



Study of the surrogate-reaction method via the simultaneous measurement of fission and gamma-decay probabilities (ACTISUR->ACTInides-SURrogate)

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Need for neutron-induced cross sections on short-lived nuclei : transmutation of nuclear waste

240Cm 27 D α : > 99.50% ϵ : < 0.50%	241Cm 32.8 D ϵ : 99.00% α : 1.00%	242Cm 162.8 D α : 100.00% SF: 6.2E-6%	243Cm 29.1 Y α : 99.71% ϵ : 0.29%	244Cm 18.1 Y α : 100.00% SF: 1.4E-4%	245Cm 8500 Y α : 100.00% SF: 6.1E-7%	246Cm 4760 Y α : 99.97% SF: 0.03%	247Cm 1.56E+7 Y α : 100.00%	248Cm 3.48E+5 Y α : 99.61% SF: 8.3E-6%
239Am 11.9 H ϵ : 99.99% α : 0.01%	240Am 50.8 M ϵ : 100.00% α : 1.9E-4%	241Am 432.6 Y α : 100.00% SF: 4E-10%	242Am 16.02 H β :-: 82.70% ϵ : 17.30%	243Am 7370 Y α : 100.00% SF: 3.7E-9%	244Am 10.1 H β :-: 100.00%	245Am 2.05 H β :-: 100.00%	246Am 39 M β :-: 100.00%	247Am 23.0 M β :-: 100.00%
238Pu 87.7 Y α : 100.00% SF: 1.9E-7%	239Pu 24.110 Y α : 100.00% SF: 3.8E-10%	240Pu 6561 Y α : 100.00% SF: 5.7E-6%	241Pu 14.290 Y β :-: 100.00% α : 2.5E-3%	242Pu 3.75E+5 Y α : 100.00% SF: 5.5E-4%	243Pu 4.956 H β :-: 100.00%	244Pu 8.00E+7 Y α : 99.88% SF: 0.12%	245Pu 10.5 H β :-: 100.00%	246Pu 10.84 D β :-: 100.00%
237Np 2.144E+6 Y α : 100.00% SF: 2E-10%	238Np 2.117 D β :-: 100.00%	239Np 2.356 D β :-: 100.00%	240Np 61.9 M β :-: 100.00%	241Np 13.9 M β :-: 100.00%	242Np 2.2 M β :-: 100.00%	243Np 1.85 M β :-: 100.00%	244Np 2.29 M β :-: 100.00%	
236U 2.342E7 Y α : 100.00% SF: 9.4E-8%	237U 6.75 D β :-: 100.00%	238U 4.468E9 Y 99.2742% α : 100.00% SF: 5.5E-5%	239U 23.45 M β :-: 100.00%	240U 14.1 H β :-: 100.00%	241U $\approx$ 5 M β -	242U 16.8 M β :-: 100.00%		
235Pa 24.44 M β :-: 100.00%	236Pa 9.1 M β :-: 100.00%	237Pa 8.7 M β :-: 100.00%	238Pa 2.27 M β :-: 100.00% SF: 2.6E-6%	239Pa 1.8 H β :-: 100.00%	240Pa $\approx$ 2 M β -			

Minor actinides

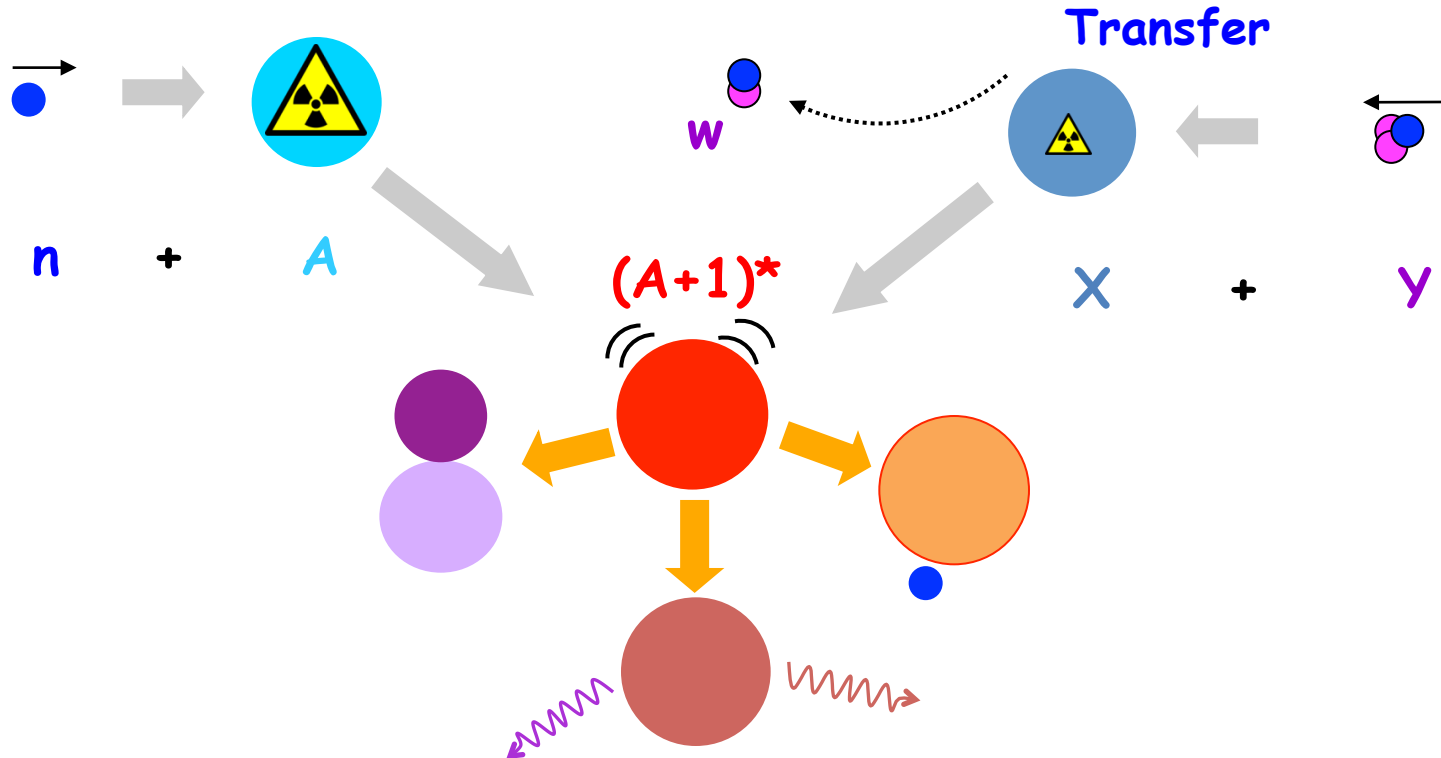
**Neutron-induced fission and capture cross sections of short-lived nuclei needed.
Very difficult or even impossible to measure!**

Surrogate-reaction method

Cramer and Britt (Los Alamos 1970...!!)

Neutron-induced reaction

Surrogate reaction



$$\sigma_{n,decay}^A(E^*) = \underbrace{\sigma_{CN}^{A+1}(E^*)}_{\text{Theory Optical model}} \cdot \underbrace{P_{decay}^{surro}(E^*)}_{\text{Experiment}}$$

Validity of the surrogate method

$$\sigma_{n,decay}^A(E^*) = \sigma_{CN}^{A+1}(E^*) \cdot P_{decay}^{surro}(E^*)$$

Neutron-induced and surrogate reaction must achieve **statistical equilibrium** (compound nucleus formation):
Decay only depends on E^* , J and π !!

$$P_{decay}^{surro}(E^*) = P_{decay}^n(E^*)$$

Populated J and π distributions
are equal

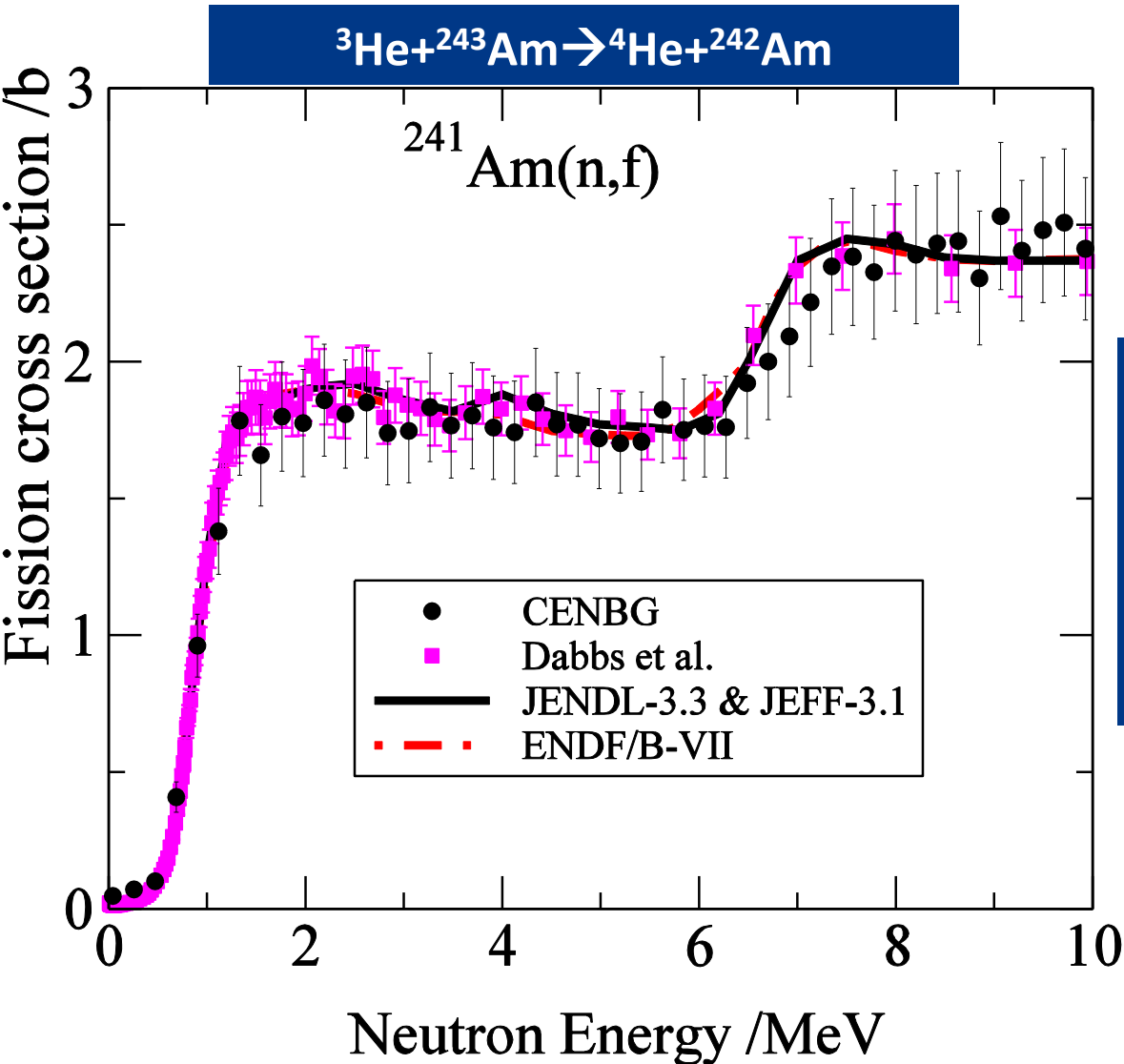
OR

Decay independent of J and π
(Only valid at high E^* in
the Weisskopf-Ewing limit)

Not possible to say a priori if a reaction meets these
conditions.

Data obtained with the surrogate method need
to be compared to neutron-induced data!

Results for fission



General finding: the cross sections obtained with surrogate method are in good agreement with n-induced data for fission!

G. Kessedjian et al., Phys. Lett. B 692 (2010) 297

G. Kessedjian et al., Phys. Rev. C 91 (2015) 044607

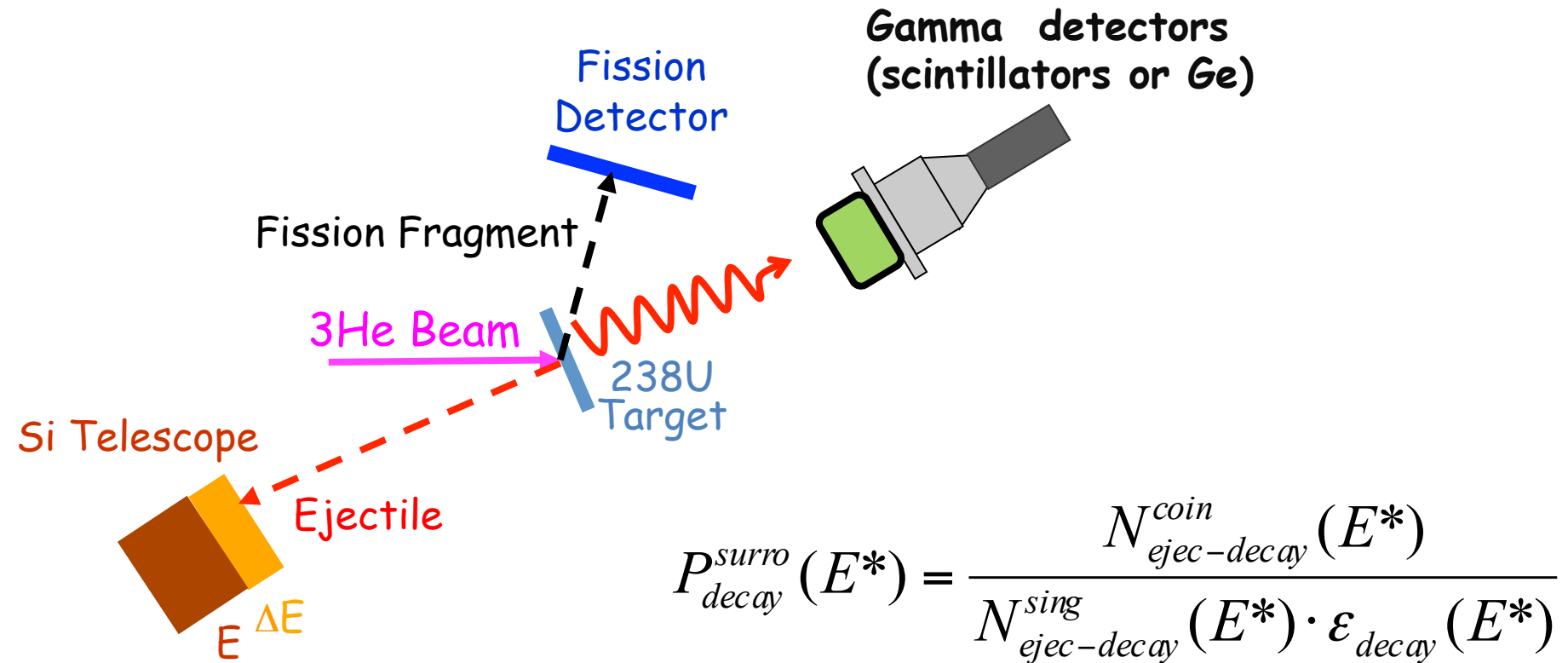
**Does the surrogate method work also for
radiative capture?**

Objective:

**Simultaneous measurement of fission and
gamma-decay probabilities.**

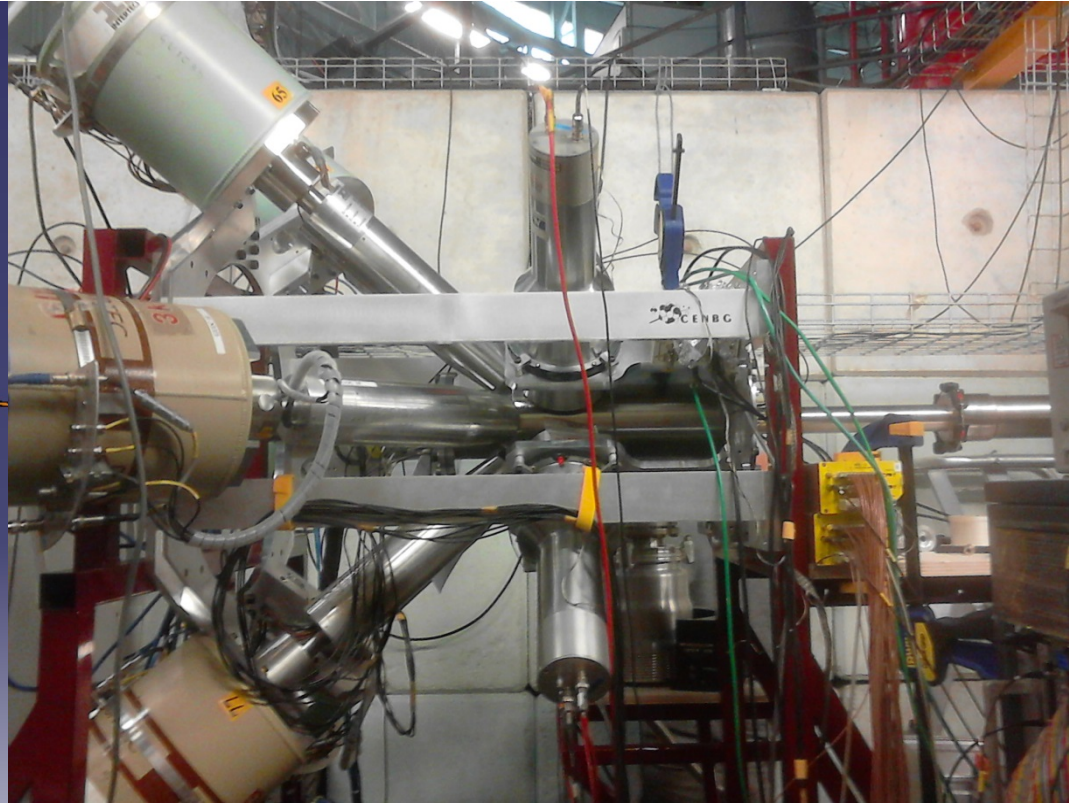
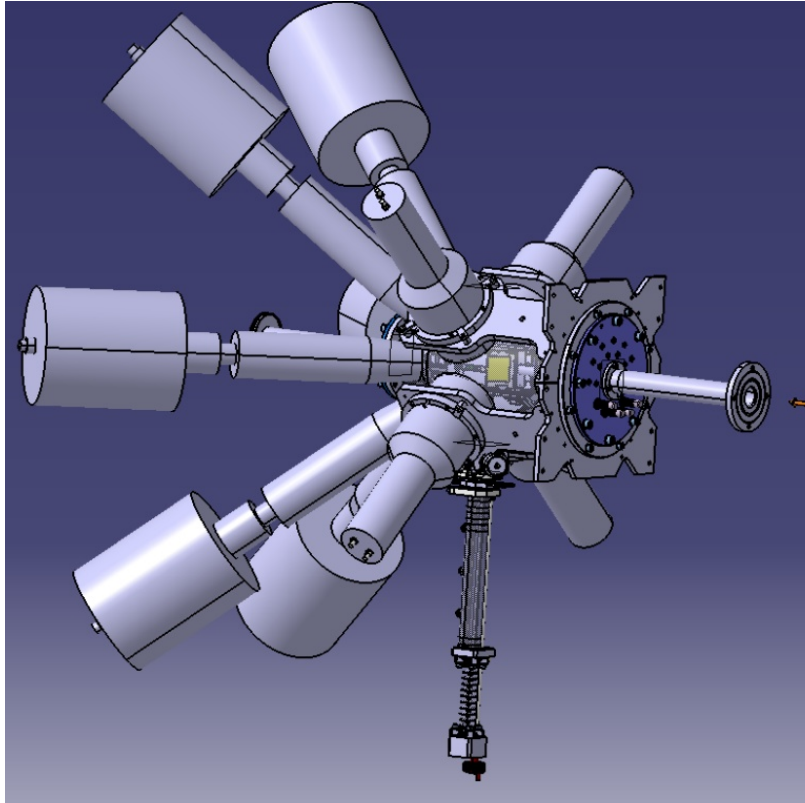
Never done before!

