Деление ядер нейтронами с энергиями 1-200 МэВ: расчёты сечений, угловых распределений осколков, смежные вопросы

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Прикладные задачи (нейтроны до 200 МэВ)

- Источники нейтронов на основе расщепления тяжёлых ядер релятивистскими протонами (Spallation Neutron Sources).
- Ядерная энергетика: трансмутация ядерных отходов, ADS–системы (ADS – accelerator-driven systems).
- Радиационное материаловедение: радиационно устойчивые материалы, радиационная безопасность, …
- Ядерная медицина: адронная терапия, производство фармпрепаратов, …

Studies of angular distributions of fission fragment in neutron-induced fission at energies above 20-30 MeV began quite recently:

Ryzhov et al. (up to 100 MeV, 2005), n_TOF and NIFFTE (up to 200 MeV, 2014-2020): ^{232}Th, ^{235}U, ^{238}U

Petersburg Nuclear Physics Institute (PNPI), Gatchina

Spallation neutron source at 1 GeV proton synchrocyclotron

and

Gatchina Neutron time-of-flight Spectrometer (GNeiS)

1-200 MeV, 2015-2020: ^{209}Bi, Pb (nat), ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{240}Pu;

2021: ^{236}U

Motivation:

- new nuclear technologies, e.g., ADS
- new information on fission process

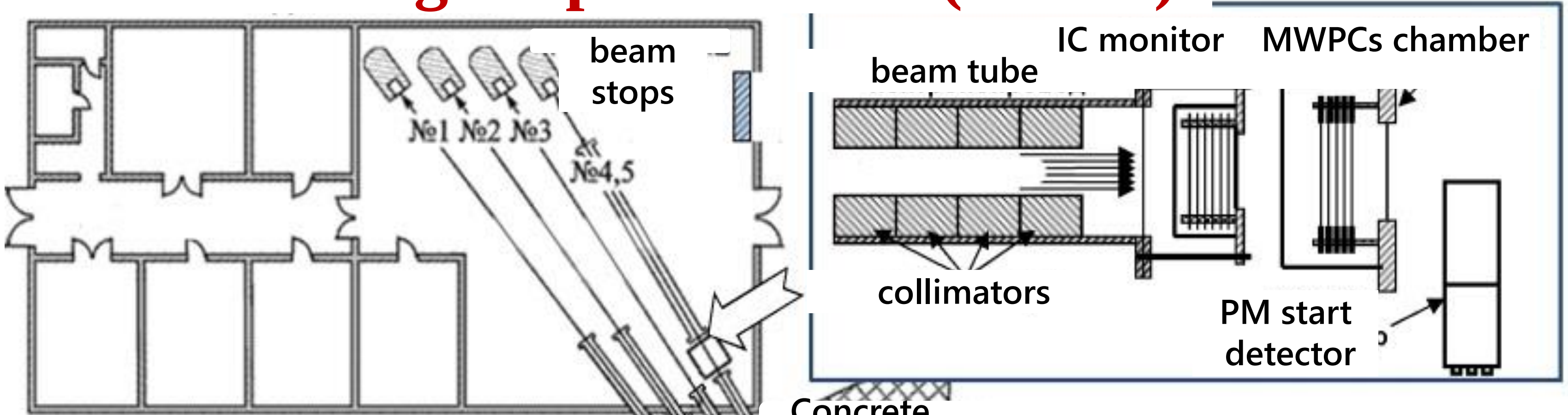
In our last publications:

A.S. Vorobyev et al. JETP Lett. 110, 242 (2019): ^{237}Np

A.S. Vorobyev et al. JETP Lett. 112, 323 (2020): ^{240}Pu

not only experimental data were presented, but results of calculation with the use of the TALYS-based code

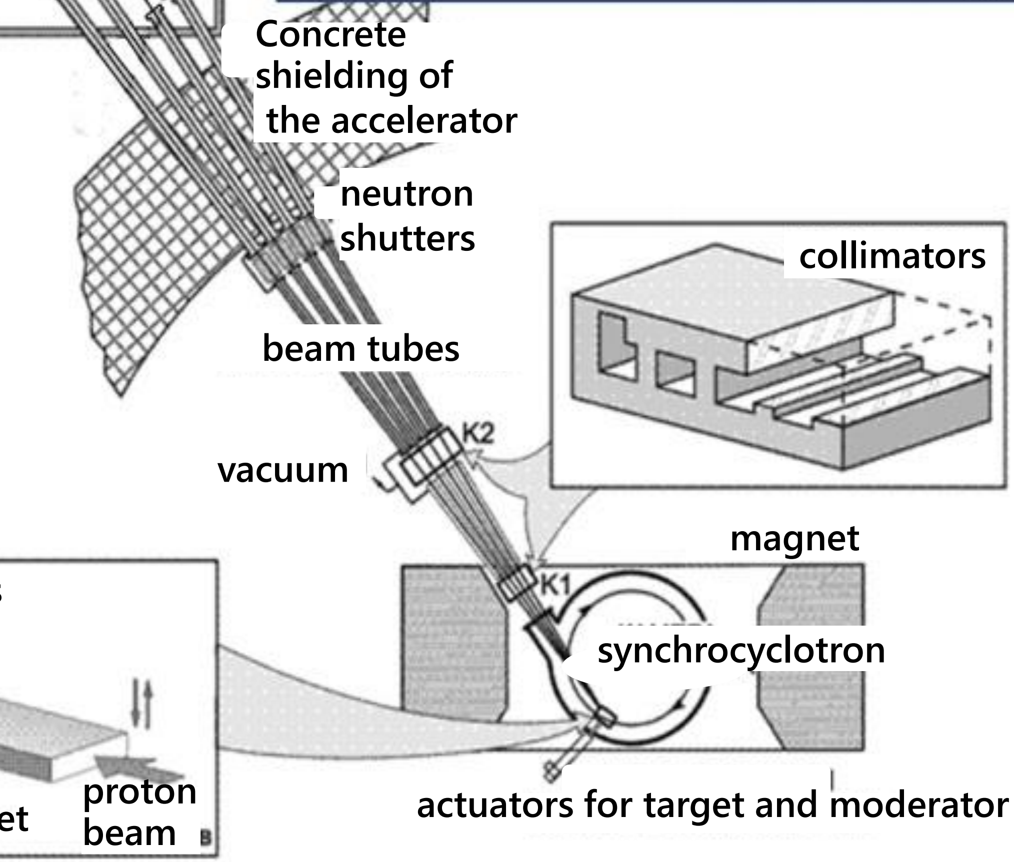
Gatchina Neutron time-of-flight Spectrometer (GNeiS)

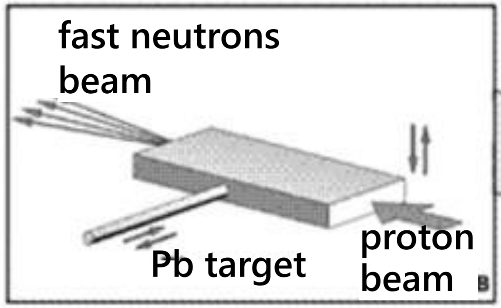


Main parameters:

$$E_p = 1\ GeV,\ \ f \approx 50\ \ Hz,\ \ \Delta t \approx 10\ \ ns,\ \ \Phi \sim 3 \cdot 10^{14}\ \frac{n}{s}$$

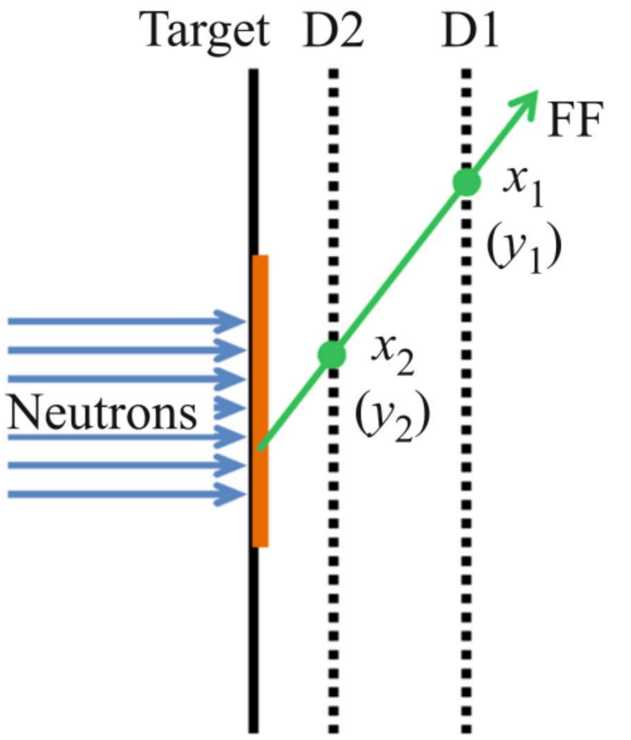
$$L = 35.5\ \ m,\ \ \frac{\Delta E}{E}(1\ \ MeV) \approx 1\%,\ \ \frac{\Delta E}{E}(200\ \ MeV) \approx 12\%$$

