedt)
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• New instruments (talk by S. Oberstedt)
• And last but not least:
  → Photo-fission at ELI-NP !
Outlook II
ELI-NP and further photo-fission physics goals

ELIADDE

8 CLOVER Ge detectors + 4 large-volume LaBr₃ detectors (3” x 3”)

ELIGANT

17 LaBr₃ + 17 CeBr₃ detectors (3” x 3”) and 33 liquid + 29 ⁶Li glass scintillation detectors
Outlook II
ELI-NP and further photo-fission physics goals

New position-sensitive twin FGIC (TU Darmstadt)
+
ELIADE
8 CLOVER Ge detectors + 4 large-volume LaBr$_3$ detectors (3” x 3”)
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17 LaBr$_3$ + 17 CeBr$_3$ detectors (3” x 3”) and 33 liquid + 29 $^6$Li glass scintillation detectors
Outlook II

ELI-NP and further photo-fission physics goals

New position-sensitive twin FGIC (TU Darmstadt)

- ELIADE
- ELIGANT

(courtesy M. Peck)

- Study of the fission fragment de-excitation process
  - measurement of fission fragments, γ rays and neutrons
  - correlations!
The collaborators


PhD students
Thank you!