Setup for simultaneous measurement of fission and gamma-decay probabilities



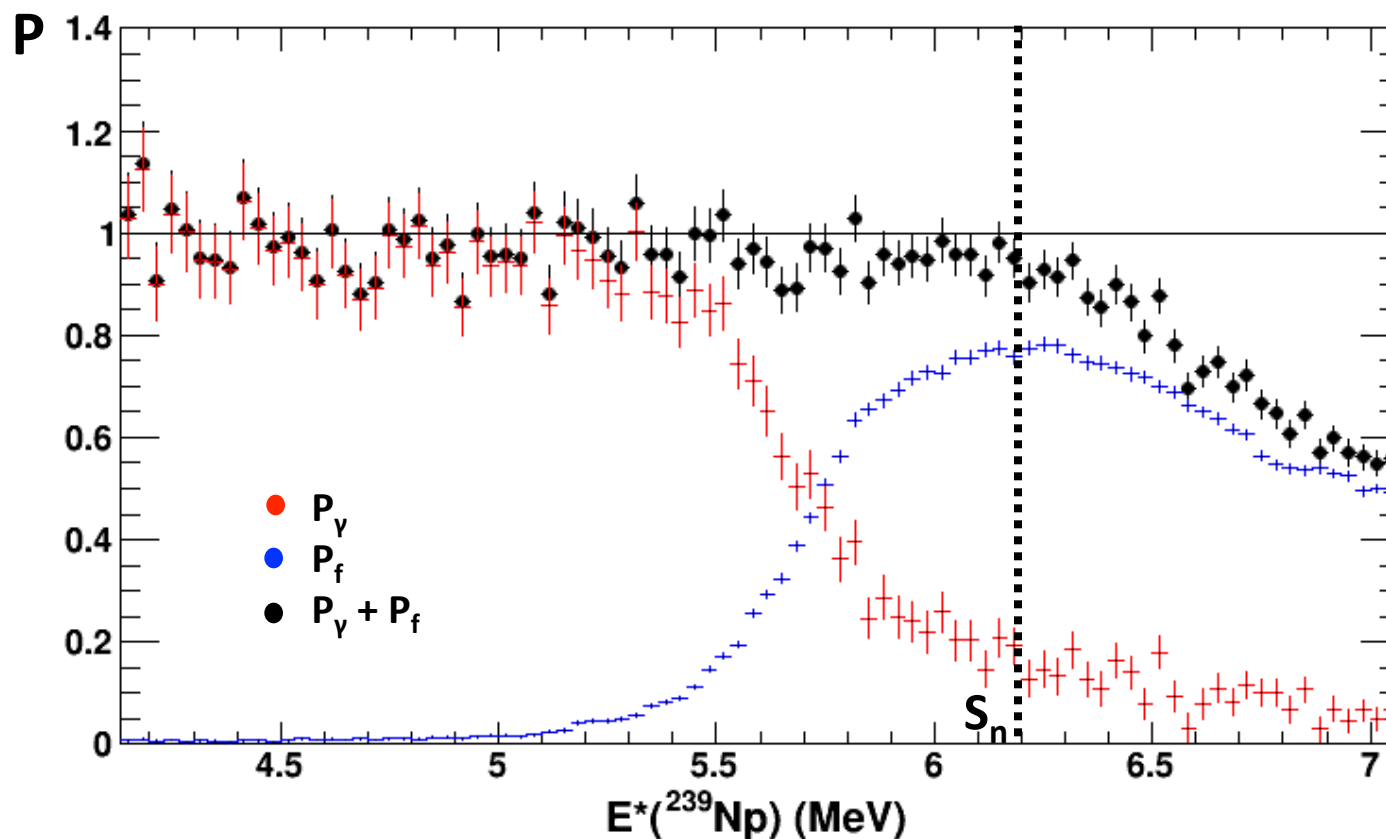
Challenge: subtraction of gamma rays emitted by the fission fragments !

Setup used for experiment at the Orsay tandem in April 2015



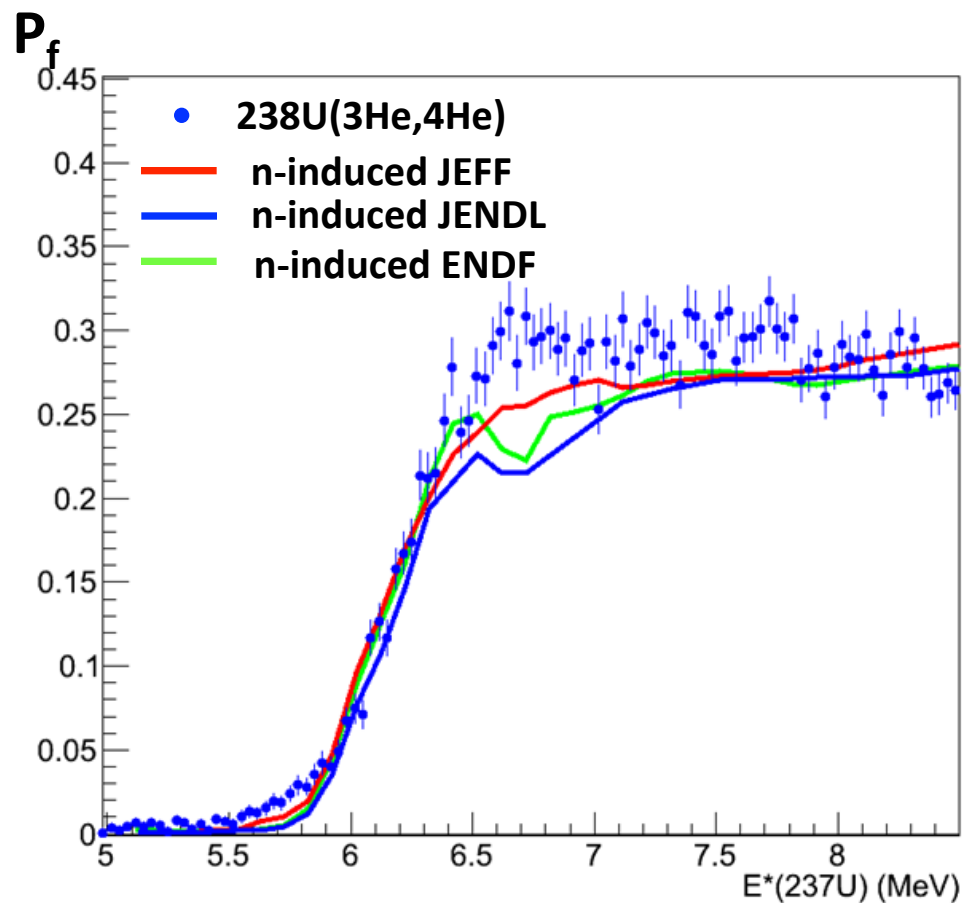
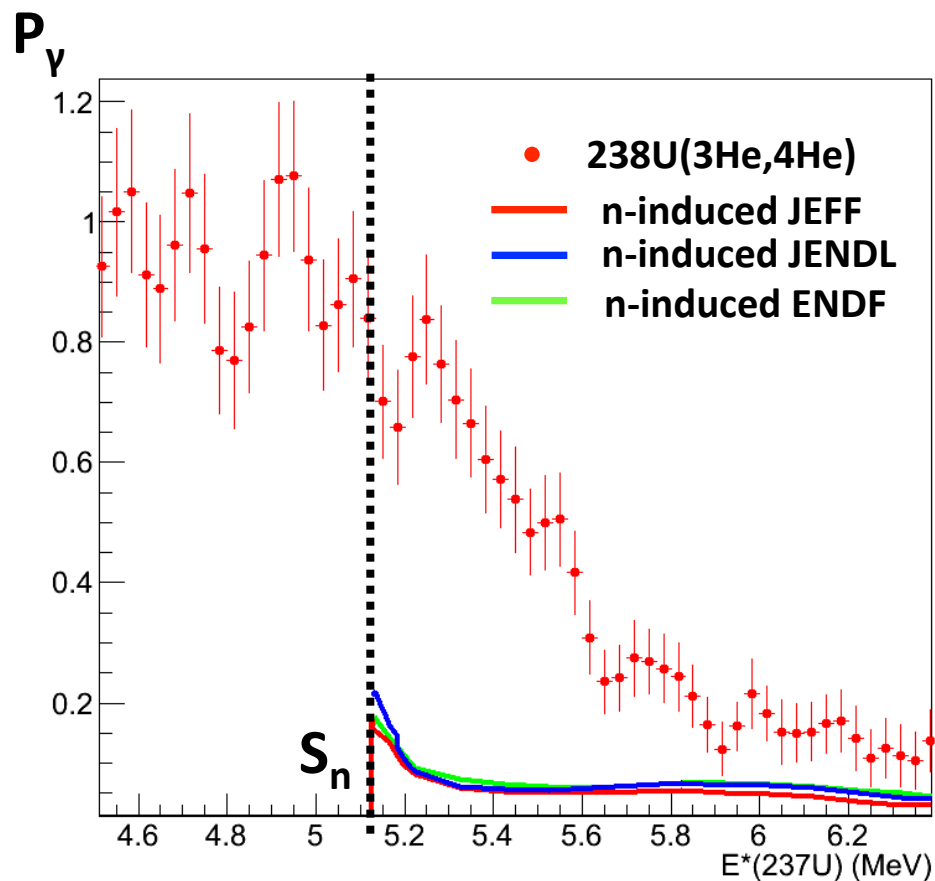
Funded by NEEDS!

Preliminary results!



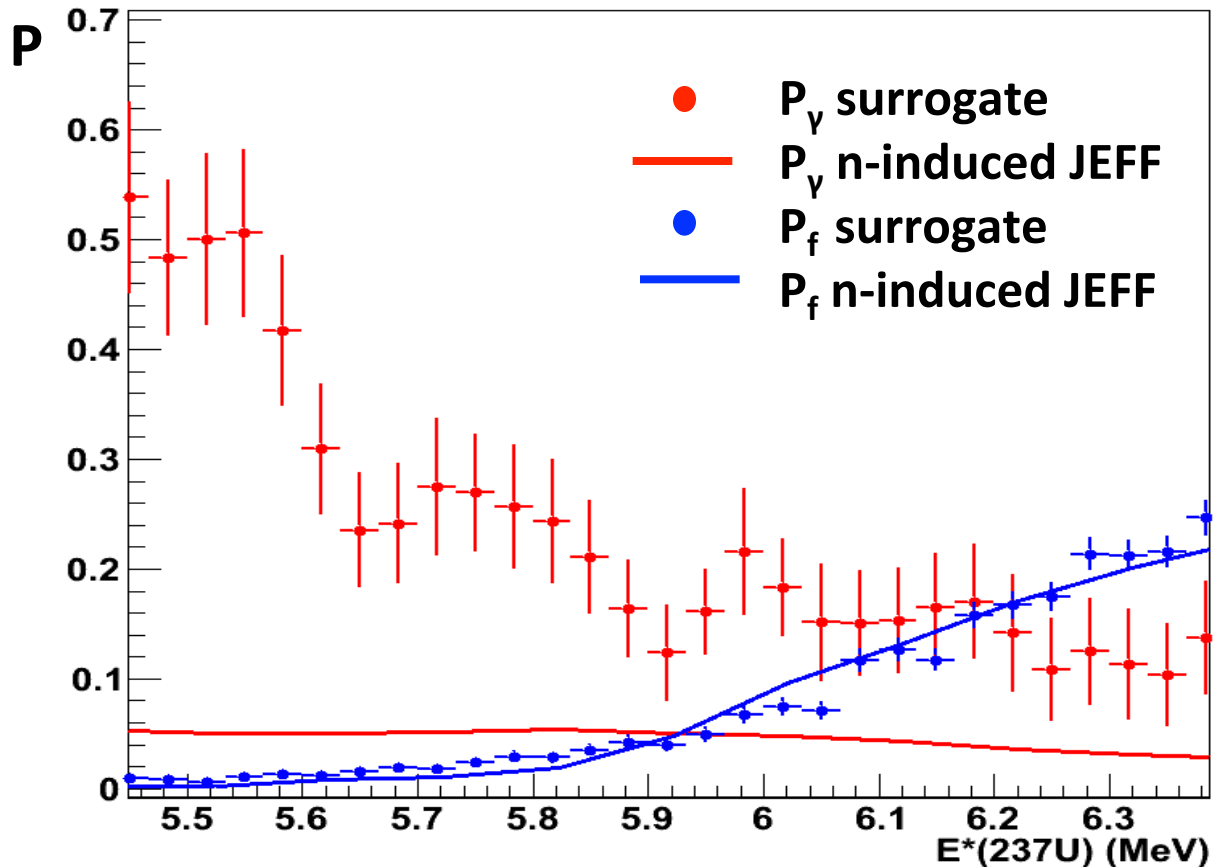
$P_f + P_\gamma = 1$ at $E^* < S_n$:
Validation of analysis procedure!

Preliminary results!



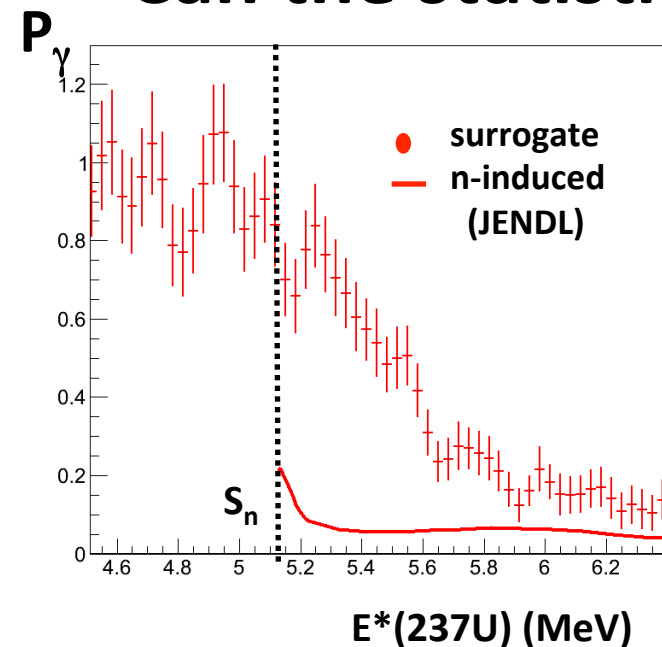
Focus on the overlap region

Preliminary results!



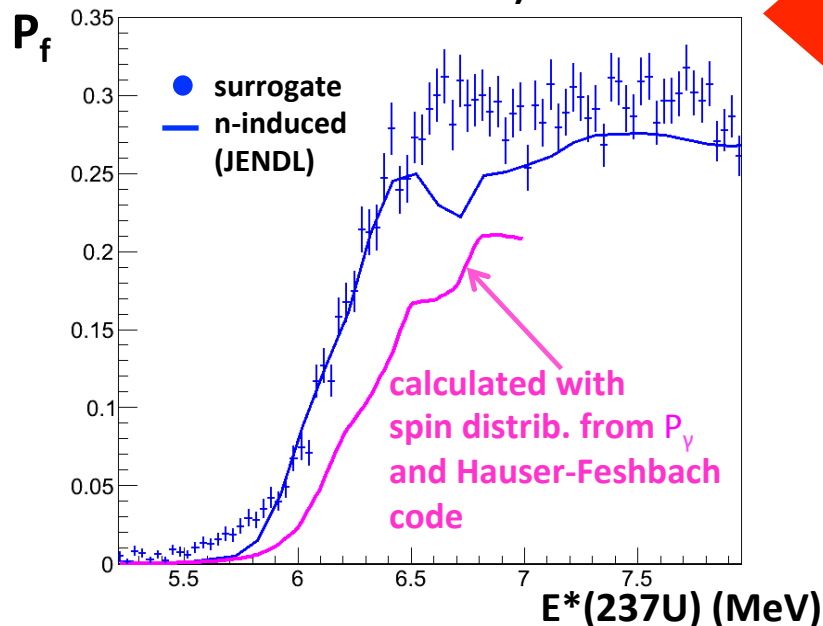
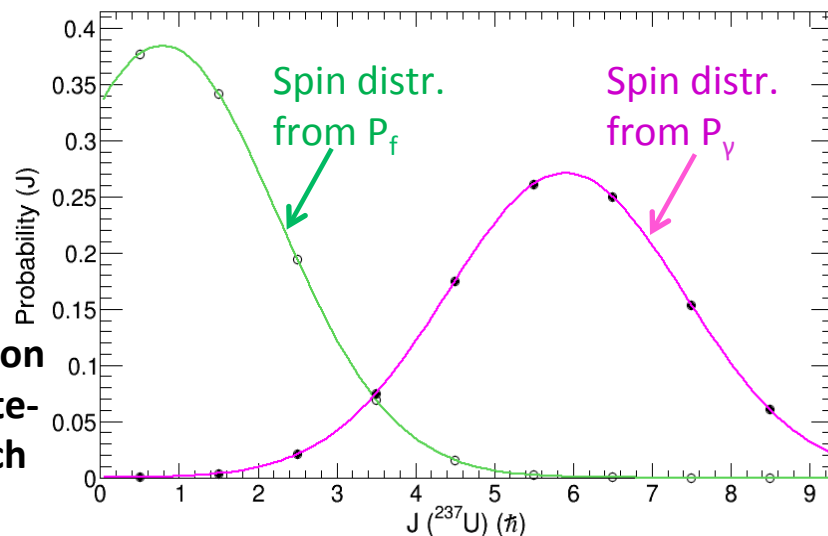
Fission is much less sensitive to the entrance channel than gamma-decay!

Can the statistical model explain these results?



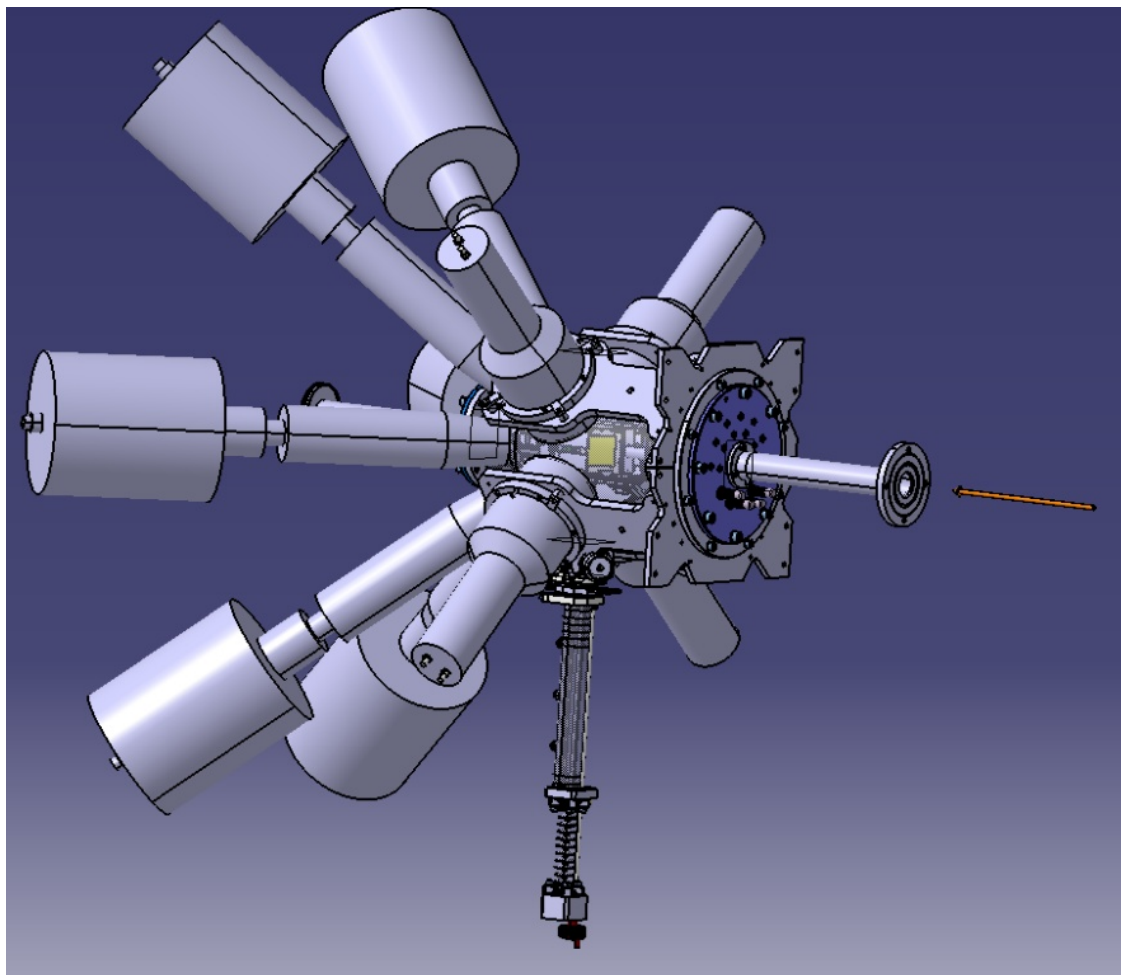
Deduce spin distribution
with the help of monte-
carlo Hauser Feshbach
code EVITA

(Based on TALYS and run
by CEA evaluator B.
Morillon)



The calculations
predict a strong
sensitivity of the
fission probability
to angular
momentum in
contradiction with
our results!