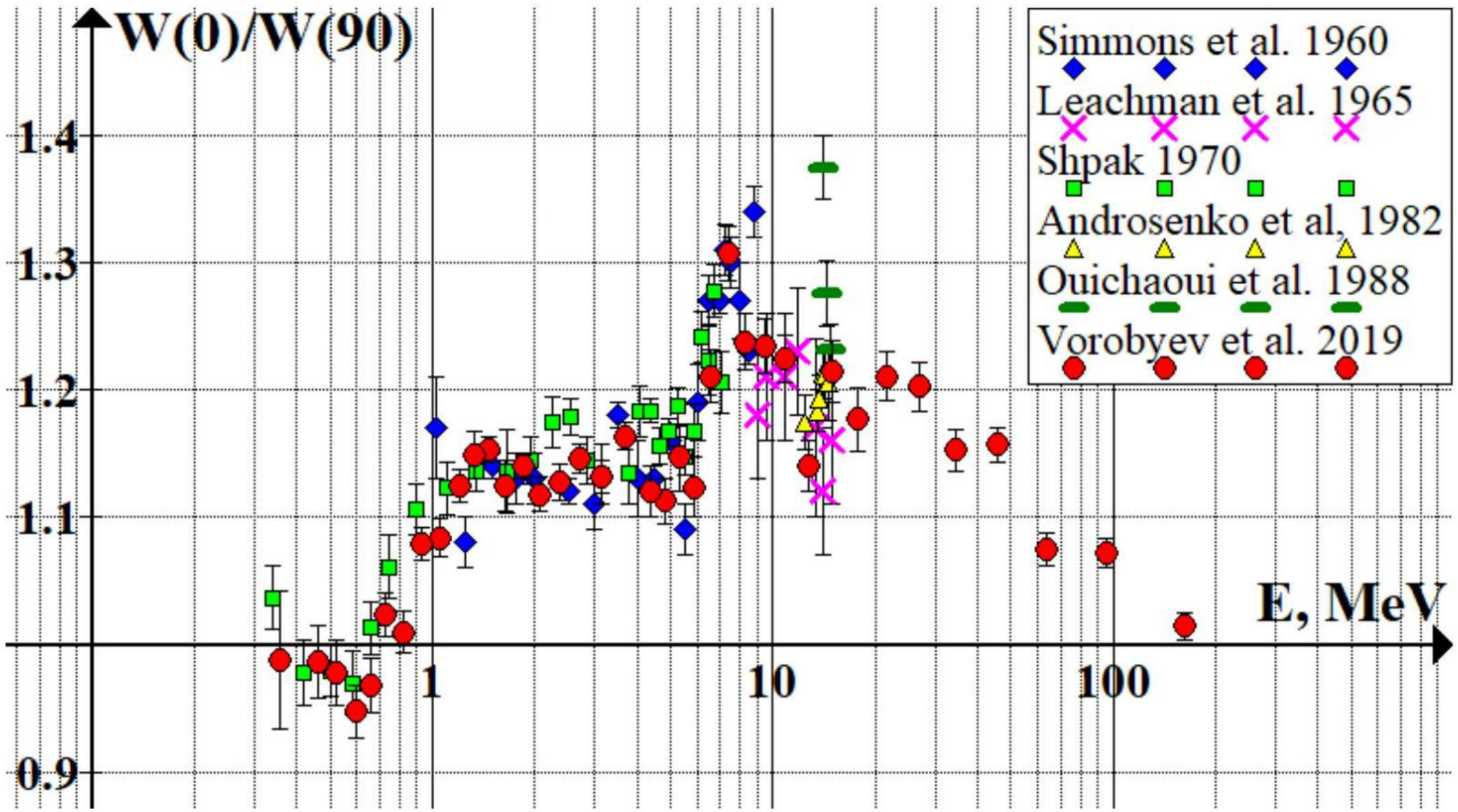




Spallation neutron source at 1 GeV proton synchrocyclotron

Vorobyev et al., 2019: $n + {}^{237}\mathrm{Np} \to$ Fission Fragments (FF) + ...





$$W(\theta) = \frac{d\sigma_f(\theta)/d\Omega}{\sigma_f} \approx \frac{1}{4\pi}\left(1 + A_2 P_2(\cos\theta) + A_4 P_4(\cos\theta)\right),$$

$$\frac{W(0)}{W(90)} = \frac{1 + A_2 + A_4}{1 - A_2/2 + 3A_4/8} \approx \frac{1 + A_2}{1 - A_2/2}$$

$$|A_4| \ll |A_2|$$

O. Bohr model of nuclear transition states on the barriers

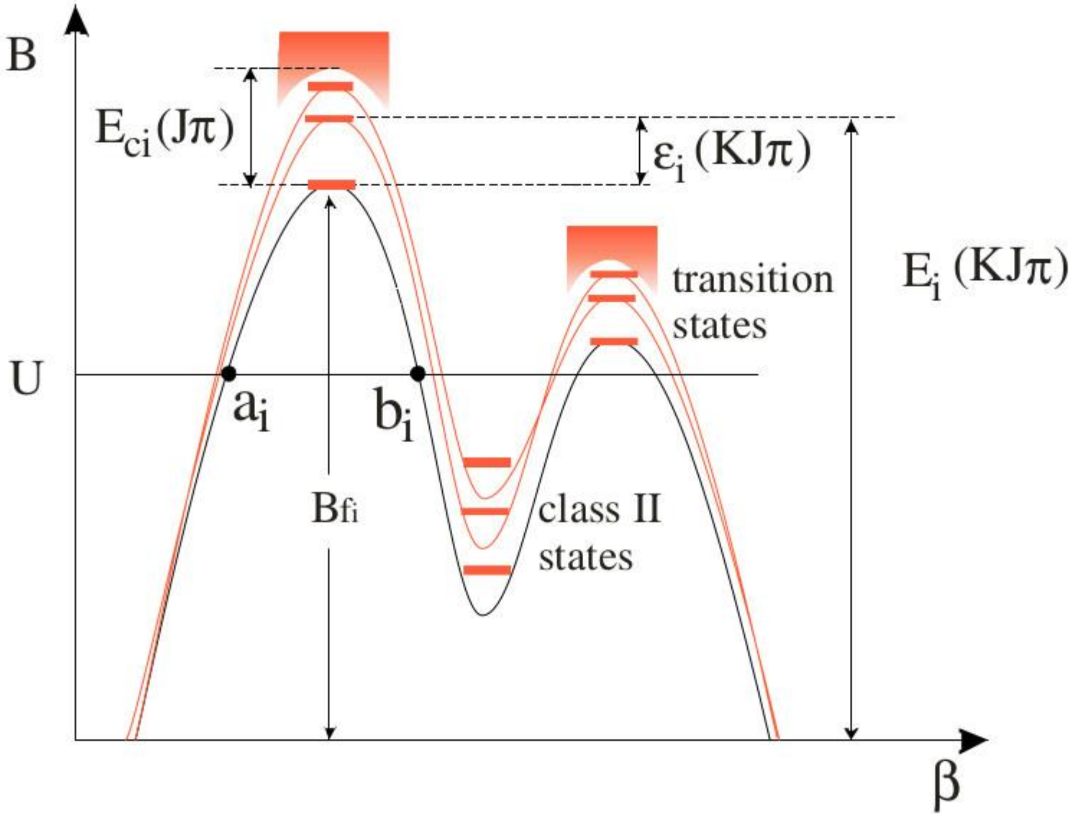


Figure: R. Capote et al. *Nuclear Data Sheets*, 2009, v. 110, p. 3107

The problem of calculating the angular distribution of fragments from nuclear fission is as old as the problem of calculating the fission cross sections.

Because in both cases we refer to the same O. Bohr model of transition states of a strongly deformed nucleus at the fission barriers.

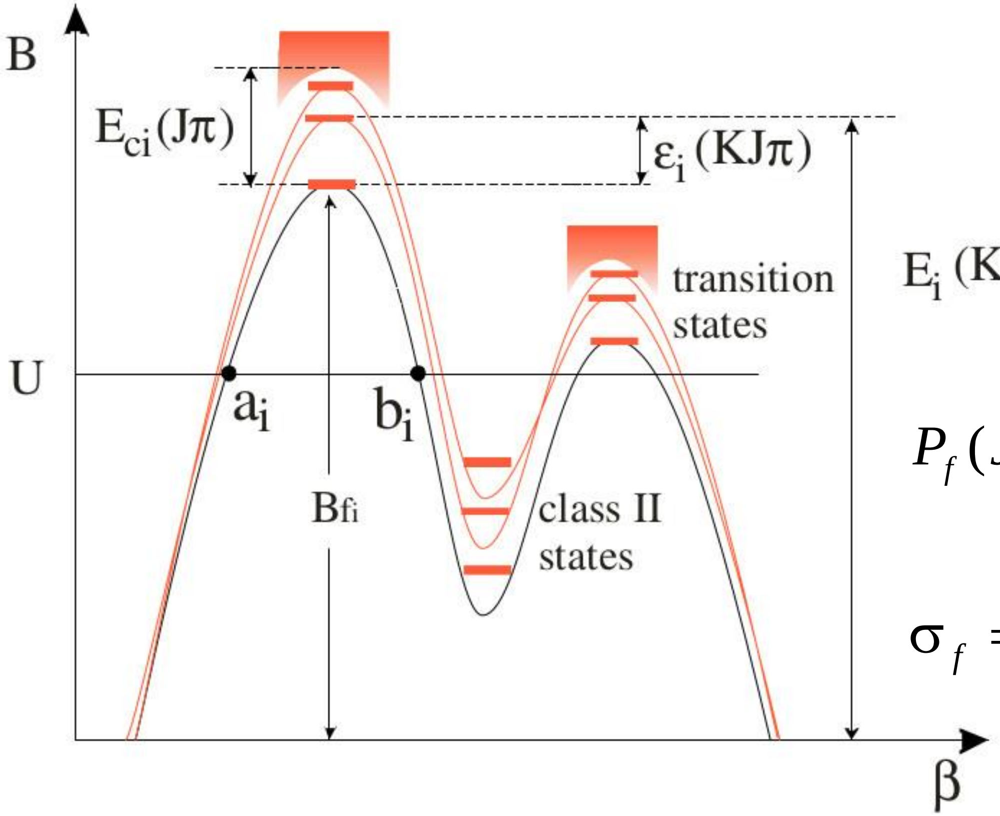
Indeed, the fission cross section is determined by the density of transition states, while the angular distribution of fragments is determined by the projections K of the fragment's spin onto the deformation axis of the same transition states.

However, we see that the current interest in these two problems - angular distributions and cross sections - is very different. There are many computer programs for nuclear reactions, such as TALYS and EMPIRE, that calculate fission cross sections, but none of them calculate the angular distribution of fragments.