Perspectives



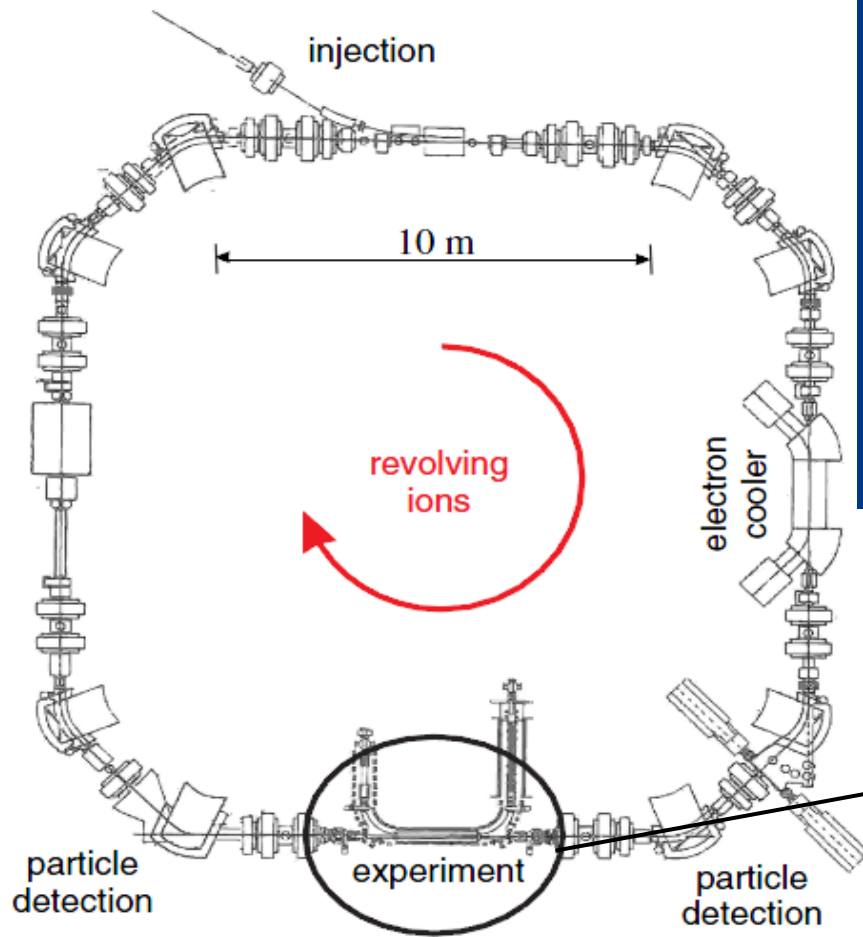
**${}^{240}\text{Pu}$: Even-even
fissioning nucleus
expected to be more
sensitive to spin/parity
differences!**

**Good-quality n-induced
data available!**

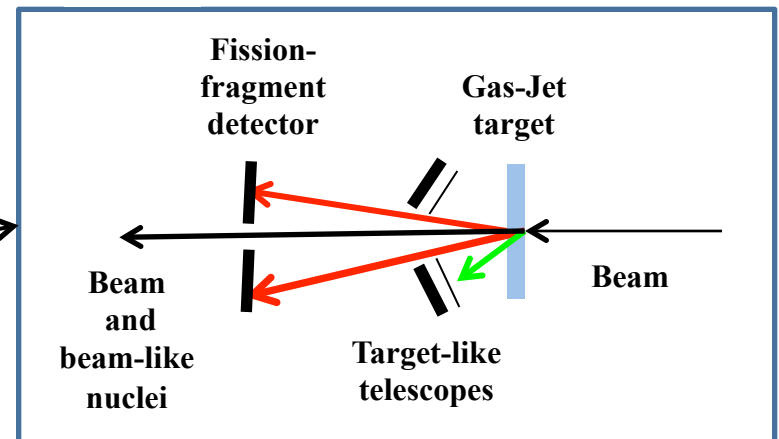
Still some uncertainties on the availability of the ${}^{240}\text{Pu}$ target!

Fission-probability measurements in inverse kinematics with radioactive ion beams

Fission-probability measurements inside a storage ring



- Beam-energy resolution of few hundreds keV, beam size 1 mm!!
- In-ring measurements with gas-jet targets (H₂, D₂, ³He, ⁴He):
Pure targets (no contaminants, no backing), limited straggling
- Increased intensity by 10^6 due to revolution frequency



**Systematic measurement of fission probabilities in other regions,
also of purified isomeric beams!!
Experiments at GSI and in the longer term HIE-ISOLDE (2023)**

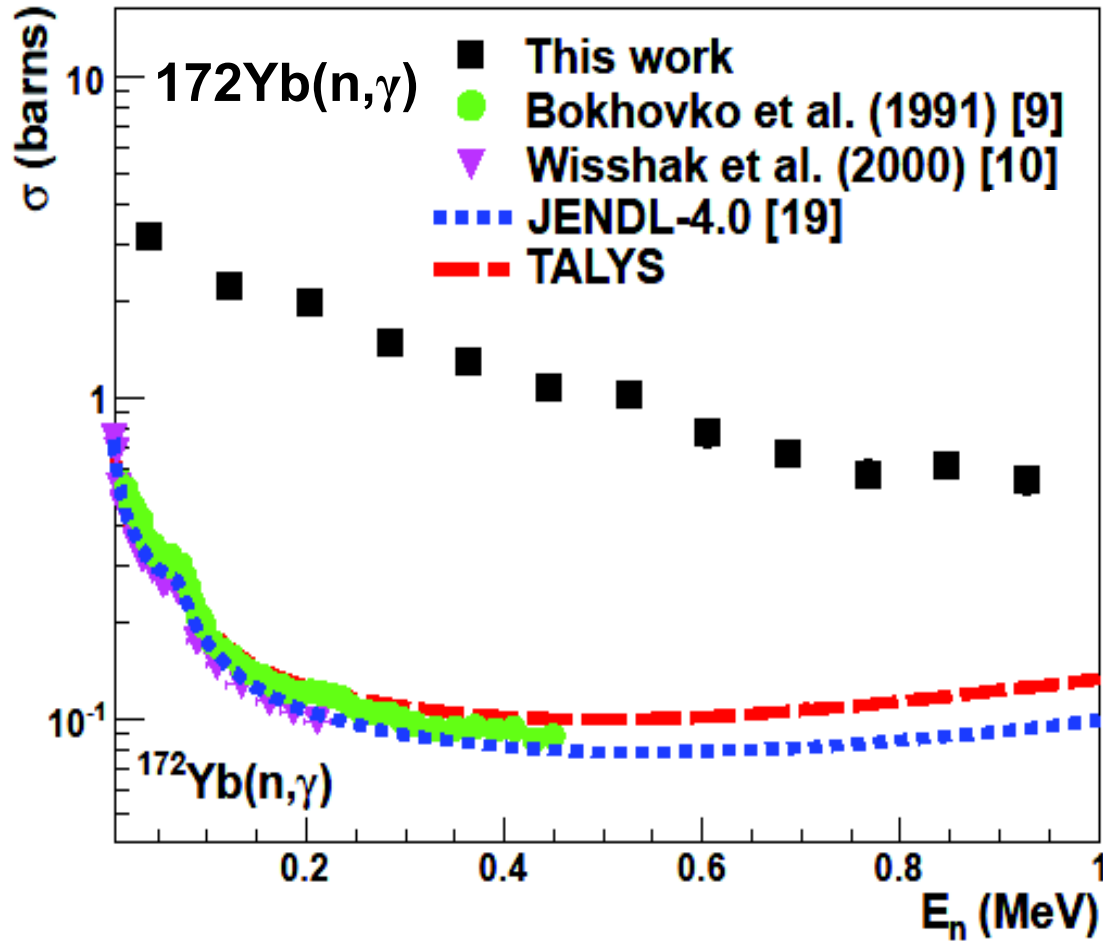
Conclusions...

- First simultaneous measurement of gamma-decay and fission probabilities for surrogate reactions
- The fission probability is much less sensitive than the gamma-decay probability to the differences in the entrance channel.
- This finding cannot be explained by statistical model calculations with standard ingredients.

... Perspectives

- Pursue theoretical efforts to understand our results
- Measurement P_f and P_γ for even-even ^{240}Pu
- Perform systematic studies in other regions of the chart of nuclei, isomeric beams
- ➔ Experiments in inverse kinematics inside storage rings with the RIBs of GSI and HIE-ISOLDE

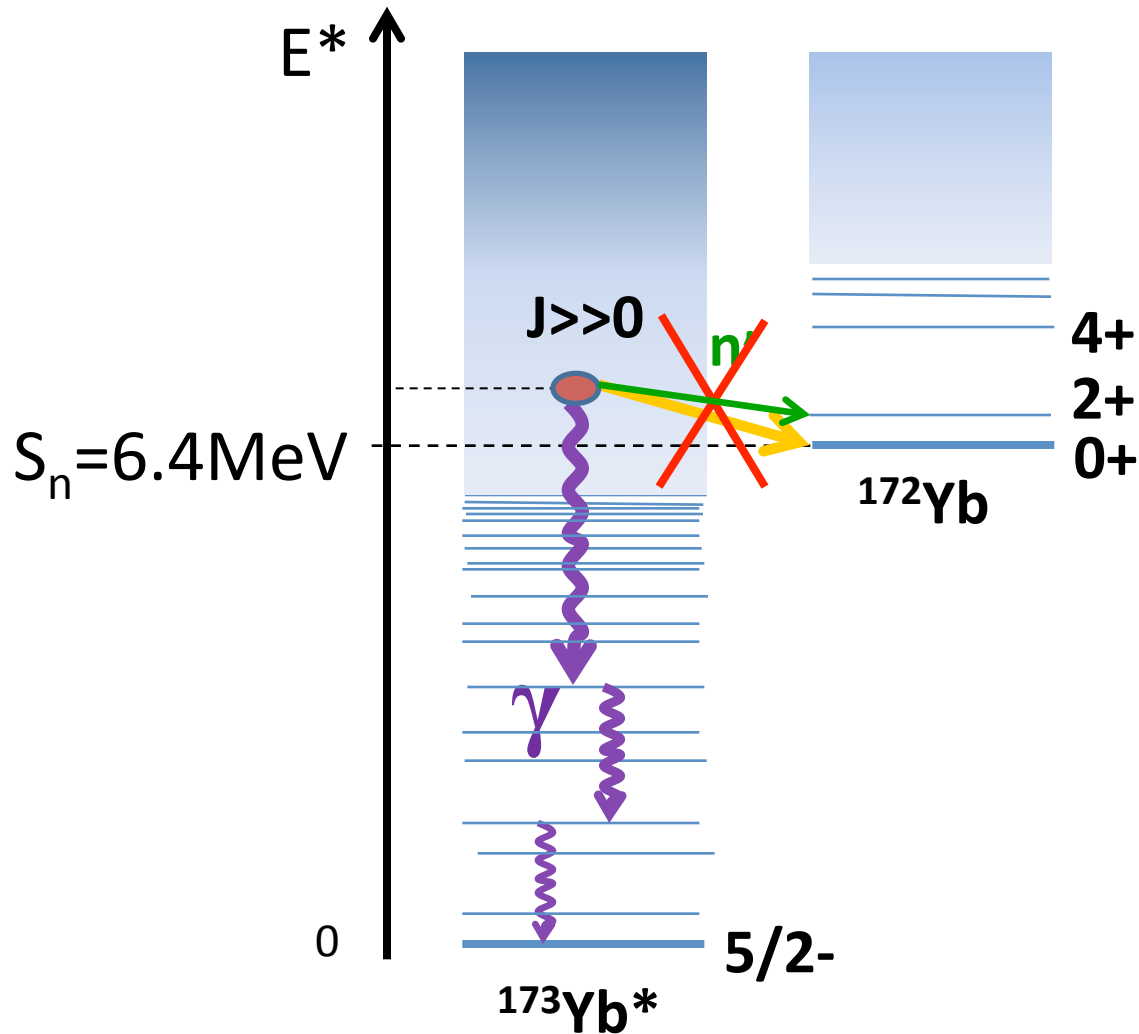
Results for radiative capture



The cross sections obtained with surrogate method are in clear disagreement with n-induced data for capture!

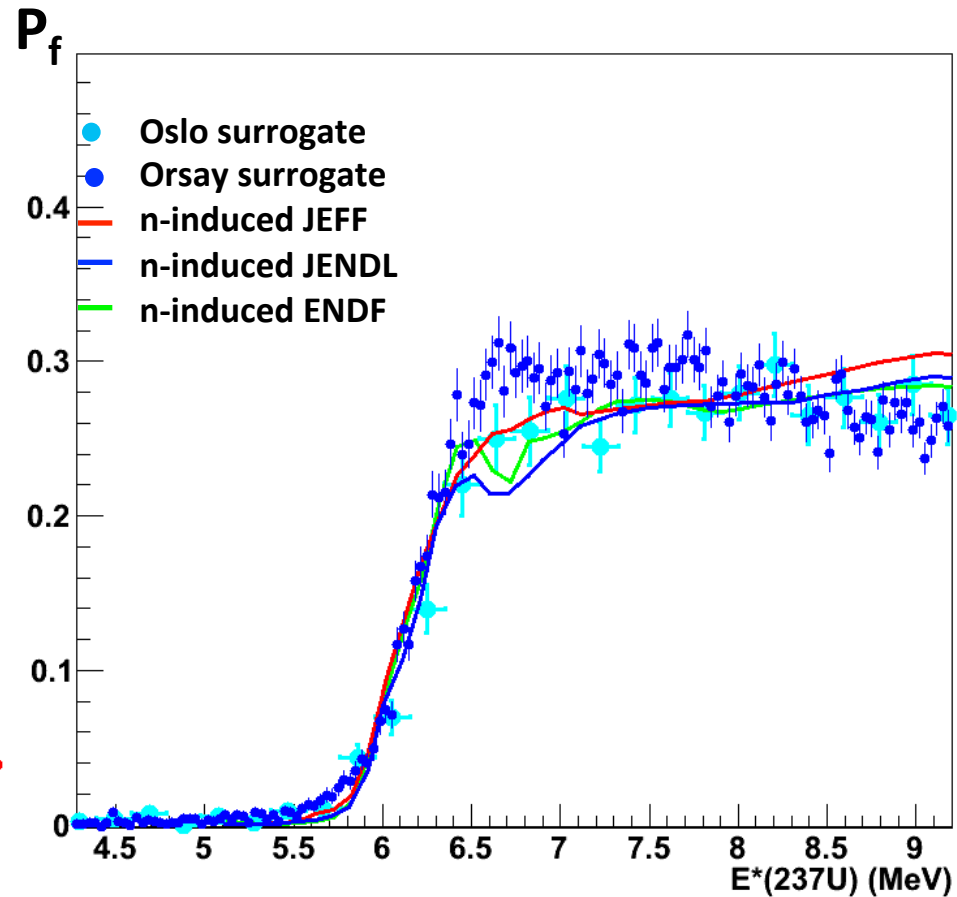
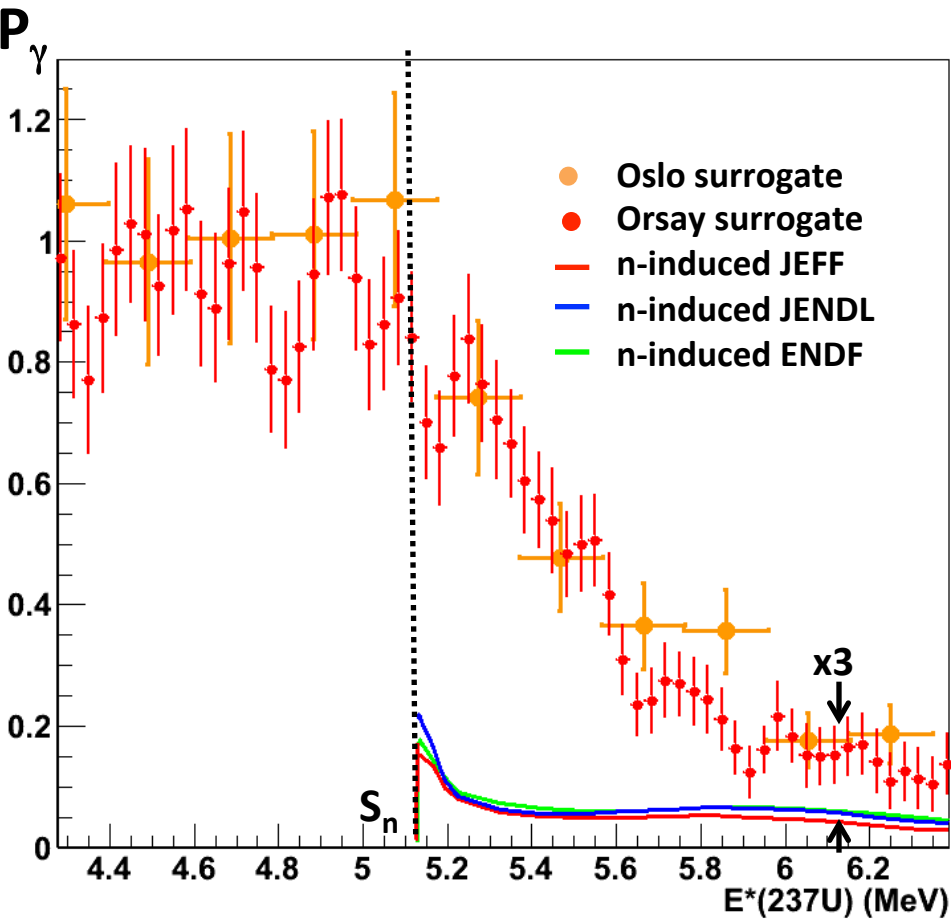
G. Boutoux et al., Phys. Lett. B 712 (2012) 319

Why do we obtain such discrepancies?



Strong sensitivity of neutron emission to $J\pi$

Preliminary results!



Transfer-induced reactions in inverse kinematics with radioactive ion beams

HIE-ISOLDE, actinide and pre-actinide beams:

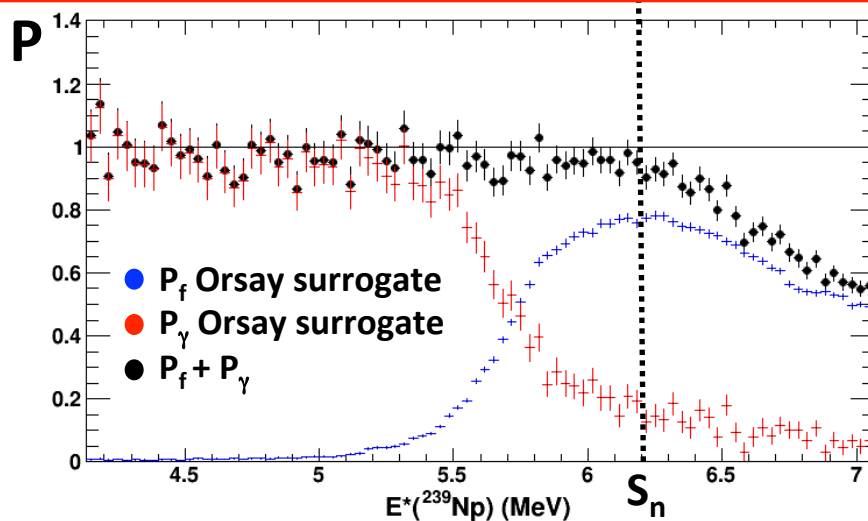
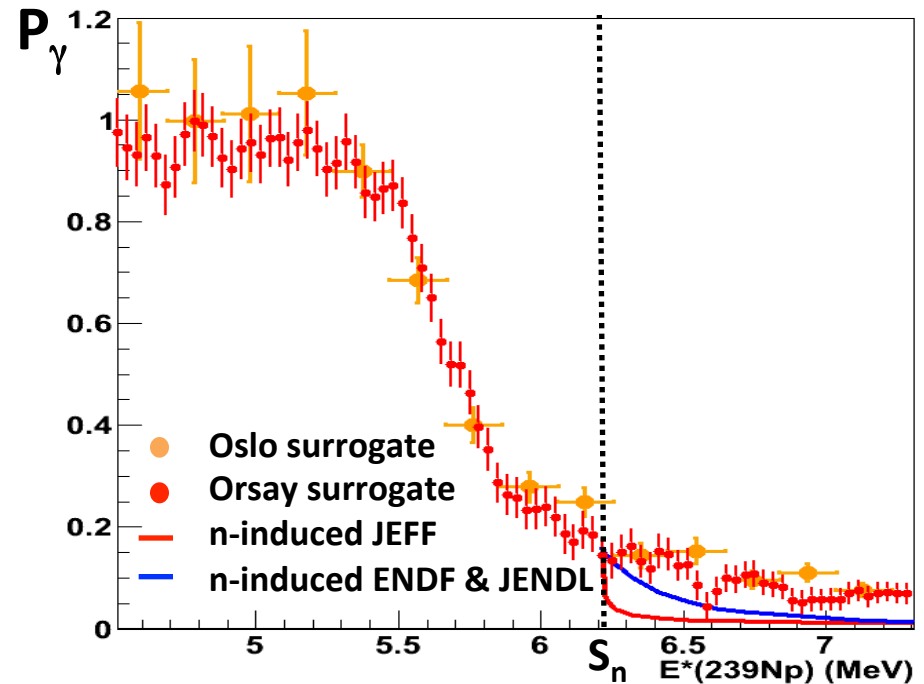
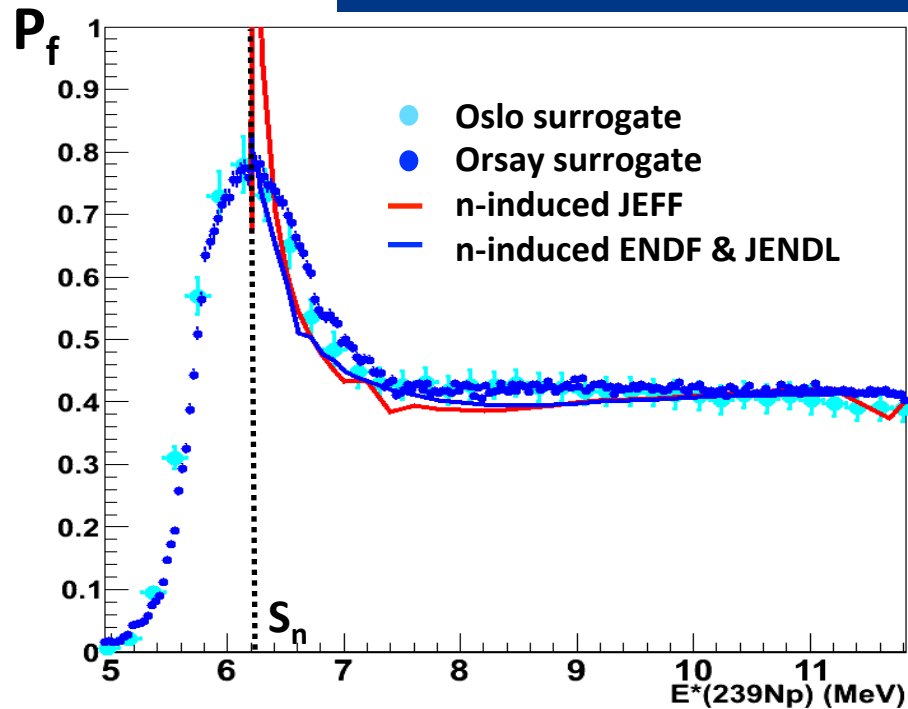
Element	Isotopic chain	Half lifes
Rn (Z=86)	$^{204-212}\text{Rn}$ $^{219-221}\text{Rn}$	$2.4\text{h} \leq T_{1/2} \leq 28.5\text{min}$ $3.96\text{s} \leq T_{1/2} \leq 25\text{min}$
Fr (Z=87)	$^{207-213}\text{Fr}$ $^{220-228}\text{Fr}$	$14.8\text{s} \leq T_{1/2} \leq 20\text{min}$ $27.4\text{s} \leq T_{1/2} \leq 21.8\text{min}$
Ra (Z=88)	$^{221-222,224-226,228}\text{Ra}$	$28\text{s} \leq T_{1/2} \leq 1600\text{y}$

10 A MeV and high intensities

BUT...

Limited beam quality: dispersion in energy > 2MeV and size > few mm

Preliminary results!



$P_f + P_\gamma = 1$ at $E^* < S_n$:
validation
of analysis procedure!!

Validity of the surrogate method: limiting cases

Neutron-induced decay probability

$$P_{neutron,decay}(E^*) = \sum_{J^\pi} P_{neutron}^{form}(E^*, J^\pi) \cdot G_{decay}(E^*, J^\pi)$$

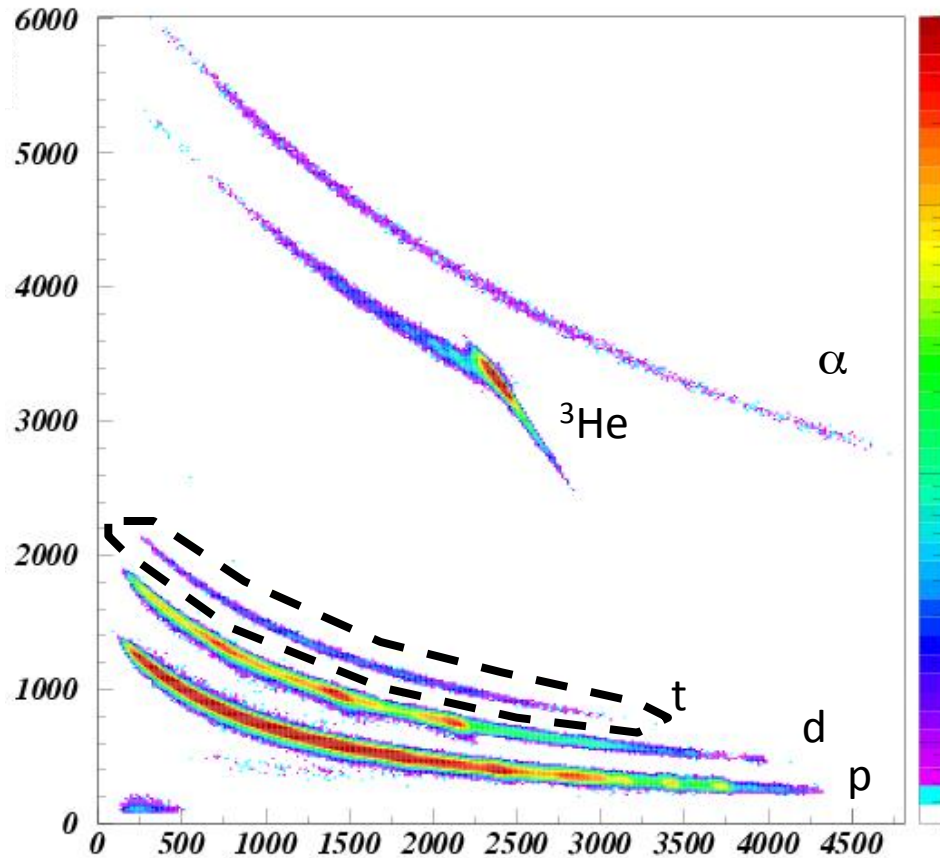
Surrogate decay probability

$$P_{surro,decay}(E^*) = \sum_{J^\pi} P_{surro}^{form}(E^*, J^\pi) \cdot G_{decay}(E^*, J^\pi)$$

$P_{n,decay}(E^*) = P_{surro,decay}(E^*)$ if $P_{neutron}^{form}(E^*, J^\pi) = P_{surro}^{form}(E^*, J^\pi)$ or $G_{decay}(E^*, J^\pi) = G_{decay}(E^*)$

Fission probability

ΔE [Ch]

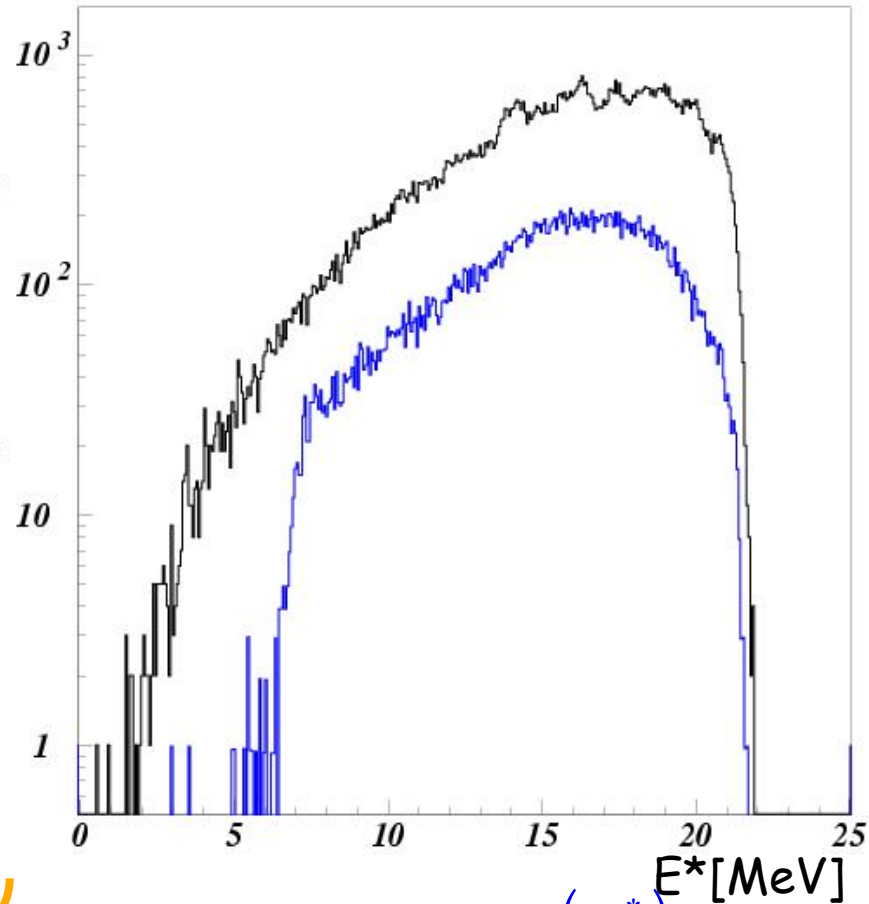


Residual energy [Ch]

Light-particle kinematics + Q-values
 E^* of the CN

Counts

Tritons

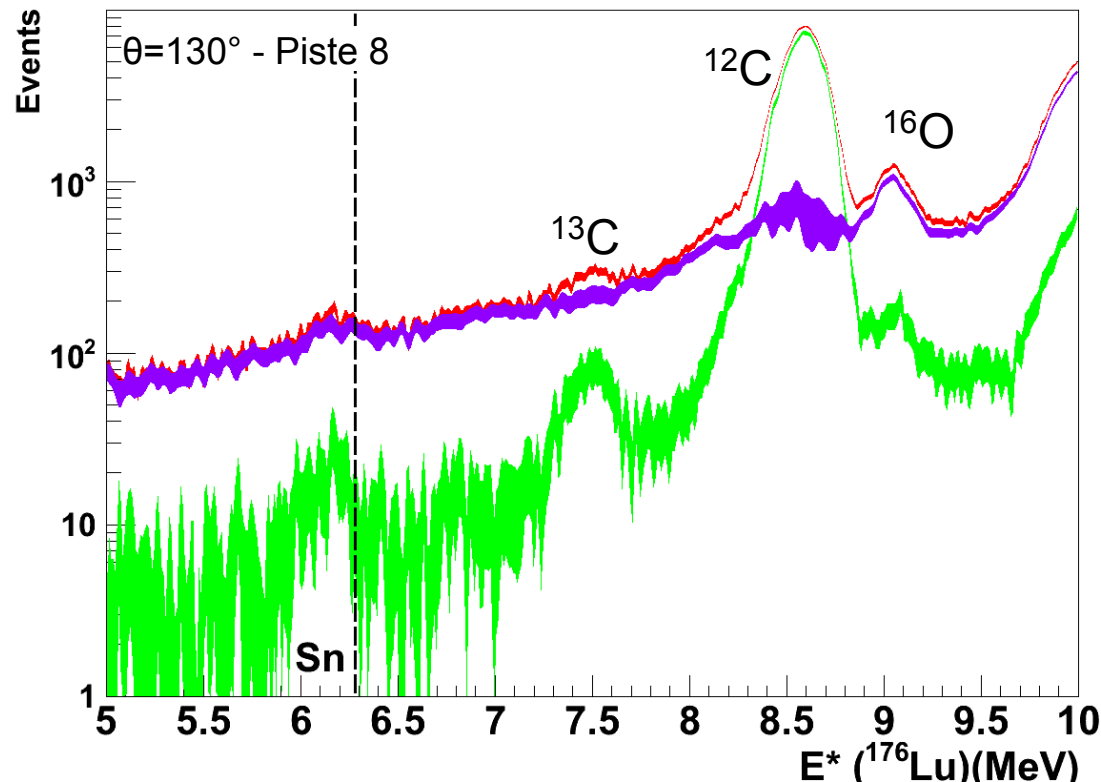


$$P_f(E^*) = \frac{N_{\text{coin}}(E^*)}{N_{\text{CN}}(E^*) \cdot \text{Eff}(E^*)}$$

$$E^* = \frac{A}{A+1} E_n + B_n$$

Soustraction des contaminants:

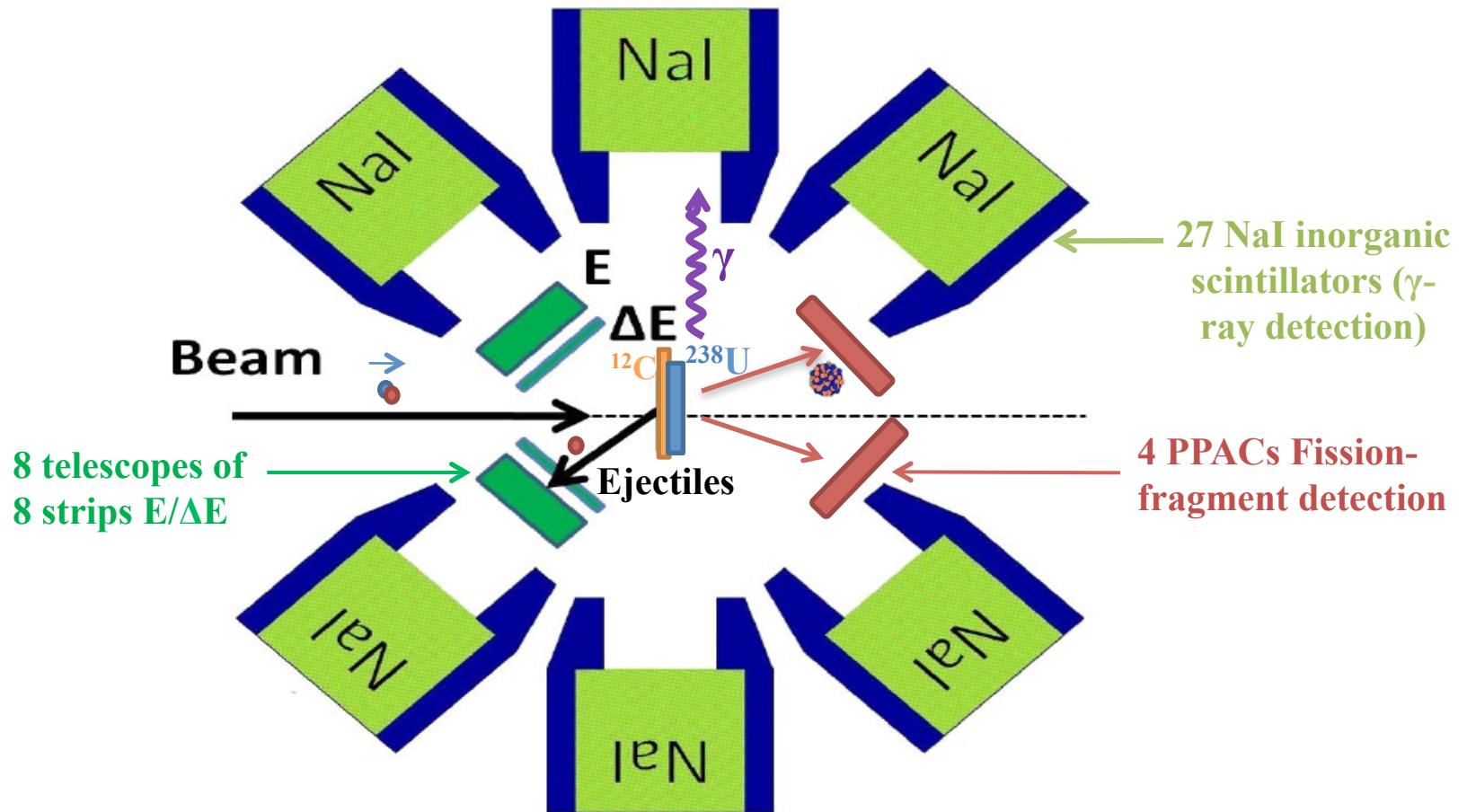
- Réactions contaminantes avec le support en C et les impuretés (O).
- Les éjectiles émis dans ces réactions donne une mauvaise identification du noyau excité!