Algorithm for calculation the fission cross section

“Nuclear fission remains the most complex topic in applied nuclear physics…”

R. Capote et al. RIPL – Reference Input Parameter Library for Calculations of Nuclear Reactions and Nuclear Data Evaluations. *Nuclear Data Sheets*, 2009, v. 110, p. 3107



$$E_i\,(KJ\pi) = B_{fi} + \varepsilon_i(K\pi) + \frac{\hbar^2}{2I_i}(J(J+1) - K(K+1))$$

$$P_f(J\pi) \sim \sum_i T_f^{J\pi}(E_{ex} - B_f - E_i(KJ\pi)) + \int_{E_c(J\pi)}^{E_{ex}} \rho(\varepsilon J\pi)\; T_f^{J\pi}(E_{ex} - B_f - \varepsilon)d\varepsilon$$

$$\sigma_f = \sum_{J\pi} \sigma(J\pi)P_f(J\pi)$$

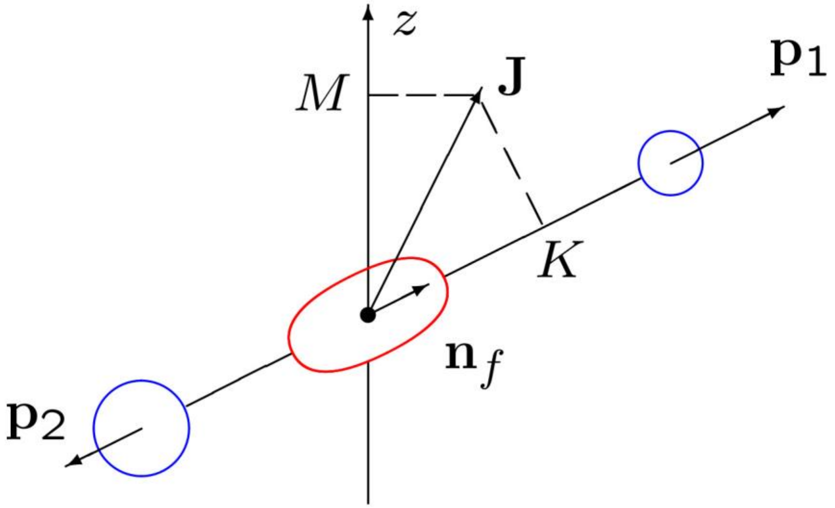
Hill-Wheeler Transmission Factor:

$$T_i(U - B_i) = \frac{1}{1 + e^{-2\pi(U - B_i)/\hbar\omega_i}}$$

“This work and the resulting database are extremely important to theoreticians involved in the development and use of nuclear reaction modelling (ALICE, EMPIRE, GNASH, UNF, TALYS) both for theoretical research and nuclear data evaluations.”

Algorithm for calculation the angular distribution of fission fragments

$$W(\theta) = \frac{1}{\sigma_f}\frac{d\sigma_f(\theta)}{d\Omega}$$



$$\Psi_{JMK} \sim D^J_{MK}(\vec{n}_f)$$

$$\frac{dw^J_{MK}(\vec{n}_f)}{d\Omega} = \frac{2J+1}{4\pi}\left|D^J_{MK}(\vec{n}_f)\right|^2$$

$$\frac{dw^{J\pi}(\vec{n}_f)}{d\Omega} = \sum_M \eta^{J\pi}(M)\sum_K \rho^{J\pi}(K)\frac{dw^J_{MK}(\vec{n}_f)}{d\Omega}$$

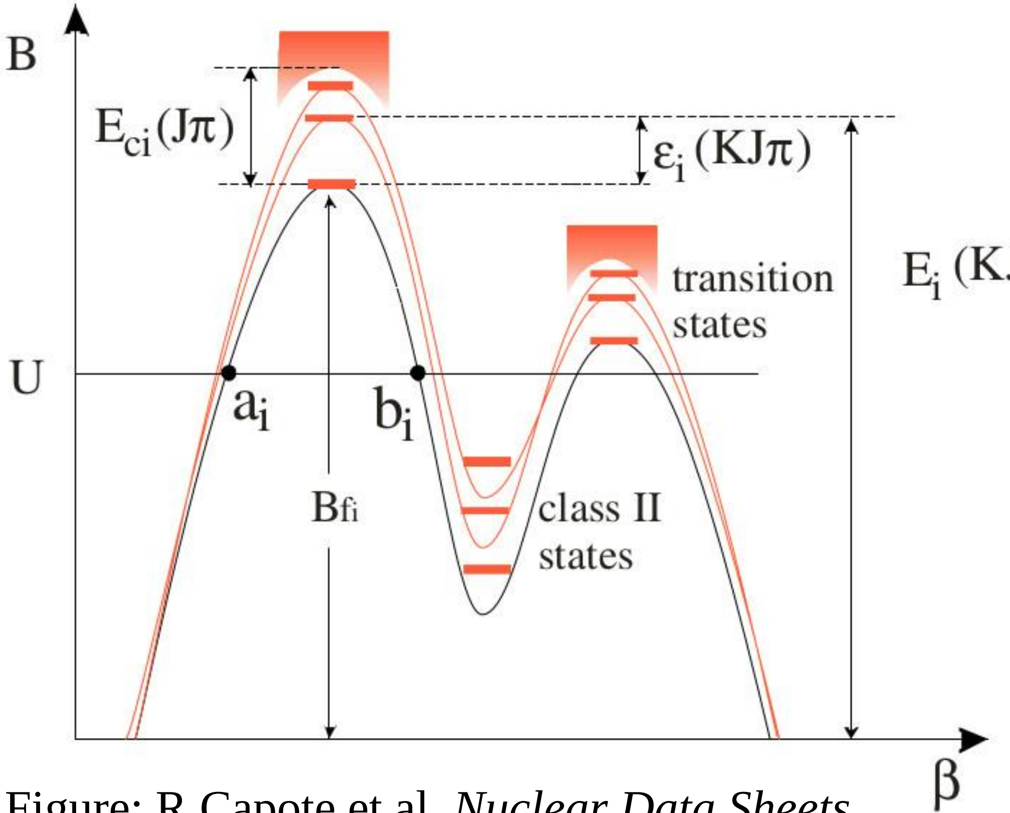
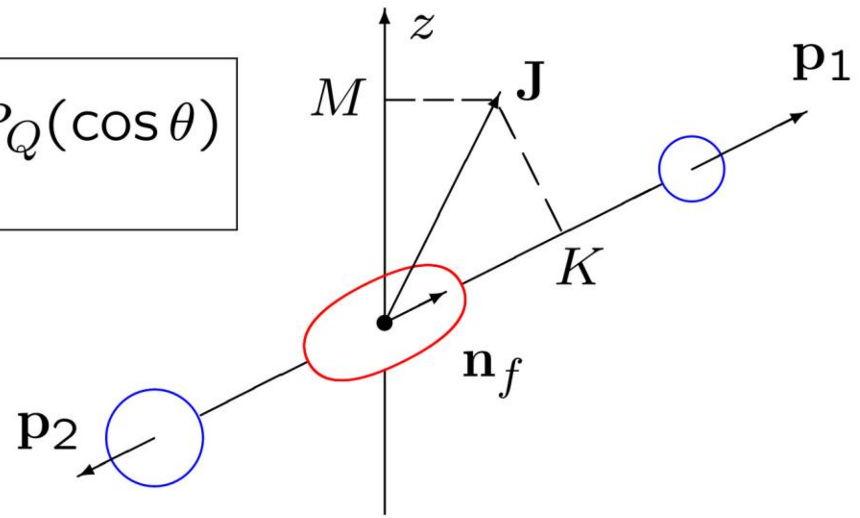


Figure: R.Capote et al. *Nuclear Data Sheets*, 2009, v. 110, p. 3107

To calculate $\frac{d\sigma_f(\theta)}{d\Omega} = \sum_{J\pi}\sigma(J\pi)P_f(J\pi)\frac{dw^{J\pi}(\theta)}{d\Omega}$ we need $\eta^{J\pi}(M)$ and $\rho^{J\pi}(K)$

Convenient transformation:

$$(2J+1)\left|D^J_{MK}(\mathbf{n}_f)\right|^2 = \sum_Q (2Q+1) C^{JM}_{JMQ0} C^{JK}_{JKQ0} P_Q(\cos\theta)$$



$$\frac{dw^J(\mathbf{n}_f)}{d\Omega} = \sum_M \eta^J(M) \sum_K \rho^J(K) \frac{2J+1}{4\pi} |D^J_{MK}(\mathbf{n}_f)|^2 \equiv$$

$$\frac{1}{4\pi} \sum_{Q=0,2,4,\ldots} (2Q+1) \underbrace{\left(\sum_M C^{JM}_{JMQ0}\, \eta^J(M)\right)}_{\substack{|||\\ \tau_{Q0}(J)}} \underbrace{\left(\sum_K C^{JK}_{JKQ0}\, \rho^J(K)\right)}_{\substack{|||\\ \beta_Q(J)}} P_Q(\cos\theta)$$

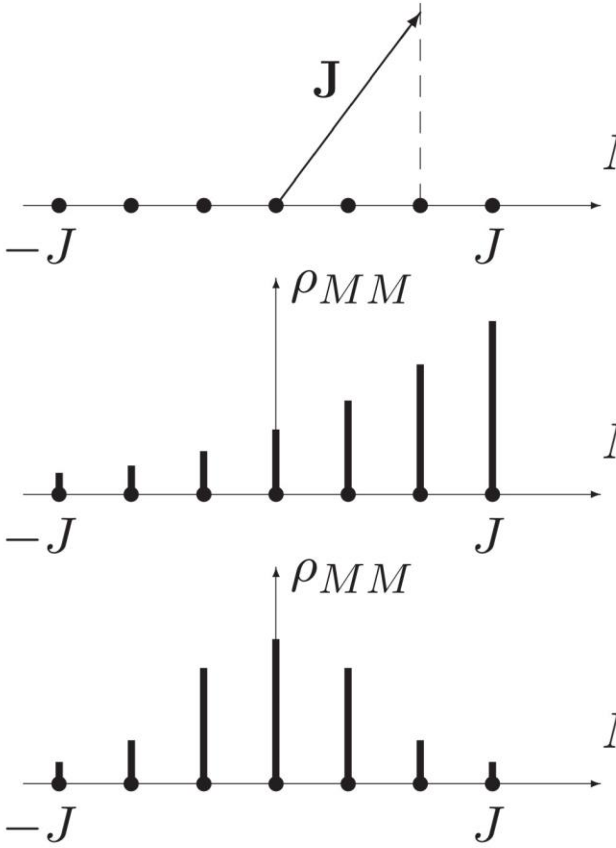
$\tau_{Q0}(J)$ — spin-tensor of orientation, $\beta_Q(J)$ — parameter of anisotropy

$$\frac{d\sigma_f(\theta)}{d\Omega} = \frac{\sigma_f}{4\pi} + \frac{1}{4\pi} \sum_{Q=2,4,\ldots} \sigma^C_{fQ} P_Q(\cos\theta) \quad \Rightarrow \quad W(\theta) = \frac{1}{\sigma_f} \frac{d\sigma_f(\theta)}{d\Omega}$$

Спиновая ориентация: матрица плотности и спин-тензоры

$$\Psi_J = \sum_M a_M \Psi_{JM}, \qquad \rho_{MM'} = \langle a_M a^*_{M'} \rangle \quad \to \quad \rho_{MM'} = \rho_{MM}\delta_{MM'}$$

$$\tau_{Qq}(J) = \sum_{MM'} C^{JM'}_{JMQq}\rho_{MM'} \quad \to \quad \tau_{Qq}(J) = \tau_{Q0}(J)\delta_{q0}(J)$$