$$N_{SINGLES} = N_{SINGLES}^{brut} - N_{SINGLES}^{support\ C}$$

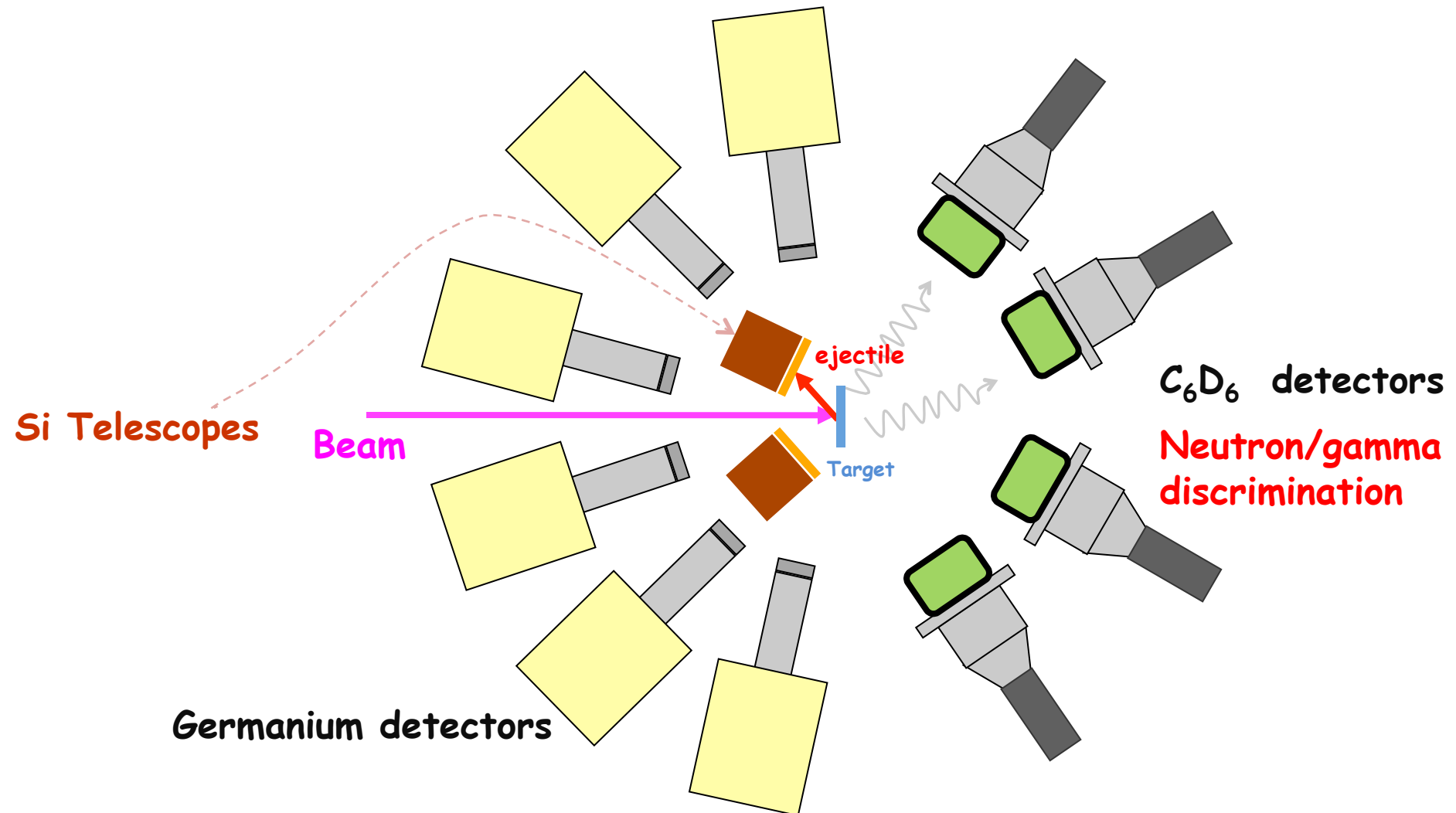
- Rejet systématique des zones contaminées par ^{12}C et ^{16}O .
- Bonne soustraction du ^{13}C dans la zone d'intérêt ($S_n < E^* < S_n + 1 \text{ MeV}$)

Experimental set-up at the Oslo cyclotron



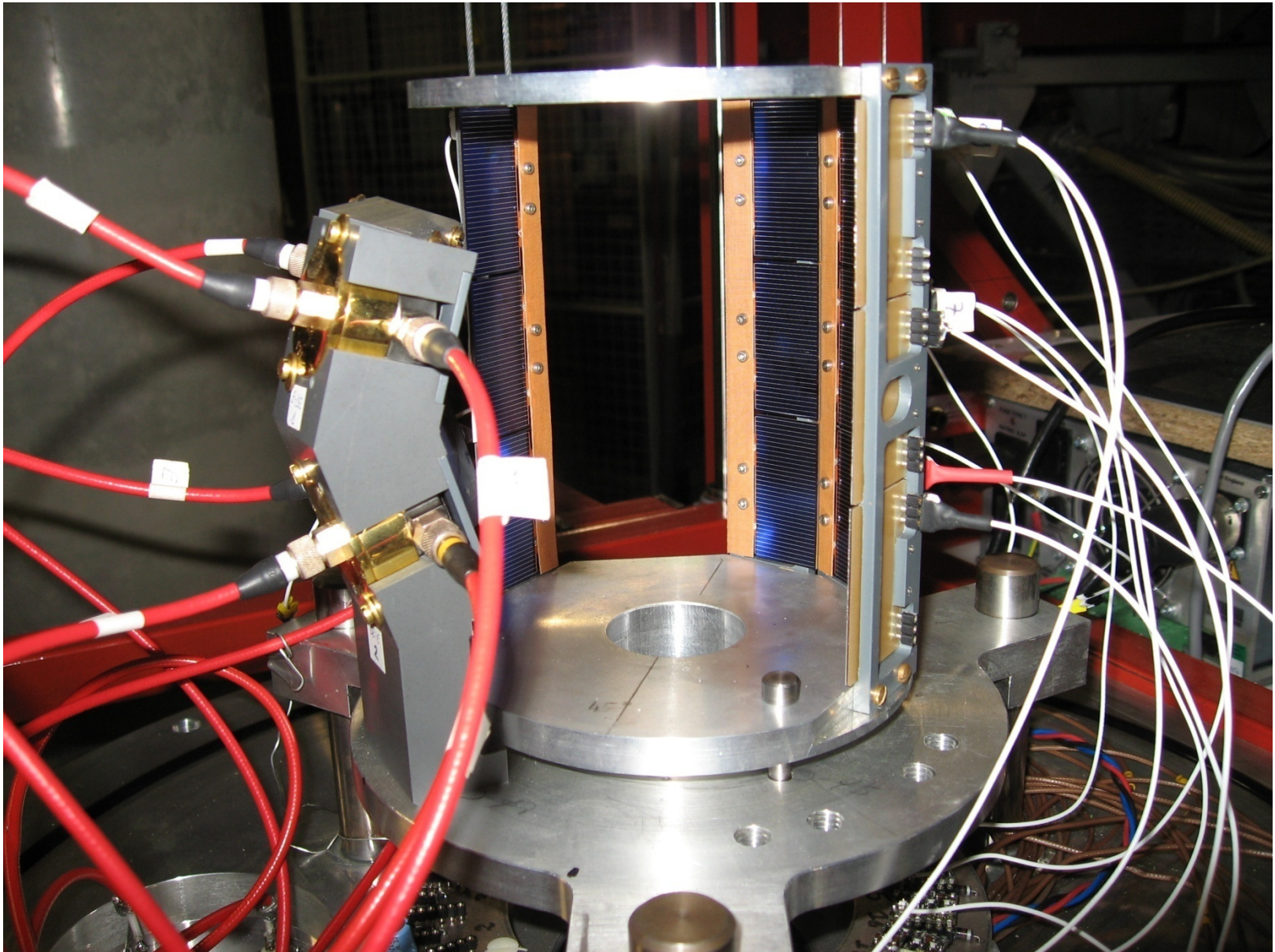
Simultaneous measurement of fission and gamma-decay probabilities!

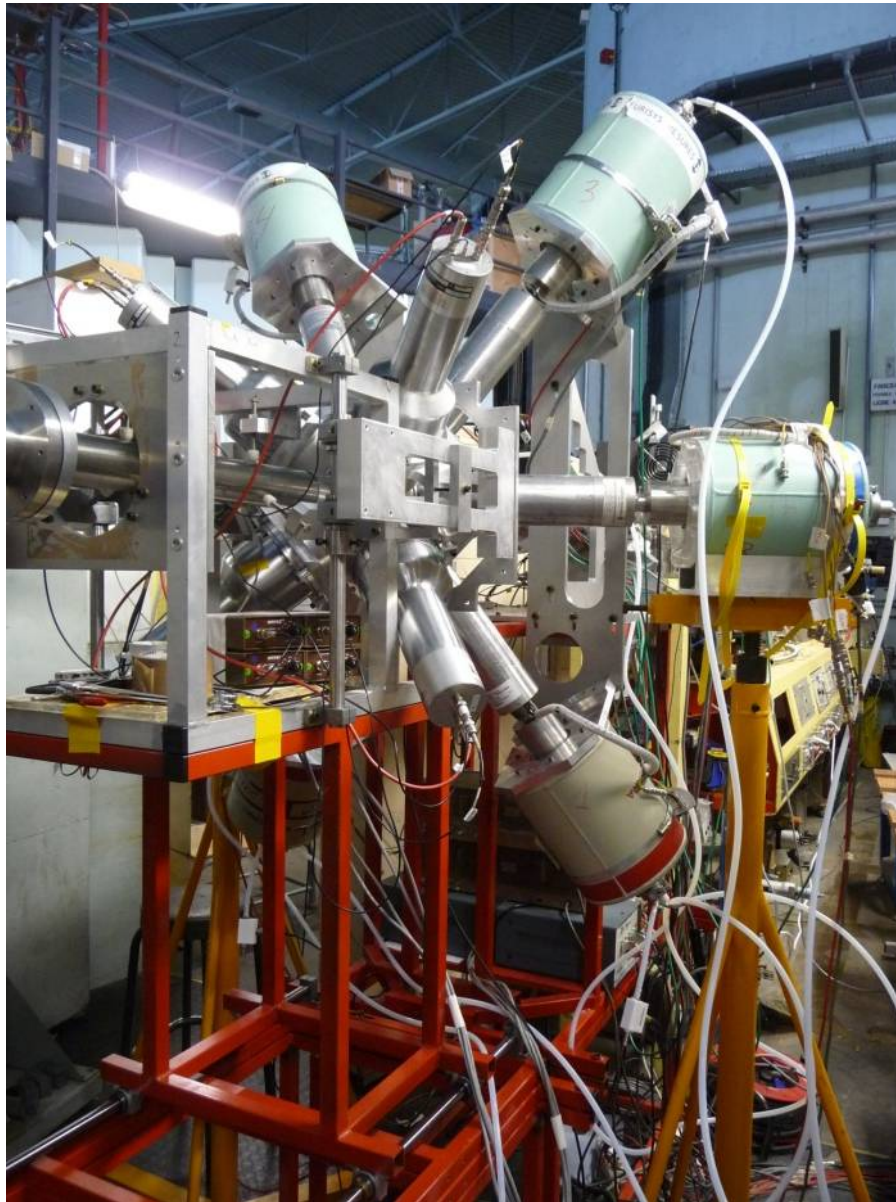
Experimental set-up for gamma-decay probability measurements at IPN Orsay



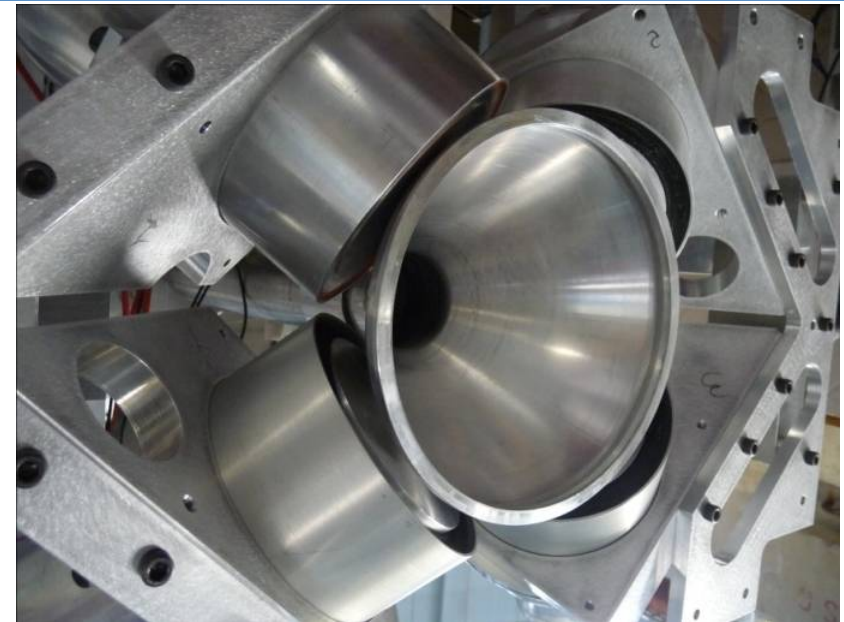
Ge --> verify the gamma-decay probabilities measured with the scintillators!
No gamma-ejectile coincidences coming from contaminants, from nucleus A-1!

Experimental set-up for fission

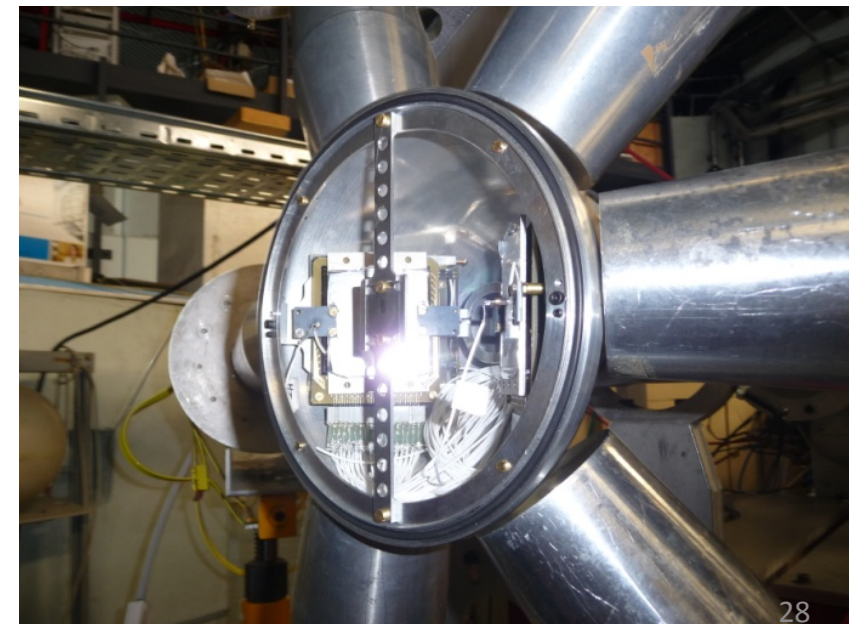




**Dispositif expérimental
@ TANDEM (IPN Orsay, FRANCE)**



Scintillateurs C6D6 x4



Détecteurs Germanium x6

Difficulties in the prediction of spin-parity distributions in surrogate reactions

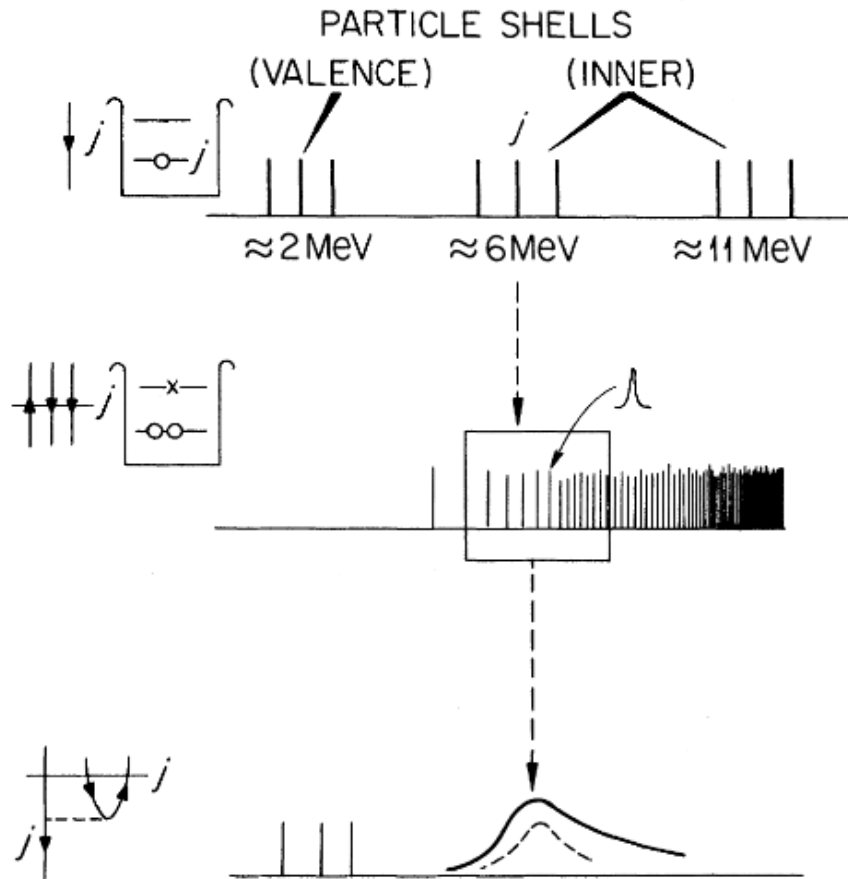


FIG. 4. Schematic illustration of the damping or spreading of the inner shell hole strength (top of figure) into the 2h-1p "sea" of levels (middle of figure) by the inelastic scattering of the hole state (bottom of figure).

Population of single particle states by the surrogate reactions.

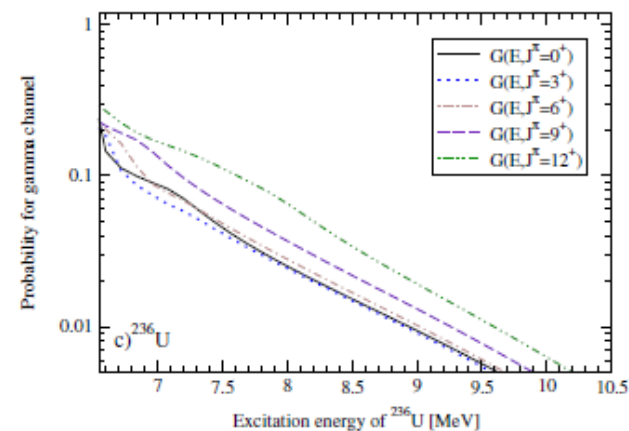
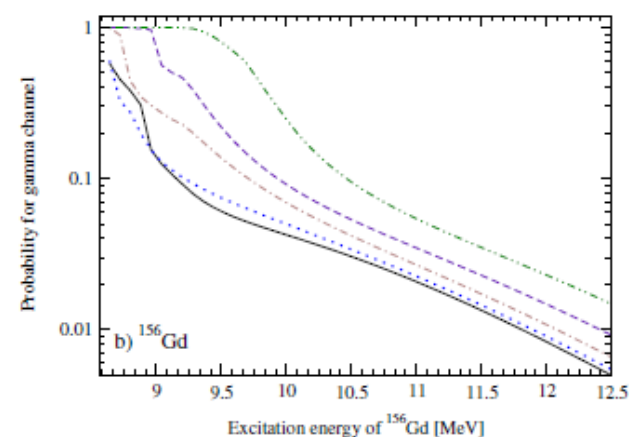
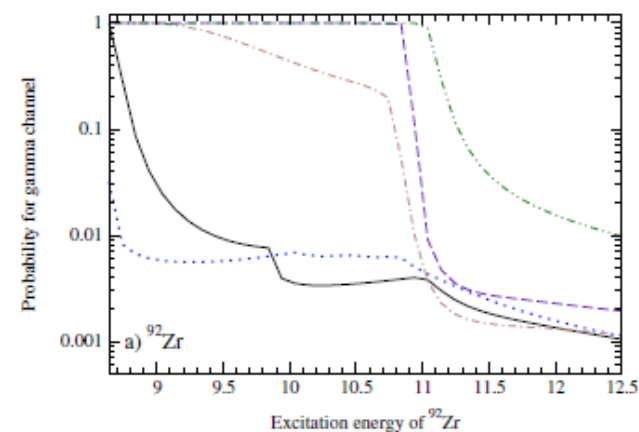
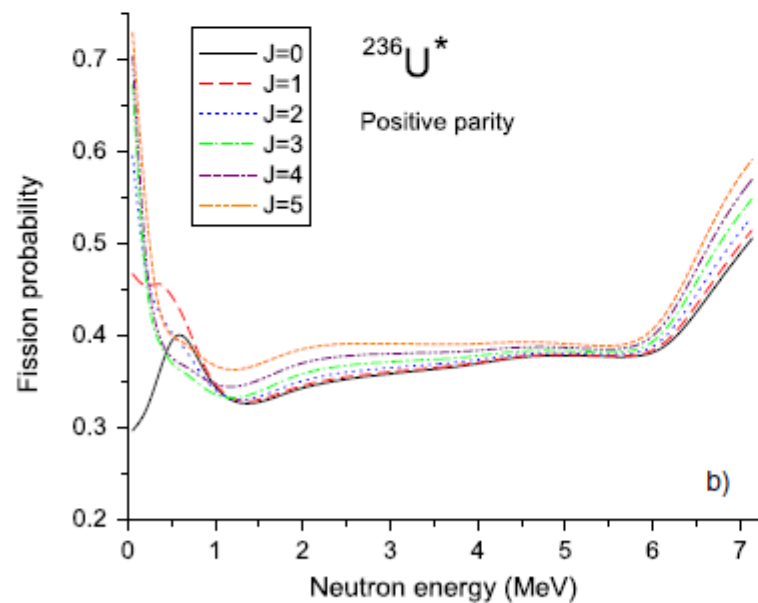
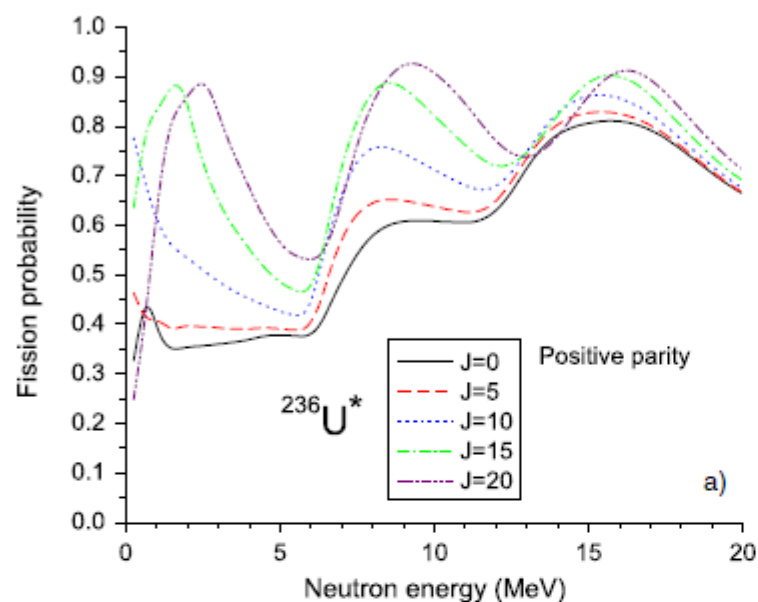
Structure information on unbound states in the continuum needed!