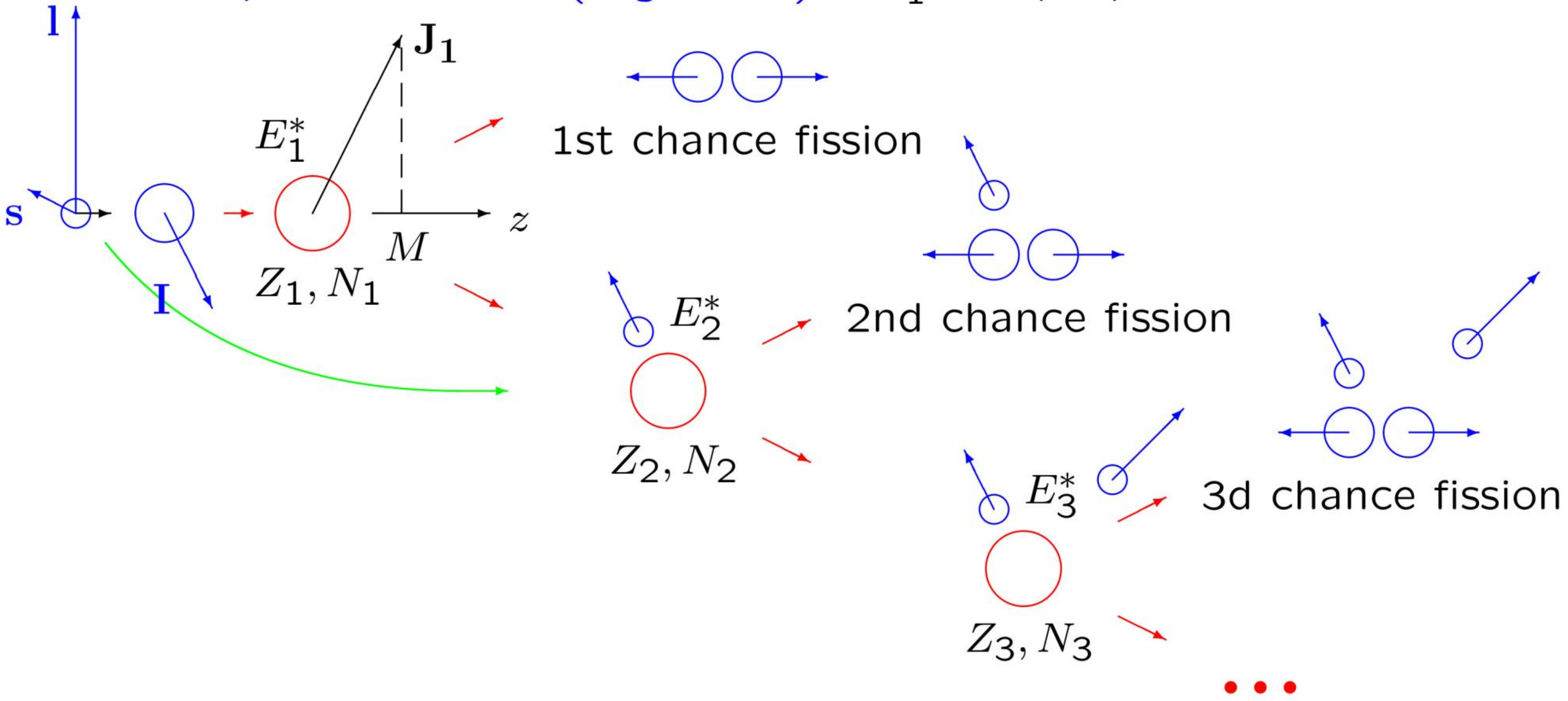
$$\tau_{10}(J) \sim \frac{\langle M \rangle}{J}$$

Поляризация

$$\tau_{20}(J) \sim \frac{3\langle M^2 \rangle - J(J+1)}{J^2}$$

Выстроенность

Nuclear spin orientation (alignment): $\mathbf{J}_1 = \mathbf{s} + \mathbf{l} + \mathbf{I}$



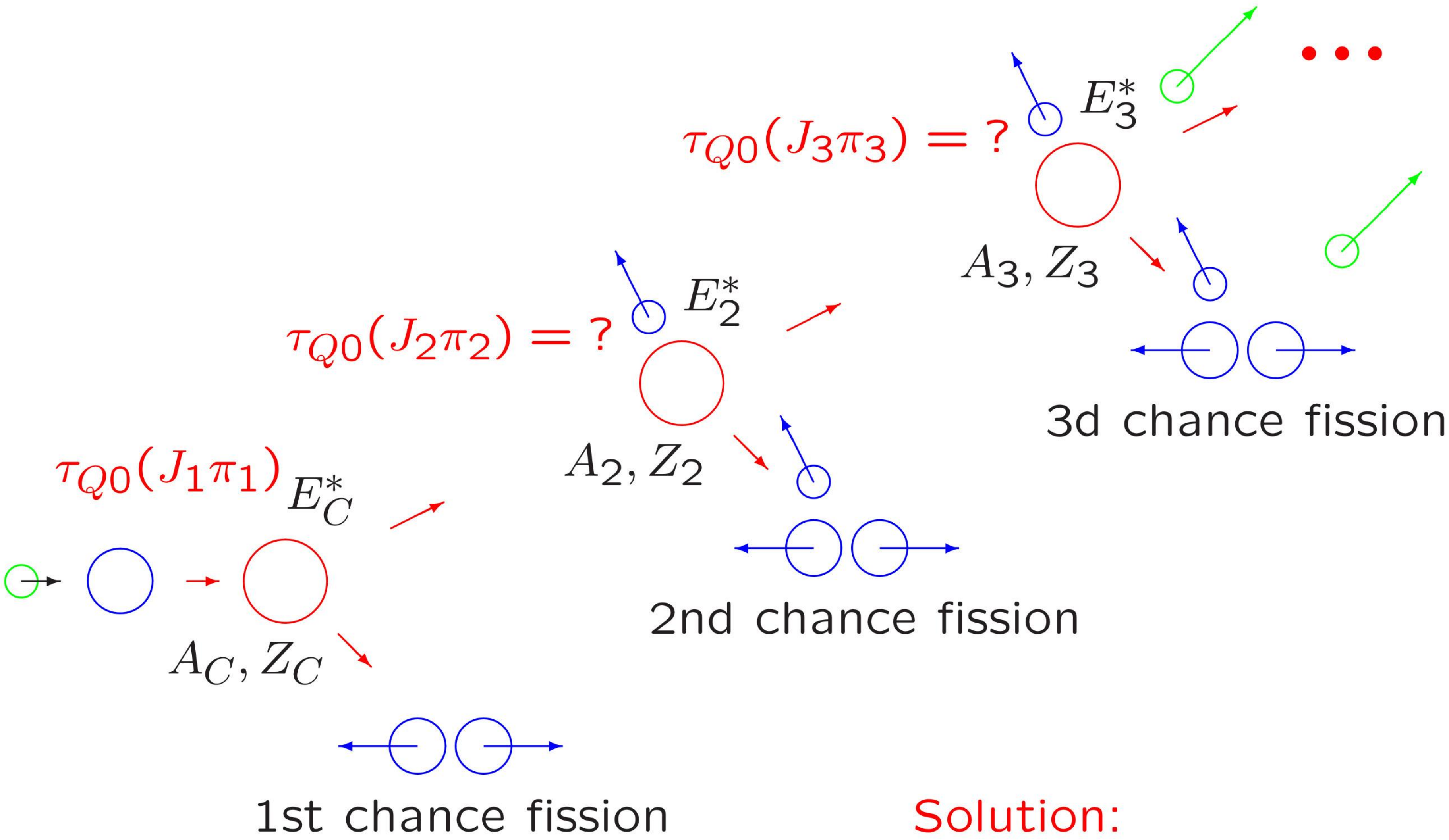
Outline of our approach for calculating the fission fragment angular distribution

$$\frac{d\sigma_f(\mathbf{n}_f)}{d\Omega} = \frac{\sigma_f^{DPE}(\mathbf{n}_f)}{4\pi} + \sum_{ZNJ\pi i} \sigma_{ZN}^{C}(J\pi i) P_f^{ZN}(J\pi i) \frac{dw_{ZN}^{J\pi i}(\theta)}{d\Omega}$$

$$\frac{dw_{ZN}^{J\pi i}(\theta)}{d\Omega} = \sum_M \eta_{ZN}^{J\pi i}(M) \sum_K \rho_{ZN}^{J\pi i}(K) \frac{dw_{MK}^{J}(\theta)}{d\Omega}, \quad \eta_{ZN}^{J\pi i}(M) = \frac{\sigma_{ZN}^{C}(J\pi M i)}{\sigma_{ZN}^{C}(J\pi i)}$$

M-dependence is calculated in the extended TALYS

Cascade deexcitation of highly excited nucleus and fission



Solution:

particle: γ, n, p, d, t=^{3}H, h=^{3}He, α, . . .

$$\tau_Q(J_2\pi_2) \sim W(jJ_1J_2Q, J_2J_1)\tau_Q(J_1\pi_1)$$

TALYS-1.9

New Edition December 21, 2017

A nuclear reaction program

User Manual

Arjan Koning
Stephane Hilaire
Stephane Goriely

Talys is a computer code system for the analysis and prediction of nuclear reactions.

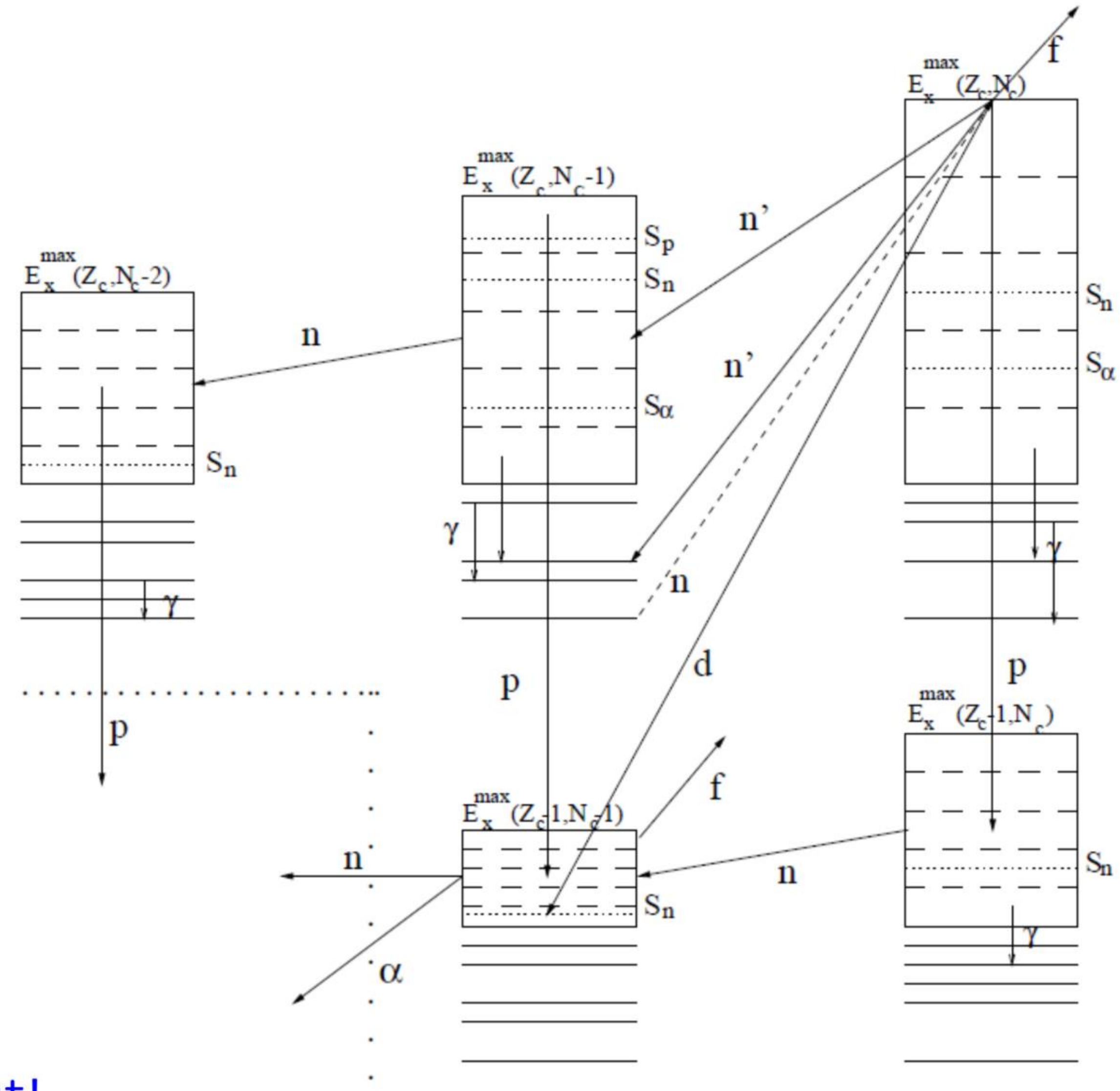
The basic objective is the simulation of nuclear reactions that involve neutrons, photons, protons, deuterons, tritons, 3He- and alpha-particles, in the 1 keV – 200 MeV energy range and for target nuclides of mass 12 and heavier.

Free use, open software, always under development: from TALYS-1.0 — December 2007 to TALYS-1.9 — December 2017.

More than 300 subroutines, more than 100 000 lines (commands), more than 500 pages in the Manual.

Completely integrated optical model and coupled-channels calculations by the ECIS-06 code.

All partial cross sections can be found, due to



the calculation of
all transition probabilities:

$w(i \to i')$, where

$i \equiv (Z_i, N_i, E_i^*, J_i, \pi_i)$

But!

— angular distributions — only for the first step reaction: a+A $\to$ C$\to$ b + B

— angular distribution for fission fragments (even for the first step or first chance) can not be calculated

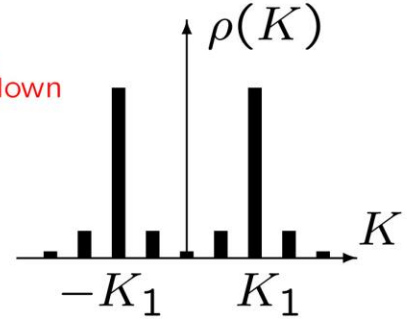
$^{237}Np(n,f)$: Results for Angular anisotropy

A.S.Vorobyev et al. JETP Lett. 110, 242 (2019)

«Low» energies:

$$E^* = E_{ex} - B_f < \Delta + U_{\text{down}}$$

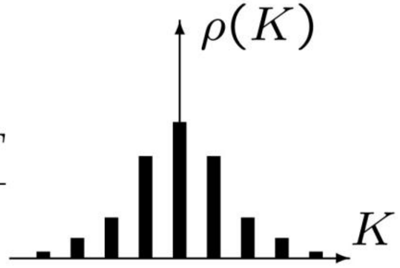
$$\rho_{ZN}^{J\pi i}(K) \sim e^{-\alpha(|K|-K_1)^2}$$



«High» energies:

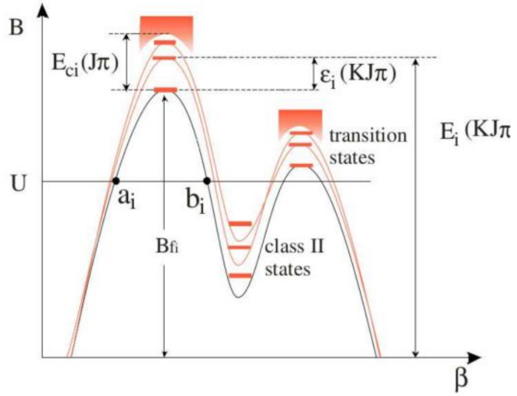
$$E^* = E_{ex} - B_f > \Delta + U_{\text{up}}$$

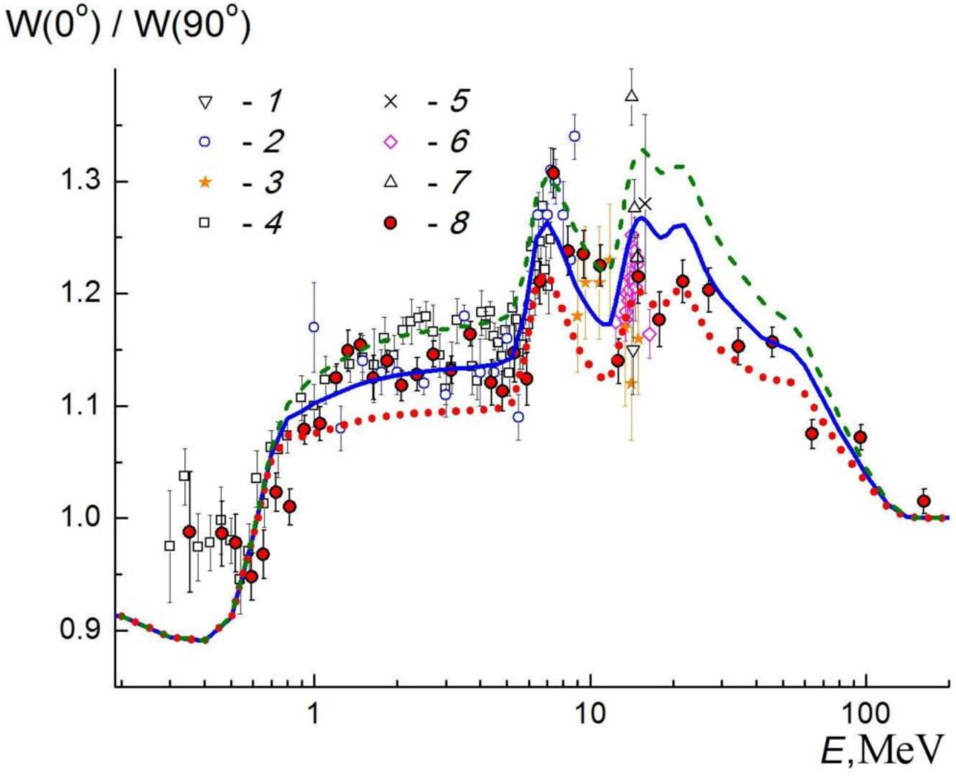
$$\rho_{ZN}^{J\pi i}(K) \sim e^{-\frac{K^2}{2K_0^2}},\ K_0^2 = \frac{J_{\text{eff}}T}{\hbar^2}$$



Additional parameters:

$$U_{\text{up}} = 0.4\ \text{MeV},\quad U_{\text{down}} = -0.1\ \text{MeV},\quad \alpha = 0.15,\quad K_1 = \begin{cases} 0.0, & {}^{238}\text{Np}, \\ 0.5, & {}^{237}\text{Np}, \\ 1.5, & \text{all other isitopes.} \end{cases}$$





$$\frac{\hbar^2}{J_{\text{eff}}} = 0.022\ \text{MeV}$$

$$\frac{\hbar^2}{J_{\text{eff}}} = 0.017\ \text{MeV}$$

$$\frac{\hbar^2}{J_{\text{eff}}} = 0.012\ \text{MeV}$$

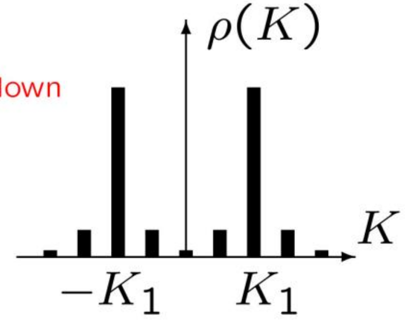
$^{240}Pu(n, f)$: Results for Angular anisotropy

A.S. Vorobyev et al. JETP Lett. 112, 323 (2020)

«Low» energies:

$$E^* = E_{ex} - B_f < \Delta + U_{\text{down}}$$

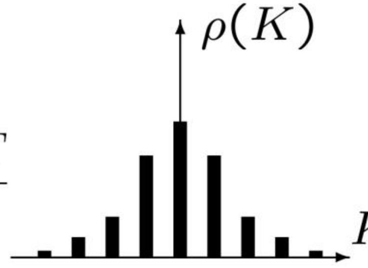
$$\rho^{J\pi i}_{ZN}(K) \sim e^{-\alpha(|K|-K_1)^2}$$



«High» energies:

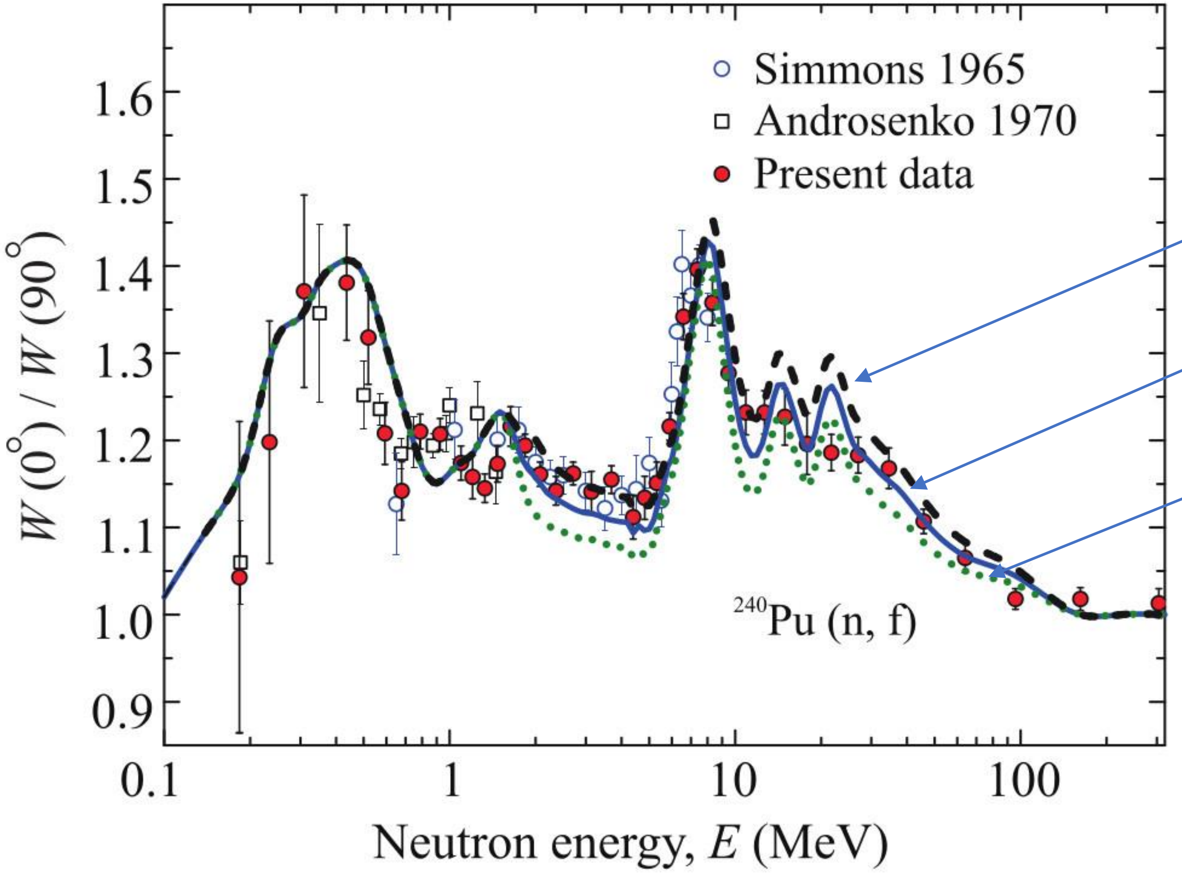
$$E^* = E_{ex} - B_f > \Delta + U_{\text{up}}$$

$$\rho^{J\pi i}_{ZN}(K) \sim e^{-\frac{K^2}{2K_0^2}}, \quad K_0^2 = \frac{J_{\text{eff}}T}{\hbar^2}$$



Additional parameters:

$$U_{\text{up}} = 0.4 \text{ MeV}, \quad U_{\text{down}} = -0.1 \text{ MeV}, \quad \alpha = 0.15, \quad K_1 = \begin{cases} \text{“special” choice, } ^{241}\text{Pu} \\ 1.9, \quad ^{240}\text{Pu} \\ 1.5, \quad \text{all other isitopes.} \end{cases}$$



$$\frac{\hbar^2}{J_{eff}} = 9 \ keV$$

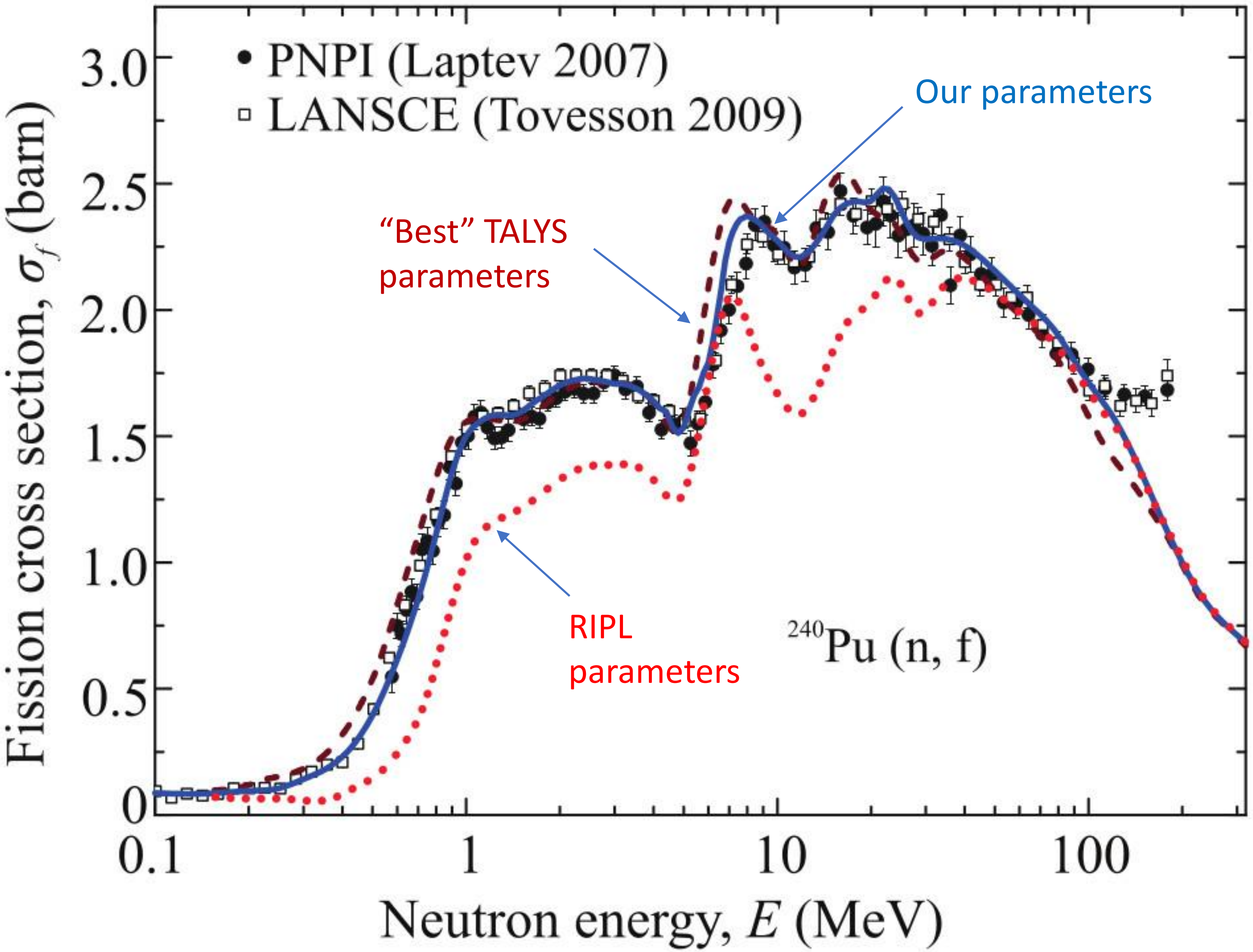
$$\frac{\hbar^2}{J_{eff}} = 7 \ keV = \frac{\hbar^2 (J_\perp - J_\parallel)}{J_\perp J_\parallel}$$

$$\frac{\hbar^2}{J_{eff}} = 5 \ keV$$

6.5 keV for TALYS parameters!

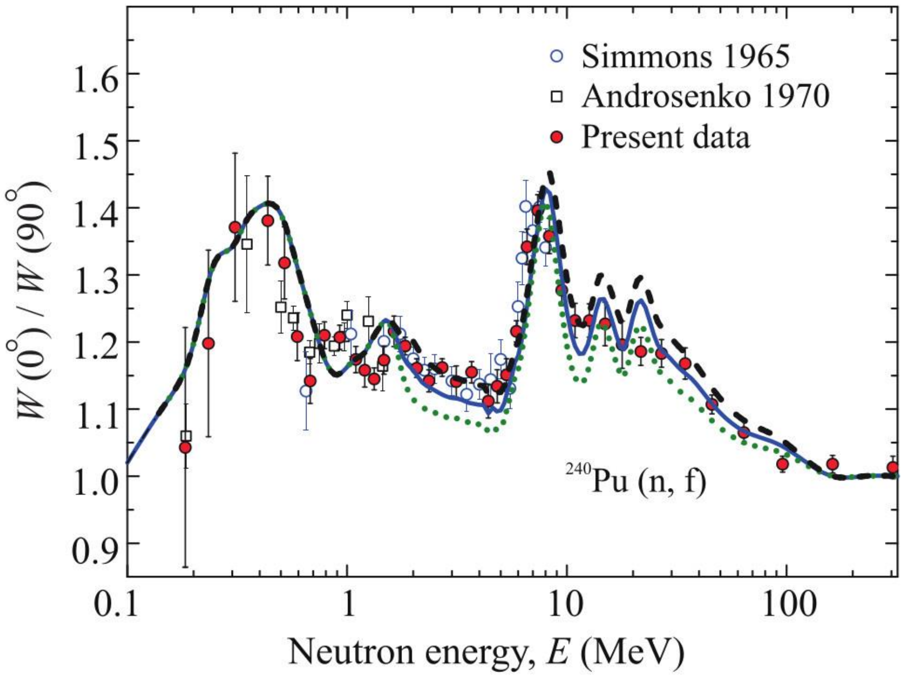
$^{240}Pu(n, f)$: Results for fission cross section