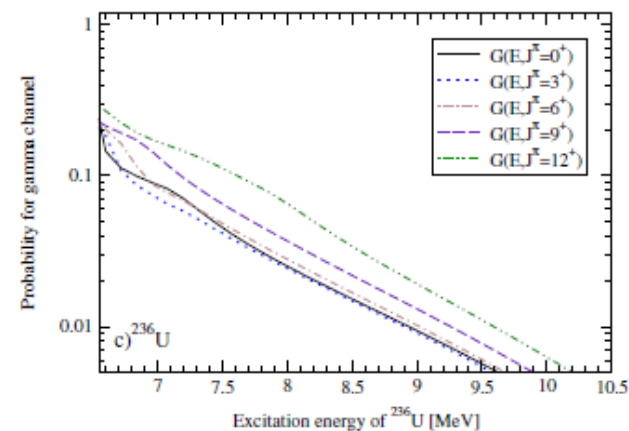
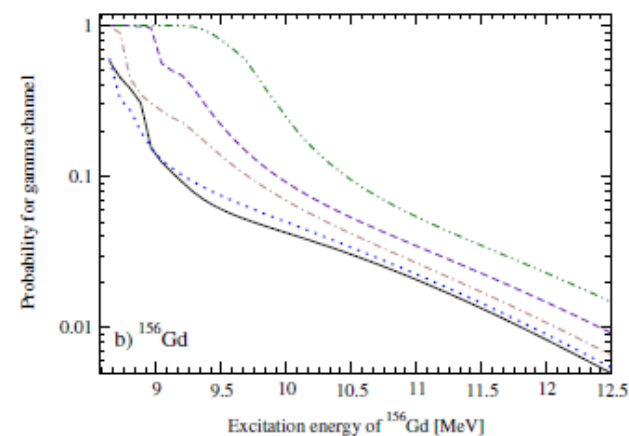
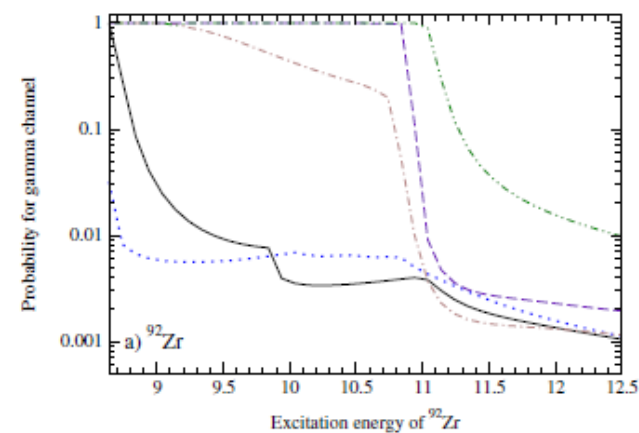
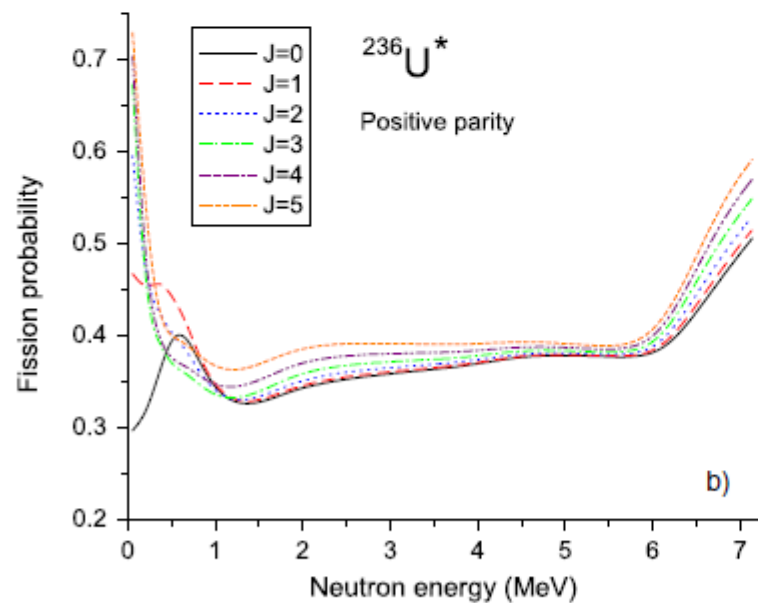
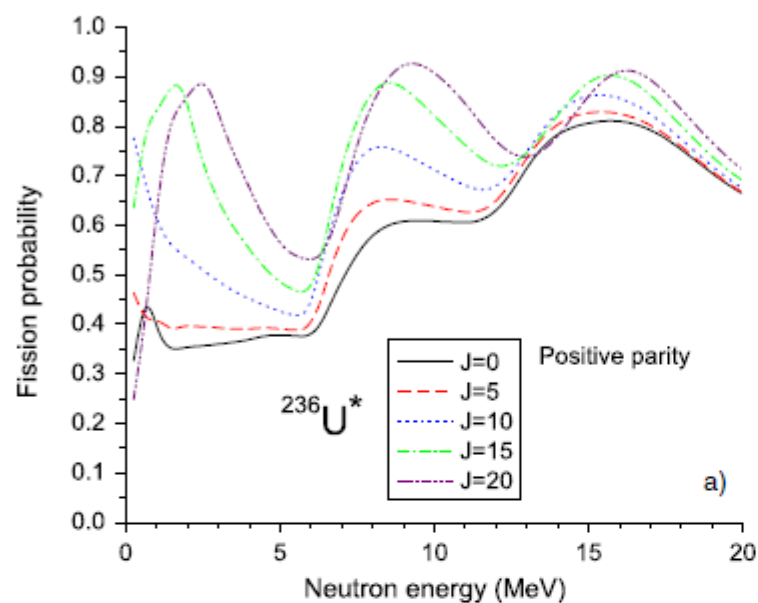
Residual interactions make these states decay into the other (many!) available states at the same energy

The initial single particle strength is fragmented!

Calculation very difficult!

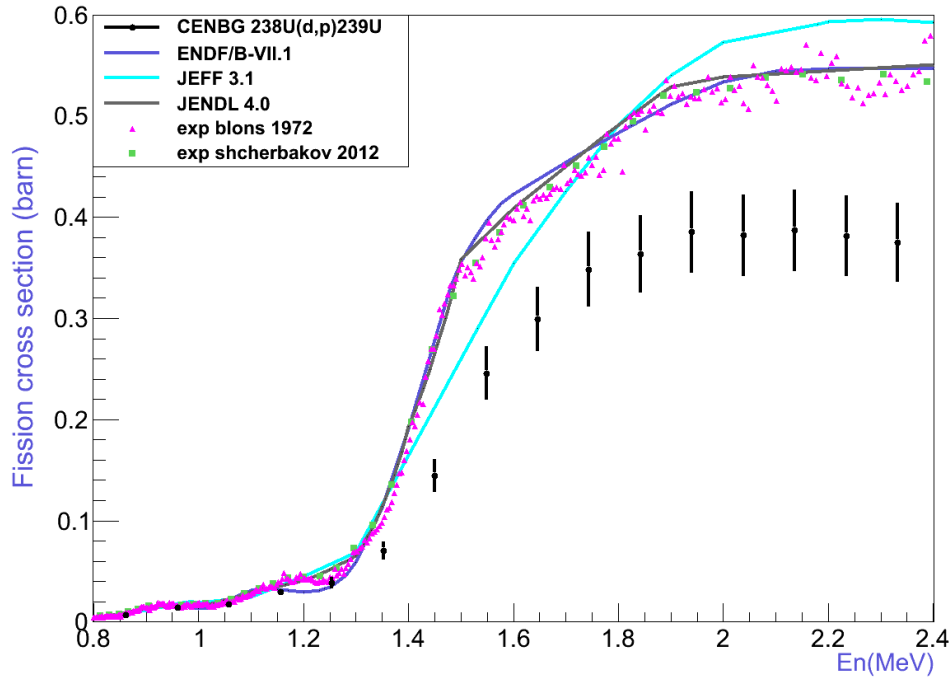
In addition one has to calculate the probability that this damping leads to the formation of a compound state.



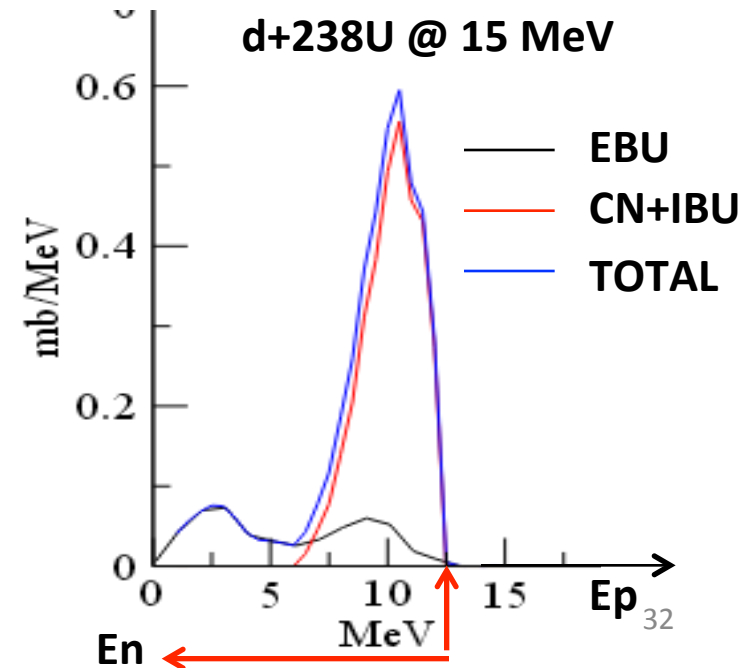
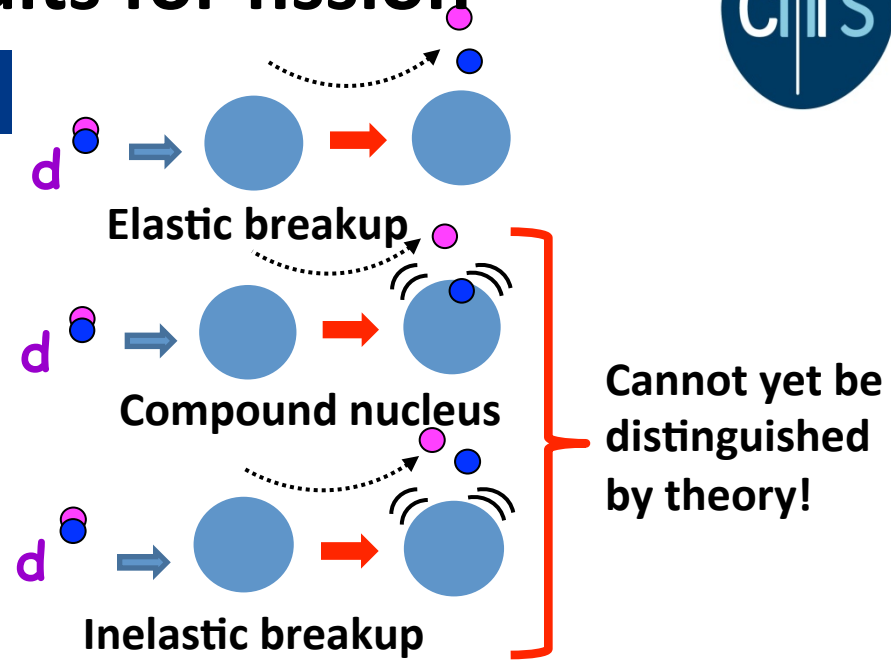


Preliminary results for fission

$^{238}\text{U}(d,p)^{239}\text{U} \leftrightarrow ^{238}\text{U}(n,f)$



Cross section lower by 30% at $E_n > 2$ MeV



Up to 10% contribution from elastic breakup!
How much inelastic breakup??

CDCC (Continuum discretized coupled-channel calculation)

Antonio Moro and Jin Lei, Univ. of Sevilla, Spain

Excitation-energy sharing between CN and ejectile

**^3He , t, d, p have no
bound excited state.
 ^4He first excited state
at 20 MeV**



**When we detect these ejectiles they are in
the ground state**

**For $E_{\text{proj}} \sim 24$ MeV
and detecting
ejectiles at backward
angles**



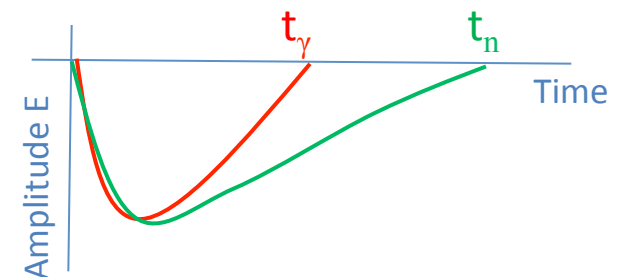
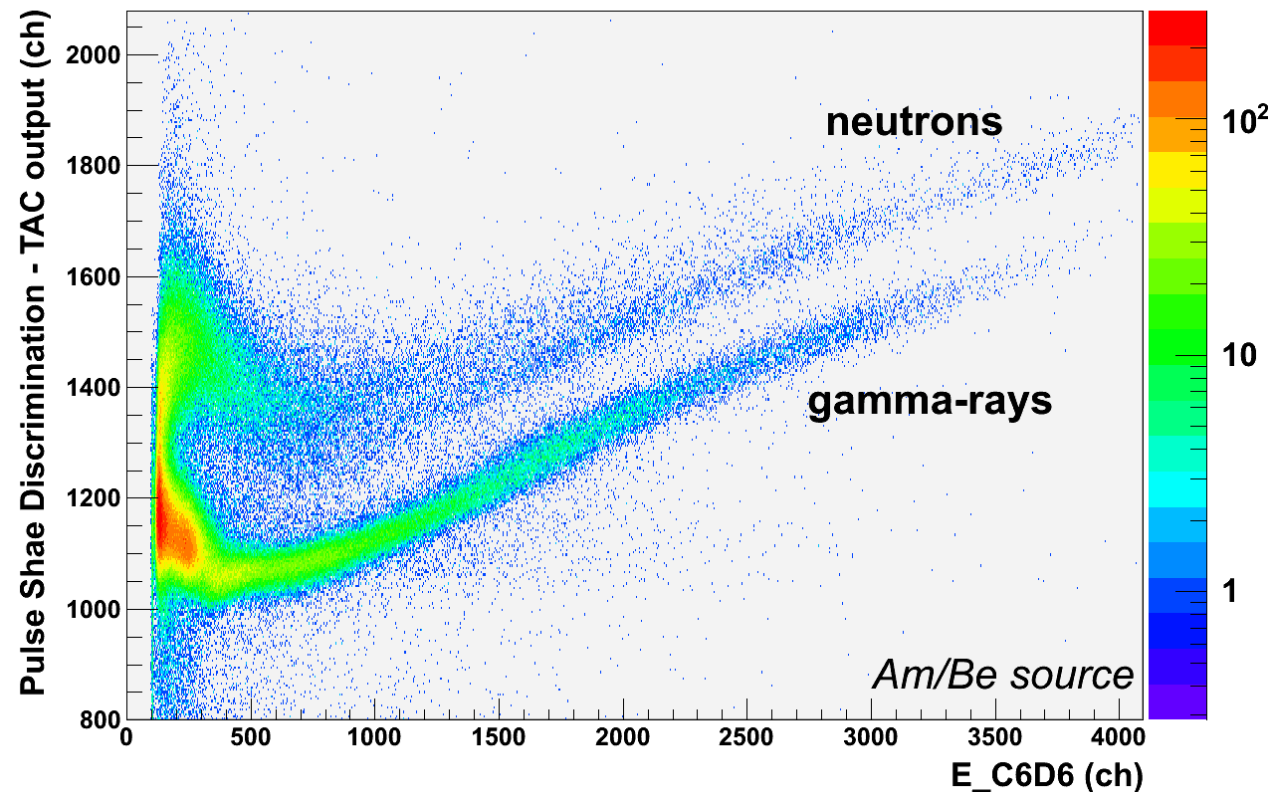
**No pollution of ($^3\text{He},d$) channel observed.
Demonstrated by Gavron et al. by comparing
fission probabilities of the same compound
nucleus formed in ($^3\text{He},t$) and ($^3\text{He},d$)
reactions.**