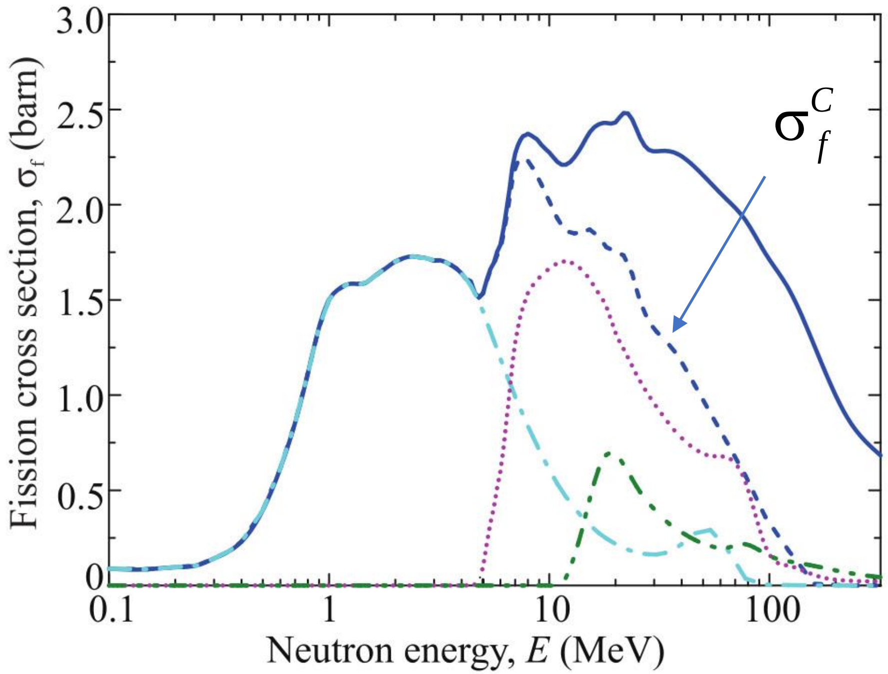
A.S. Vorobyev et al.
JETP Lett. 112,
323 (2020)



$^{240}Pu(n, f)$: Results for angular anisotropy

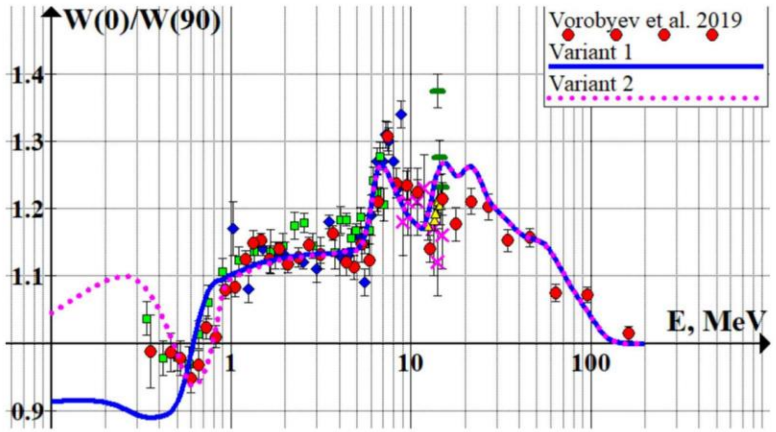
The calculated cross-section σ_f^C and the observed angular anisotropy of the fission fragments W(0)/W(90) decrease similarly at E > 20 MeV. Thus, at high energies the angular anisotropy seems be mainly related to the decay of primary compound nucleus.





Multichance fission for $n + {}^{237}$Np at $E_n = 80$ MeV:

$$\sigma_f = \underbrace{\sigma_f^{DPE}}_{\sim 80\%} + \underbrace{\sigma_f^{C}}_{\sim 20\%}, \quad a = \frac{W(0^\circ)}{W(90^\circ)} - 1 = 0.078$$



Main decay chaines of compound nucleus:

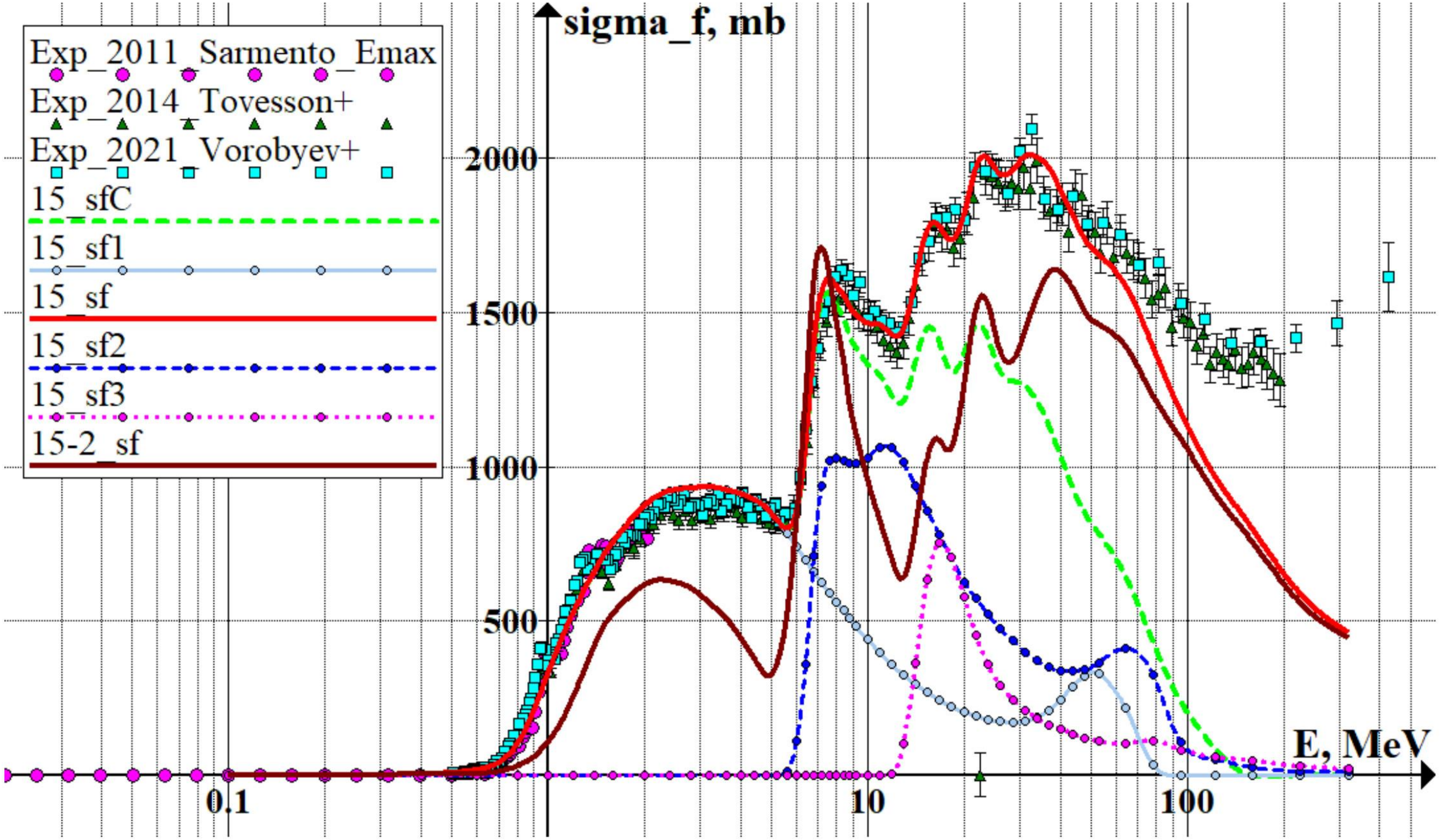
$$\begin{array}{ccccccccccccccccccc}
{}^{238}\mathbf{Np} & \to & {}^{237}\mathrm{Np} & \to & {}^{236}\mathrm{Np} & \to & {}^{235}\mathrm{Np} & \to & {}^{234}\mathrm{Np} & \to & {}^{233}\mathrm{Np} & \to & {}^{232}\mathrm{Np} & \to & {}^{231}\mathrm{Np} & \to & {}^{230}\mathrm{Np} & \to & \ldots \\
 & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & \\
 & & {}^{237}\mathrm{U} & \to & {}^{236}\mathbf{U} & \to & {}^{235}\mathbf{U} & \to & {}^{234}\mathrm{U} & \to & {}^{233}\mathbf{U} & \to & {}^{232}\mathbf{U} & \to & {}^{231}\mathbf{U} & \to & {}^{230}\mathbf{U} & \to & \ldots \\
 & & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & \\
 & & & & {}^{236}\mathrm{Pa} & \to & {}^{235}\mathrm{Pa} & \to & {}^{234}\mathbf{Pa} & \to & {}^{233}\mathrm{Pa} & \to & {}^{232}\mathrm{Pa} & \to & {}^{231}\mathrm{Pa} & \to & {}^{230}\mathrm{Pa} & \to & \ldots \\
 & & & & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & \\
 & & & & & & {}^{235}\mathrm{Th} & \to & {}^{234}\mathrm{Th} & \to & {}^{233}\mathrm{Th} & \to & {}^{232}\mathrm{Th} & \to & {}^{231}\mathrm{Th} & \to & {}^{230}\mathrm{Th} & \to & \ldots \\
 & & & & & & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & & \searrow & \\
 & & & & & & & & \ldots & \to & \ldots & \to & \ldots & \to & \ldots & \to & \ldots & \to & \ldots
\end{array}$$

8 isotopes, ^{238}Np, ^{236}U, ^{235}U, ^{233}U, ^{232}U, ^{231}U, ^{230}U, ^{234}Pa, give ~ 80 % to σ_f^C and a,