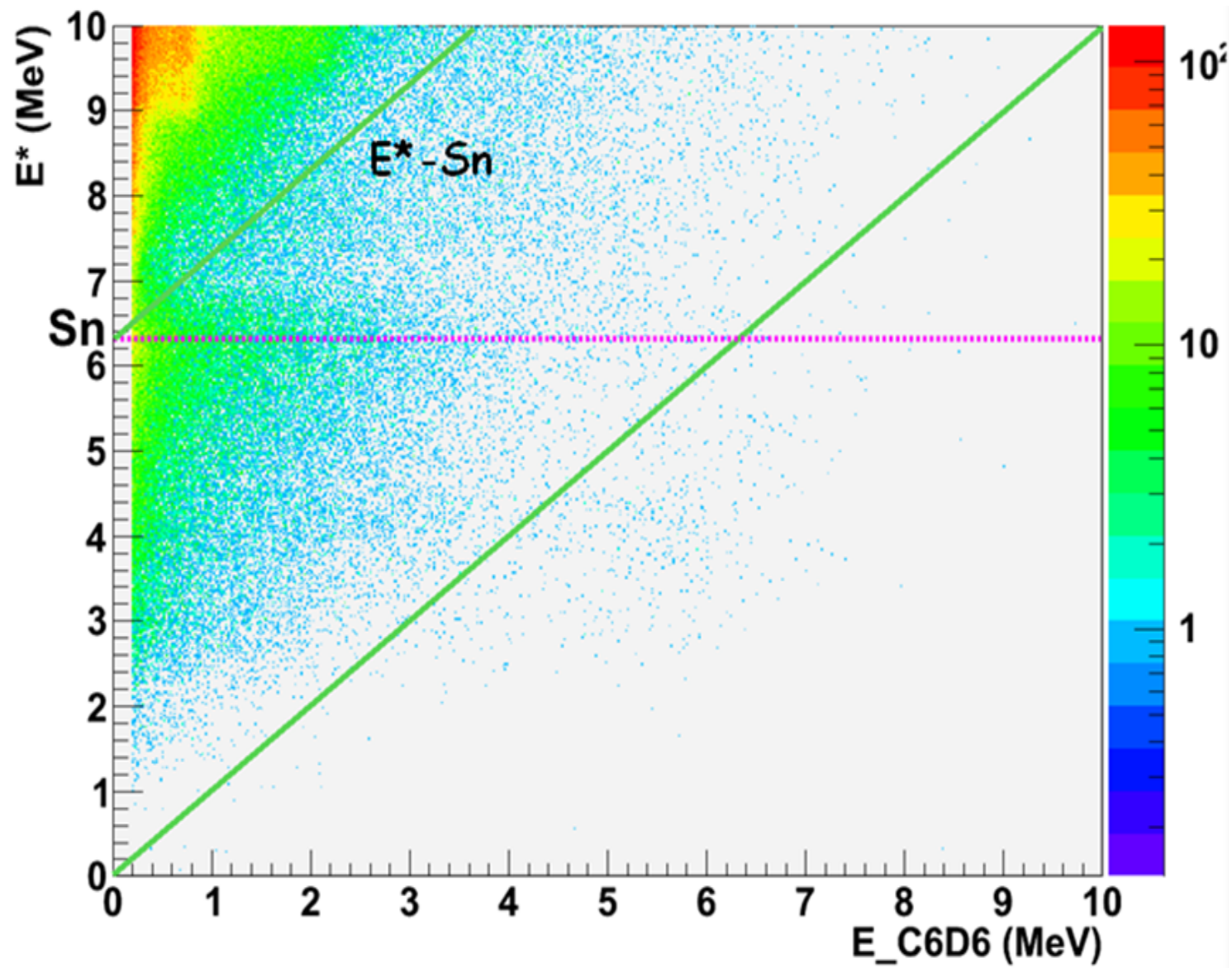
→ Neutrons thermiques:
D minimize les captures neutronique
au sein même du scintillateur

$$P_{\gamma}(E^*) = \frac{N_{p-\gamma}(E^*)}{N_p(E^*) \cdot \varepsilon_{\gamma}(E^*)}$$

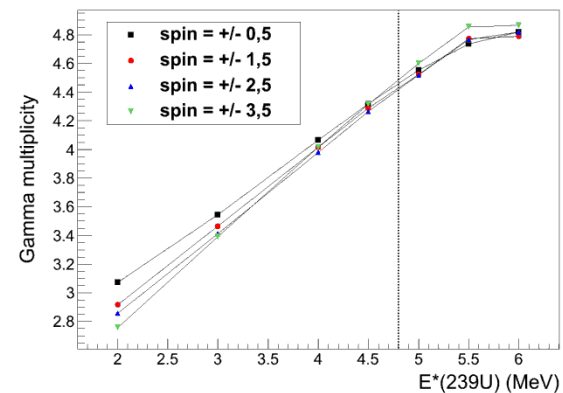
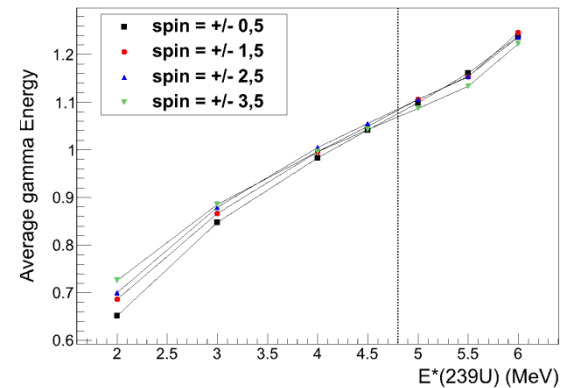
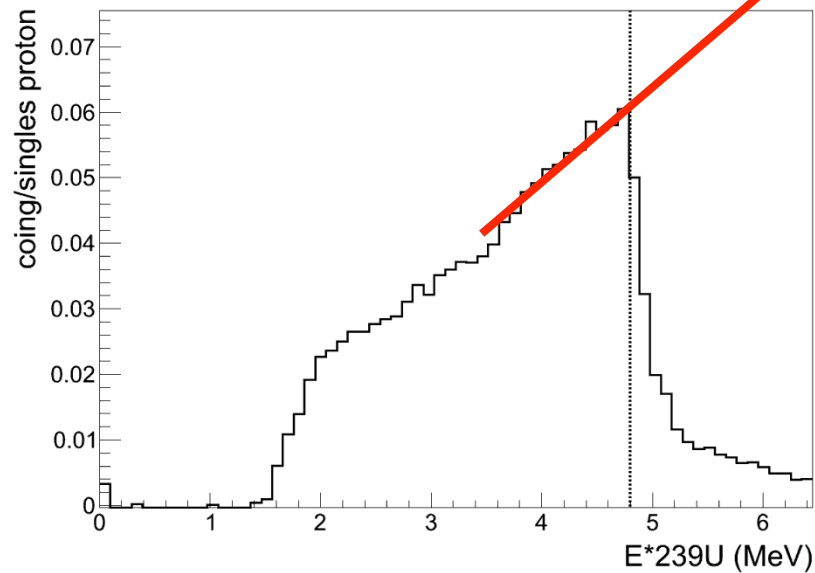
Identifier la particule qui
interagit dans le
scintillateur
→ **Discrimination n-γ**

→ Neutrons rapides:
Séparation par analyse de
forme (temps de
relaxation du signal)





EXtrapolation Efficiency Method (EXEM)



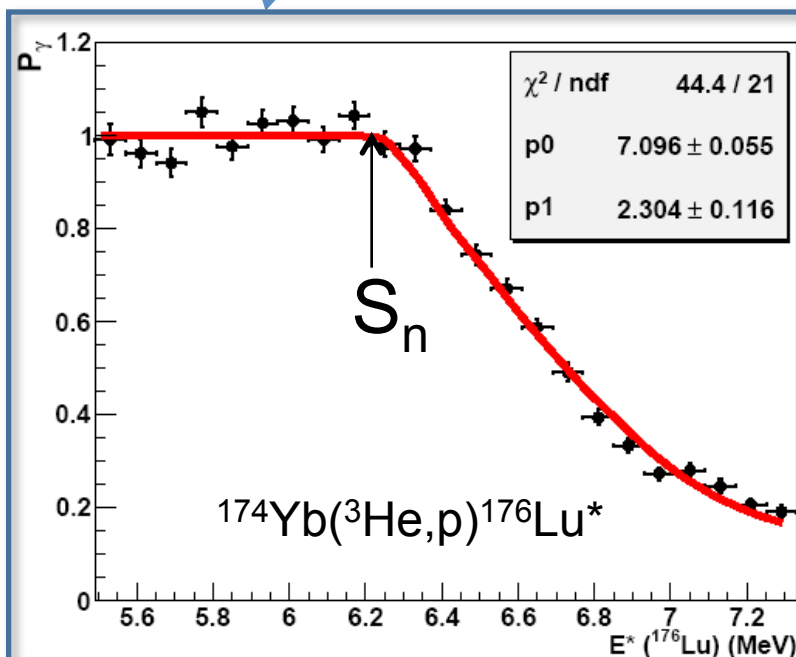
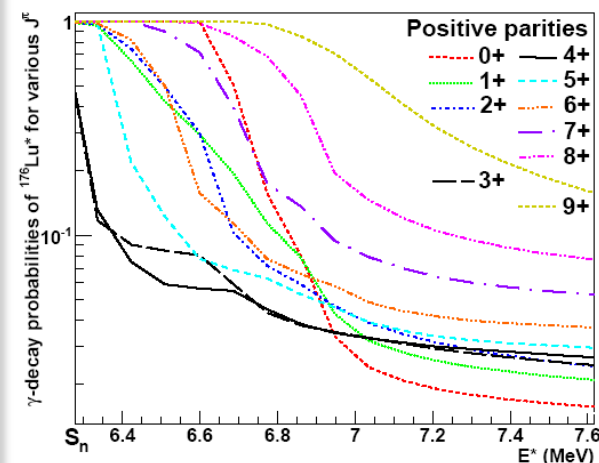
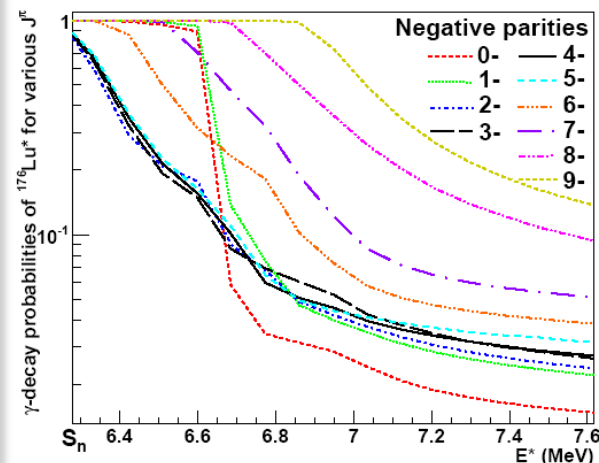
$$P_{\chi}(E^*) = \sum_{J^{\pi}} F_s^{CN}(E^*, J^{\pi}) \cdot G_{\chi}(E^*, J^{\pi})$$

FIT

Distribution de spin modélisée
par une gaussienne
indépendante de l'E*

$$\frac{0.5}{\sqrt{2\pi}\sigma} e^{-\frac{(J-\bar{J})^2}{2\sigma^2}}$$

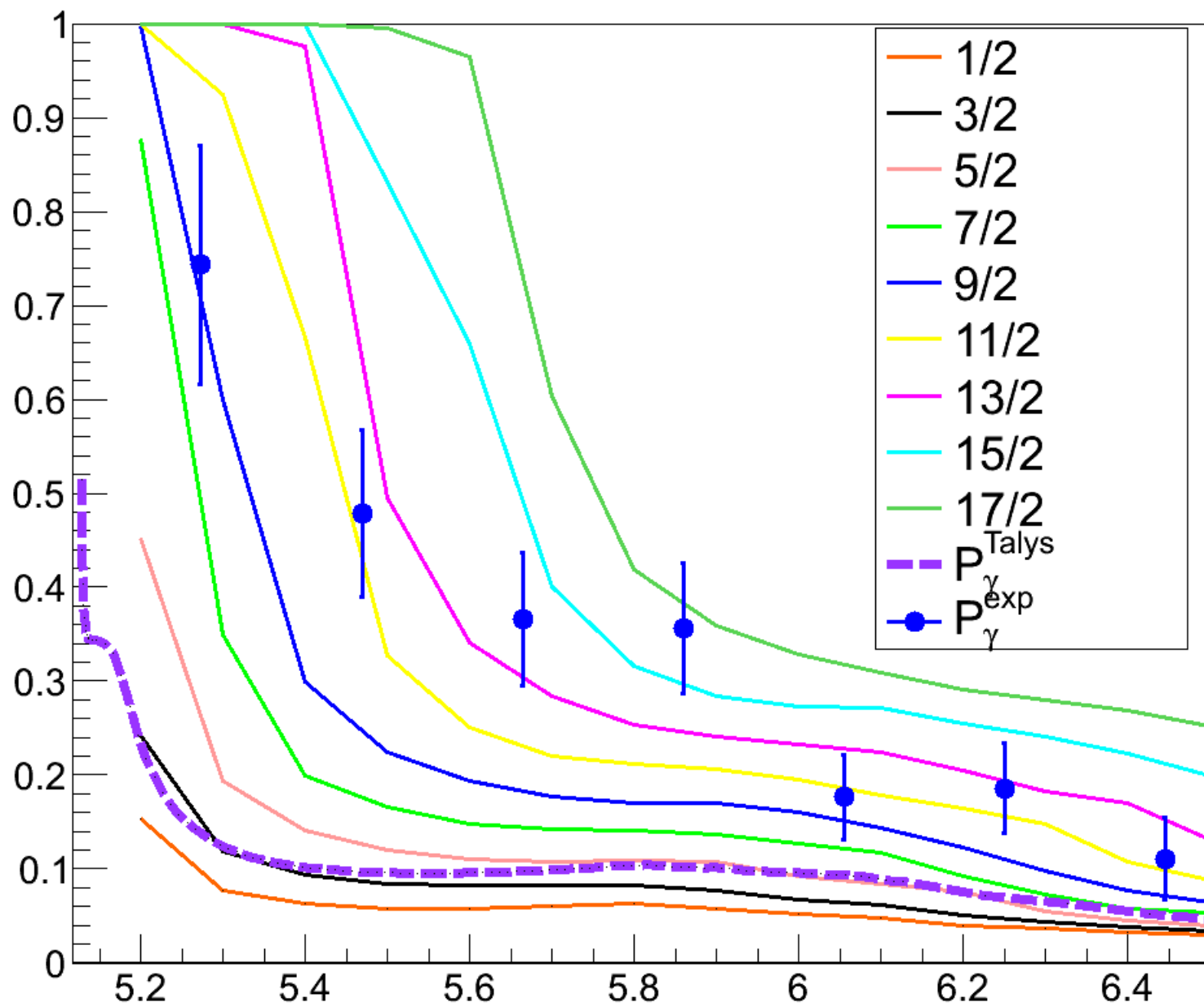
Rapport d'embranchement
calculé avec TALYS



$$\langle J \rangle = 7 \hbar$$

$$\sigma = 2.3 \hbar$$

Parity 50/50



Conditions $T_{1/2} > 10$ microseconds $DE > 30$ keV

Possible isomers:

^{234}Pa : GS, $4+$, 6.7 h

$^{234\text{m}}\text{Pa}$: 74 keV, $0-$, 1.1. m

^{216}Th : GS, $0+$, 26.0 ms

$^{216\text{m}}\text{Th}$: 2.0400 MeV, $8+$, 134 μs

^{213}Ra : GS, $1/2-$, 2.73 m

$^{213\text{m}}\text{Ra}$: 1.7700 MeV, $17/2-$, 2.20 ms

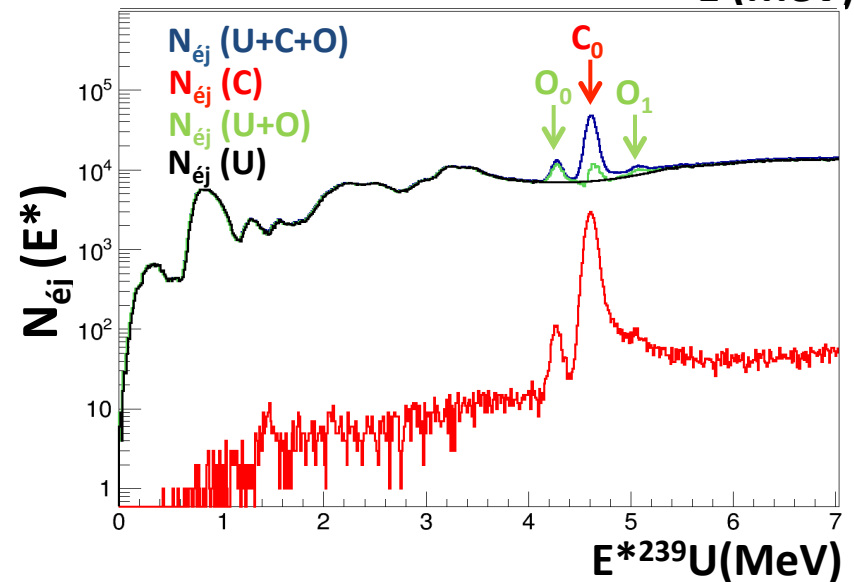
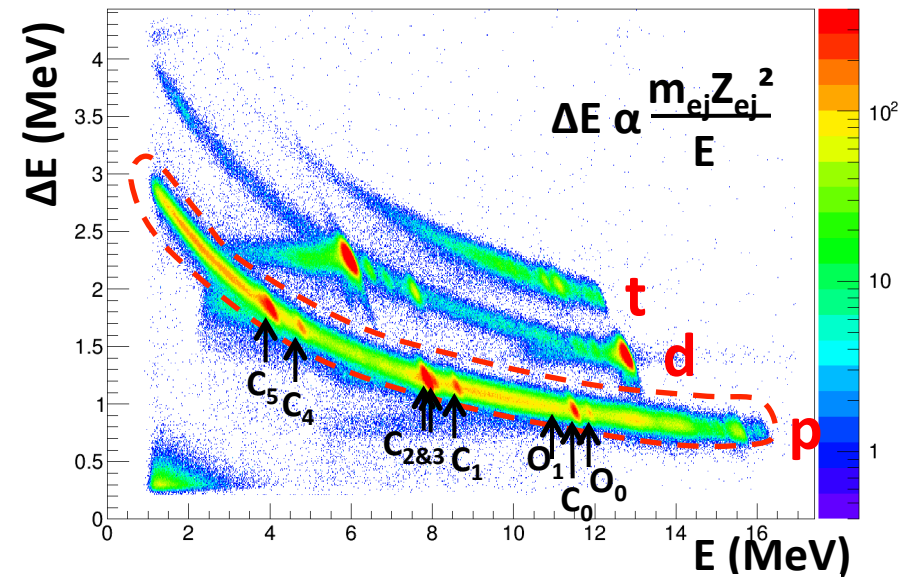
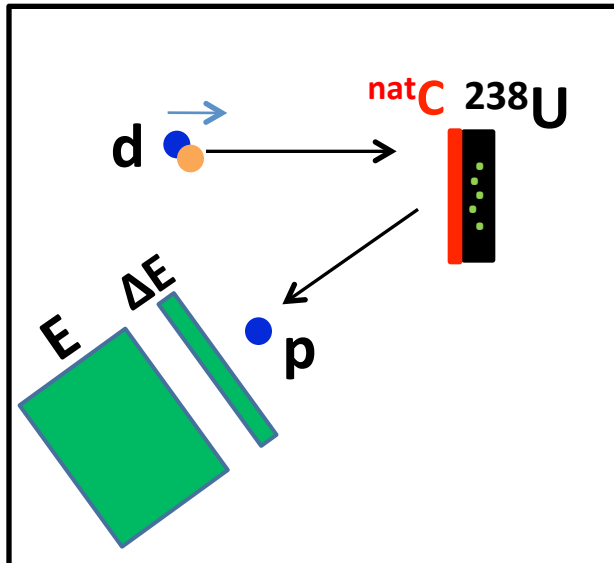
III) Mesures des probabilités d'émission gamma et de fission

Analyse des données : détermination de $P_{s,\chi}(E^*) = N_{\chi}^{\text{coinc}}(E^*) / N_{\text{éj}}(E^*) \cdot \varepsilon_{\chi}(E^*)$

- Identification du noyau composé associé à la voie de substitution grâce à la matrice d'identification des éjectiles
- Cinématique des réactions de transfert :