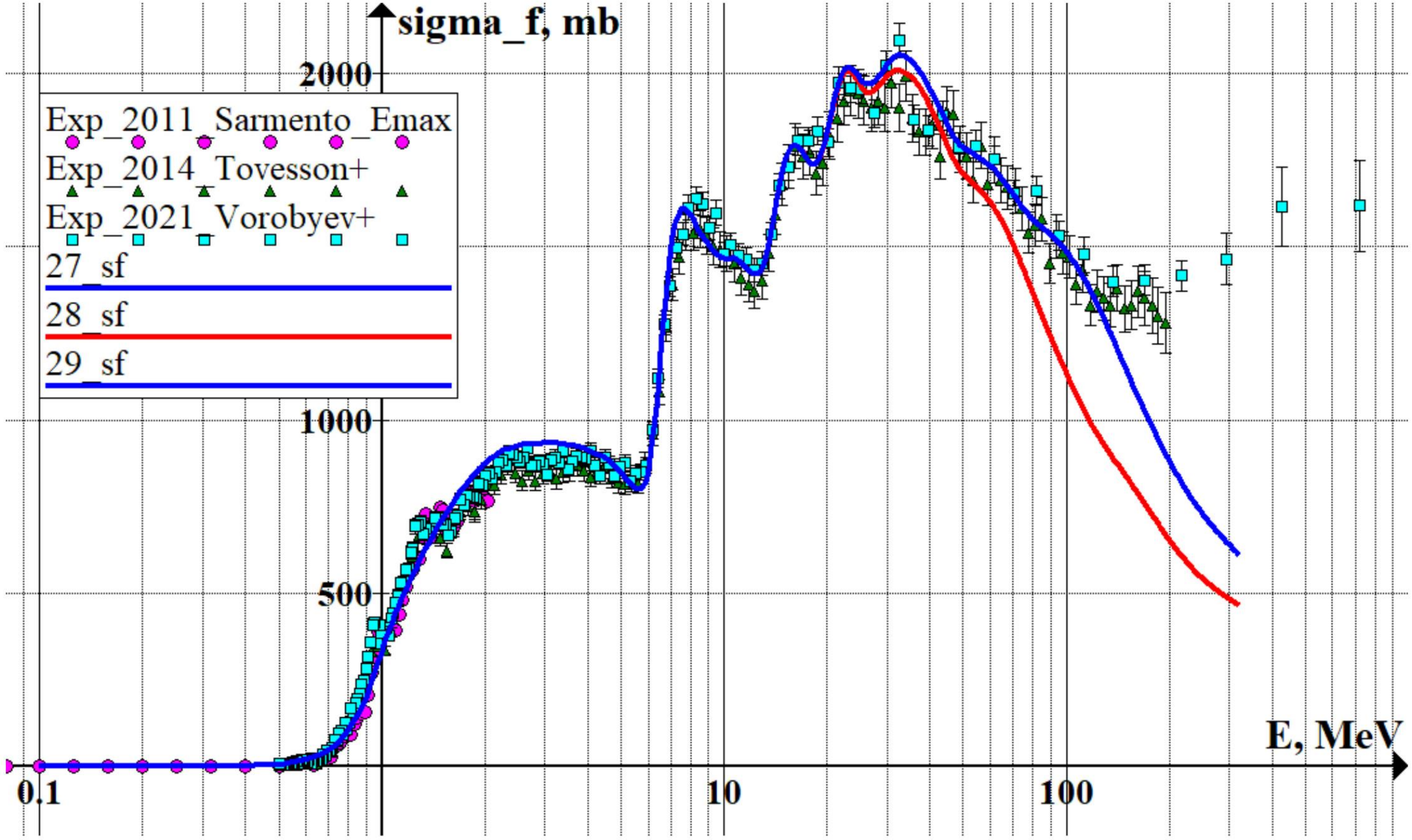
1 isotope, ^{230}**U** ($T_{1/2} = 20.23$ d), gives ~ 30 % to σ_f^C and a!

The used effective moment of inertia I_{eff} is the average of the moments of involved isotopes. Really I_{eff} depends at least on Z, N, E^*.

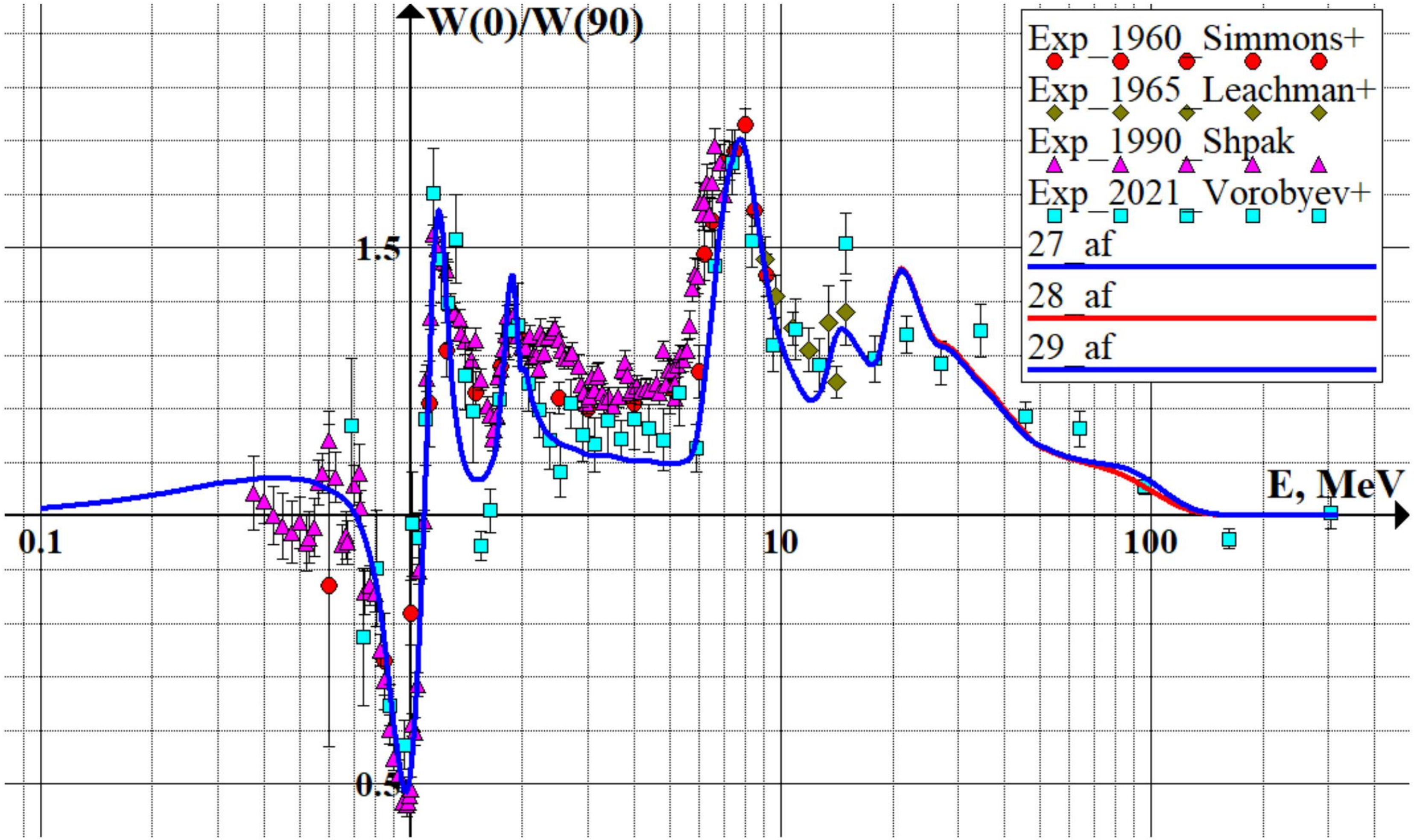
Предварительные результаты по (n,f)^{236}U



Предварительные результаты по (n,f)^{236}U



Предварительные результаты по $(n,f)^{236}U$



Заключение (чем интересны нейтроны 1-200 МэВ)

- Исследование конкуренции равновесных и предравновесных процессов посредством измерения угловой анизотропии осколков деления (деление – полностью равновесный процесс).
- Исследование характеристик деления изотопов , удалённых от линии стабильности (нейтронодефицитных ядер).
- Исследование характеристик деления ядер с высокими барьерами деления (Bi, Pb, …).
- Включение в TALYS возможности вычисления угловых распределений осколков позволит провести «ревизию» огромного массива ранее полученных результатов по угловой анизотропии осколков деления, полученных не только на нейтронах, но и на заряженных частицах. Полученная информация о переходных состояниях расширит наши возможности по вычислению сечений деления.
- Включение в TALYS возможности вычислению угловых распределений не только осколков, но и любых других частиц, в том числе гамма-квантов, расширит наше понимание каскадных ядерных процессов.

$^{240}Pu(n, f)$: Results for fission cross section

A.S. Vorobyev et al.
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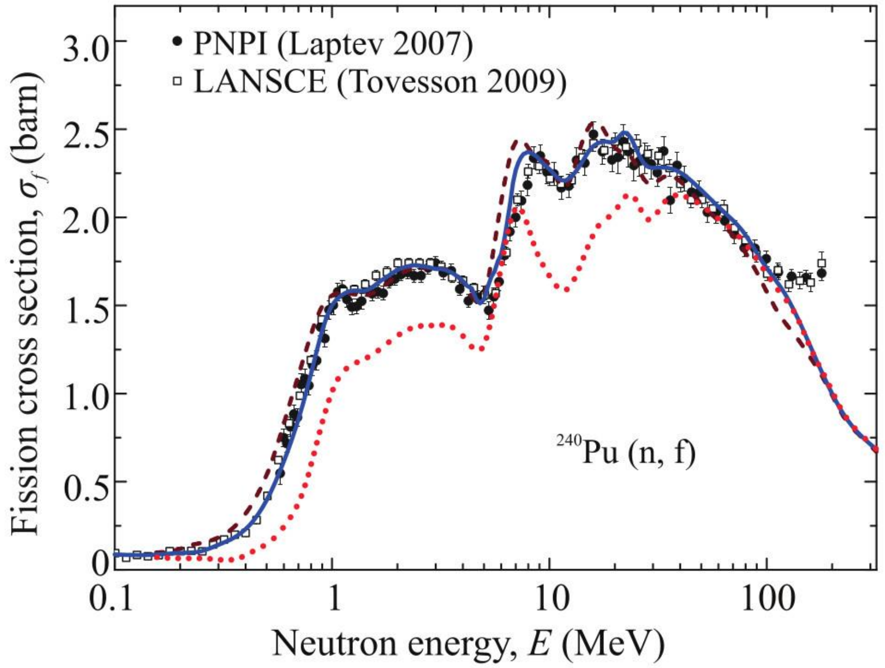

Our parameters for barriers and level densities at barriers

	1				2			
A	B	$\hbar\omega$	R_{tm}	K_{rc}	B	$\hbar\omega$	R_{tm}	K_{rc}
241	6.05	0.78	0.7	1.5	5.4	0.5	1.0	1.5
240	6.07	0.9	8.0	2.0	5.05	0.6	8.0	4.0
239	6.1	0.8	0.7	1.5	5.6	0.5	1.0	1.5
238	5.6	0.9	4.0	1.0	5.0	0.6	4.0	2.0