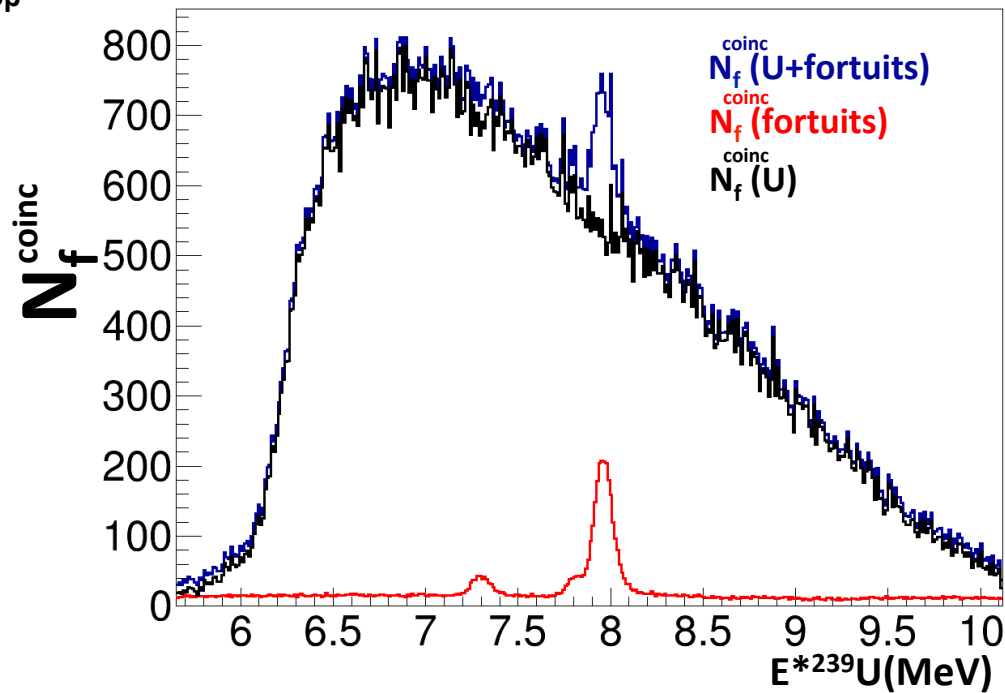
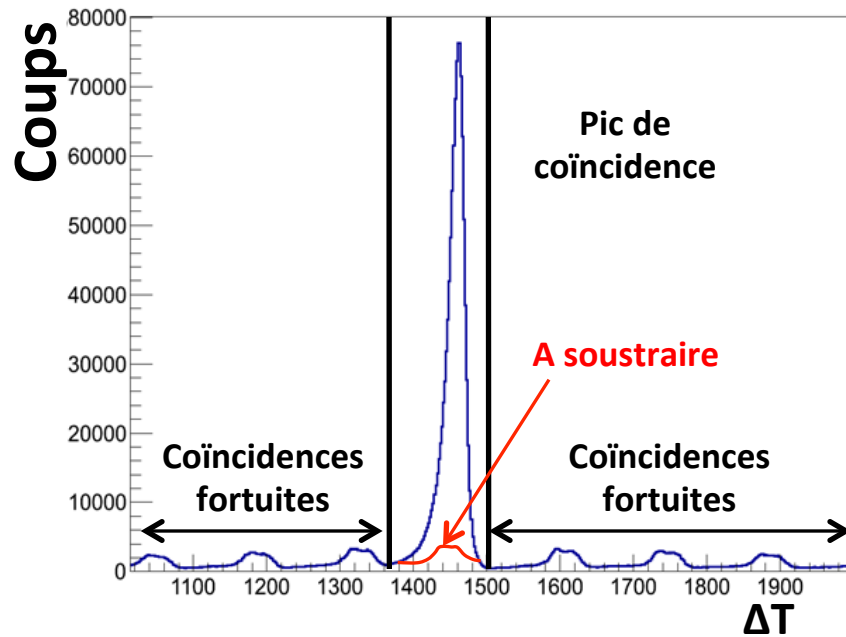
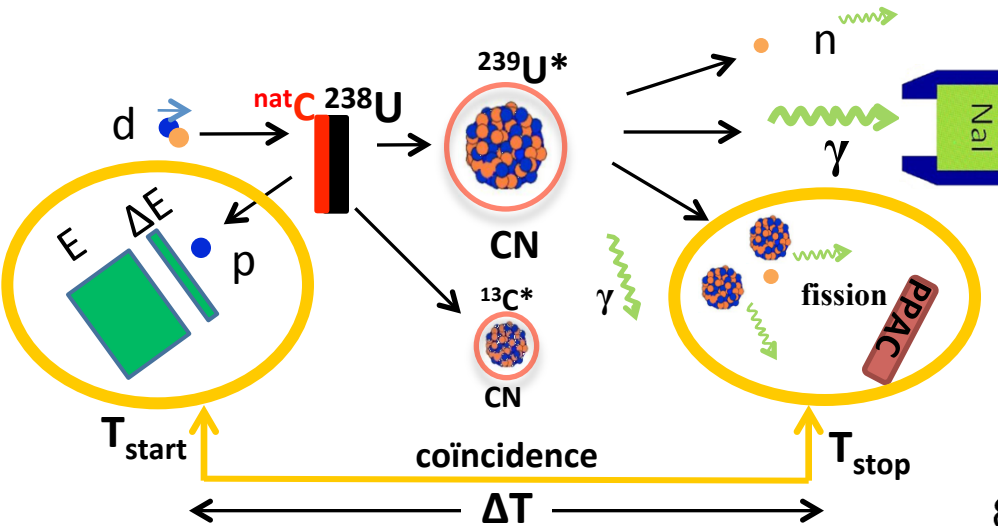
$$E^*(^{239}\text{U}) = E_{\text{faisceau}} + Q - E_{\text{recul}}(m_{\text{éj}}, \Theta_{\text{éj}}) - E_{\text{éj}}$$

Soustraction du fond Carbone et de la contamination à l'oxygène



III) Mesures des probabilités d'émission gamma et de fission

Analyse des données : détermination de $P_{s,f}(E^*) = \frac{N_f^{coinc}(E^*)}{N_{éj}(E^*) \cdot \epsilon_f(E^*)}$



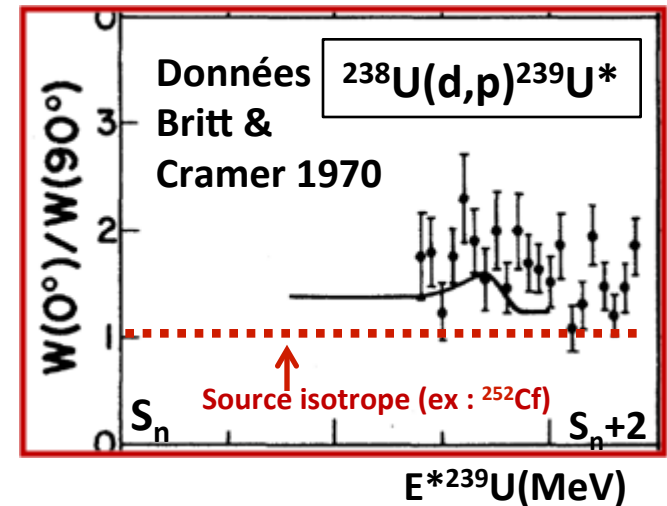
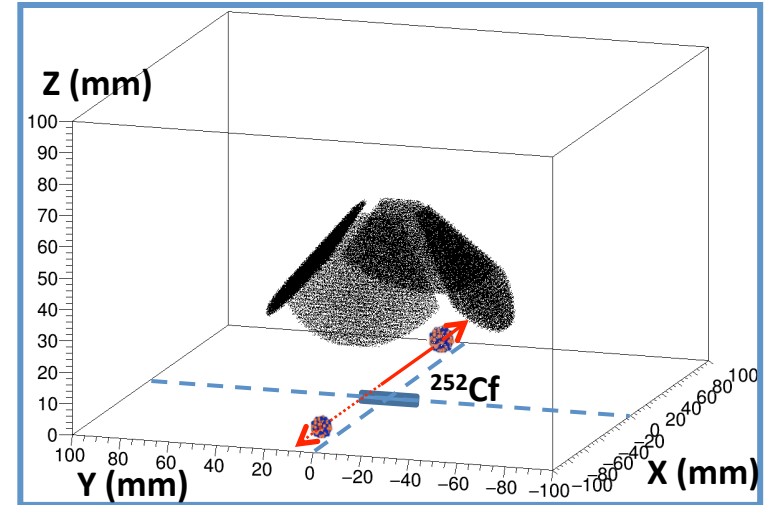
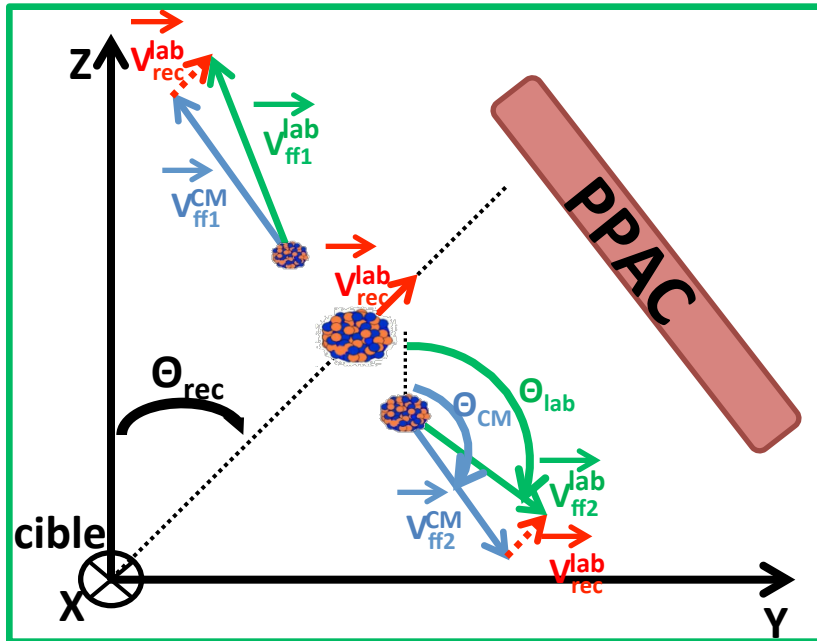
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$$\epsilon_f^{tot} = f(\epsilon_f^{géo}, \epsilon_f^{anisotropie}, \epsilon_f^{entraînement})$$

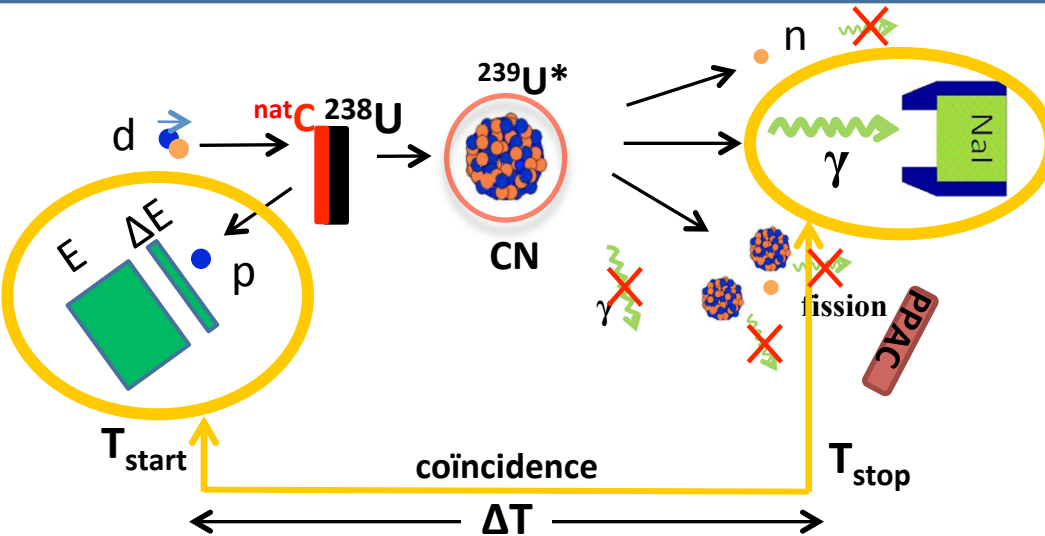
$$48 \% = 41 \% + 4 \% (1-7\%) + 3 \%$$

- Mesure de $\epsilon_f^{géo}$ avec une source isotrope d'émission de fragments de fission de ^{252}Cf d'activité connue → Simulation
- Inclusion de $\epsilon_f^{anisotropie}$ dans le Centre de Masse (CM) dans la simulation avec les données de Britt et Cramer
- Inclusion de $\epsilon_f^{entraînement}$ dans la simulation en prenant en compte l'effet d'entraînement du noyau de recul



III) Mesures des probabilités d'émission gamma et de fission

Analyse des données : détermination de $P_{s,\gamma}(E^*) = \frac{N_{\gamma}^{\text{coinc}}(E^*)}{N_{\text{éj}}(E^*) \cdot \varepsilon_{\gamma}(E^*)}$



Suppression des gammas « **post-neutron** » ●

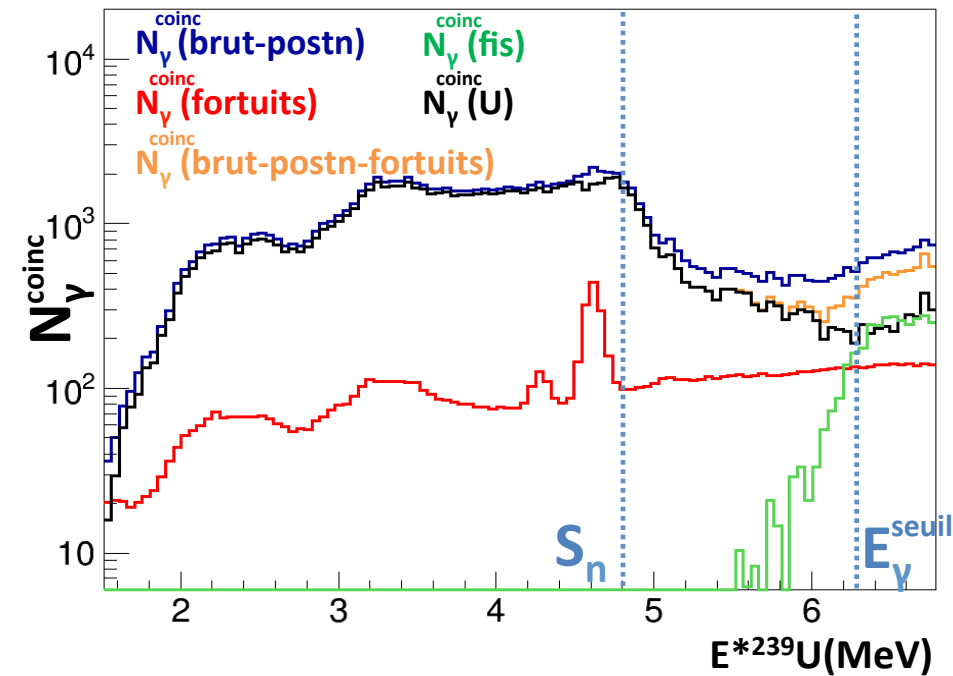
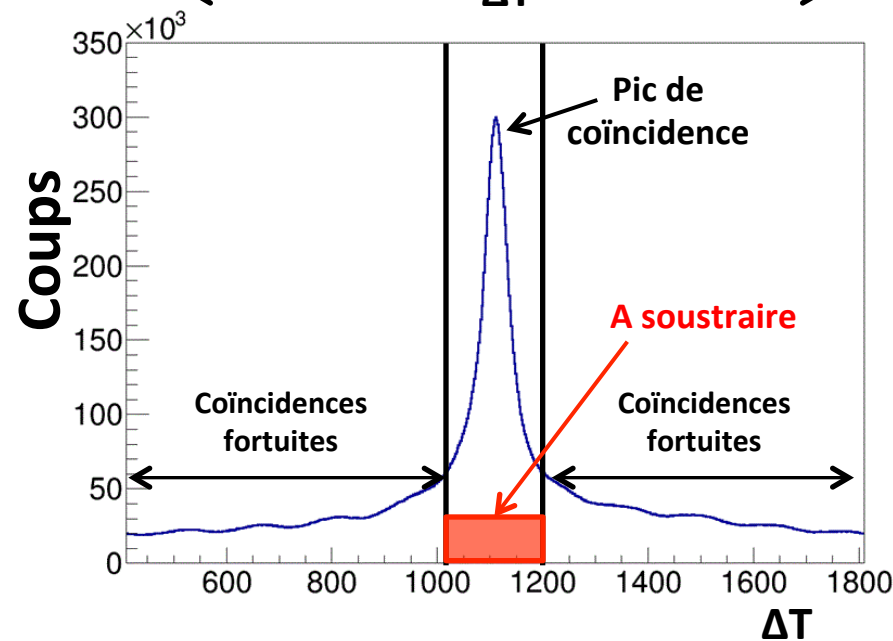
$$E_{\gamma, \text{post-n}}^{\text{max}} = E^* - S_n$$



$$E_{\gamma}^{\text{seuil}} = E_{\gamma, \text{post-n}}^{\text{max}} = 1,5 \text{ MeV}$$

Suppression des coïncidences **fortuites** ●

Suppression des gammas de **fission** ●



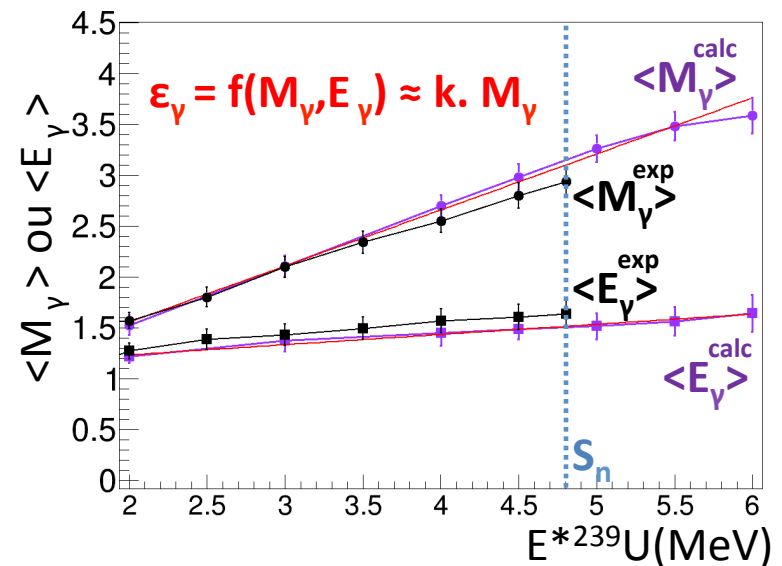
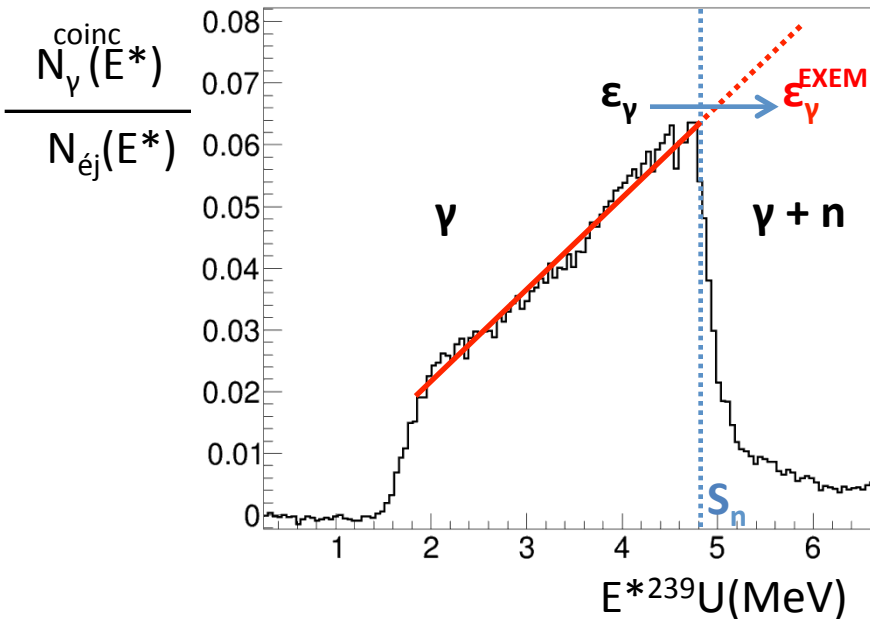
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Méthode EXEM (EXtrapolated Efficiency Method) :

$$P_{\gamma}(E^*) = \frac{N_{\gamma}^{\text{coinc}}(E^*)}{N_{\text{éj}}(E^*) \times \epsilon_{\gamma}(E^*)} = 1 \text{ (pour } E^* < S_n) \Rightarrow \epsilon_{\gamma}(E^*) = \frac{N_{\gamma}^{\text{coinc}}(E^*)}{N_{\text{éj}}(E^*)}$$

Hypothèse de la méthode : la tendance de $\epsilon_{\gamma}(E^*)$ se poursuit à plus haute E^*



- Les calculs EVITA basés sur le modèle statistique confortent l'hypothèse de la méthode
- Méthode EXEM validée par le calcul de $\epsilon_{\gamma}(E^*)$ par la méthode des fonctions de poids