Prompt Fission Neutrons in Coincidence with Well-Defined Fission Fragments: A Dedicated Experiment to Disclose Their Origin

N. Carjan[1,2], M. Rizea[2]

[1] **Joint Institute for Nuclear Research, Dubna, Russia**

[2] **National Institute of Physics and Nuclear Engineering, Bucharest, Romania**

Plan

- Historical remark on the evaporation hypothesis
- Alternative explanation of the main properties of the prompt fission neutrons (PFN)
- Dynamical scission model (short presentation)
- Typical results: angular and energy distributions for a given configuration at scission (i.e., a given fragment-mass ratio and TKE) present structures
- Recent experimental data suitable for comparison
- Synopsis

Historical remark

The main properties of the prompt fission neutrons (PFN) (a) an almost Maxwellian distribution of their energies and (b) an angular distribution with respect to the light fragment direction asymmetrically peaked at 0° and 180° (i.e., $\nu(0^\circ)/\ \nu(180^\circ) > 1$)) were measured during the Manhattan Project.

The simplest explanation was immediatelly embraced: they are evaporated from *moving fission fragments*.

It is worth stressing that the nuclear evaporation theory (Weisskopf) existed at that time . What a bargain! On the contrary nothing was known about fission dynamics. So there was no choice.

Historical remark (2)

Neutron spectra were fitted with Maxwellian and Watt distributions and, as a bonus, information about the nuclear temperature of the fission fragments was extracted. Concerning the angular distribution, it turned out that what we observe is not just a kinematic anisotropy, as it was anticipated. The asymmetry with respect to 90° could not be quantitatively explained by unequal fragment velocities ($v_L > v_H$) but no problem: a parameter was added and fitted to the experimental curve. The conclusion was: for some reason the light fragment evaporates 30$\%$ more neutrons than the heavy one. Nobody has revendicated the evaporation hypothesis! Nowadays it is referred to as the "Los Alamos" model although the Madland-Nix paper was published 30 years later.

Neutrons released at scission: an alternative

The emission mechanism of scission neutrons (SN) is the diabatic coupling between the neutron degree of freedom and the changing neutron-nucleus potential during the scission process (i.e., from the neck rupture at finite radius r_{min} to the absorption of the neck stubs by the fragments). This tiny diabatic part of the fission process was investigated using the time-dependent Schrödinger equation with time-dependent potential in M. Rizea, N. Carjan, Nucl. Phys. A **909** (2013) 50.
The scission process is defined by the nuclear shapes just before (α_i) and immediatelly after scission (α_f) and by the duration ΔT of the transition between these two shapes. These quantities are not really known.
Briefly, our approach is dynamical, microscopic and quantum mechanical.

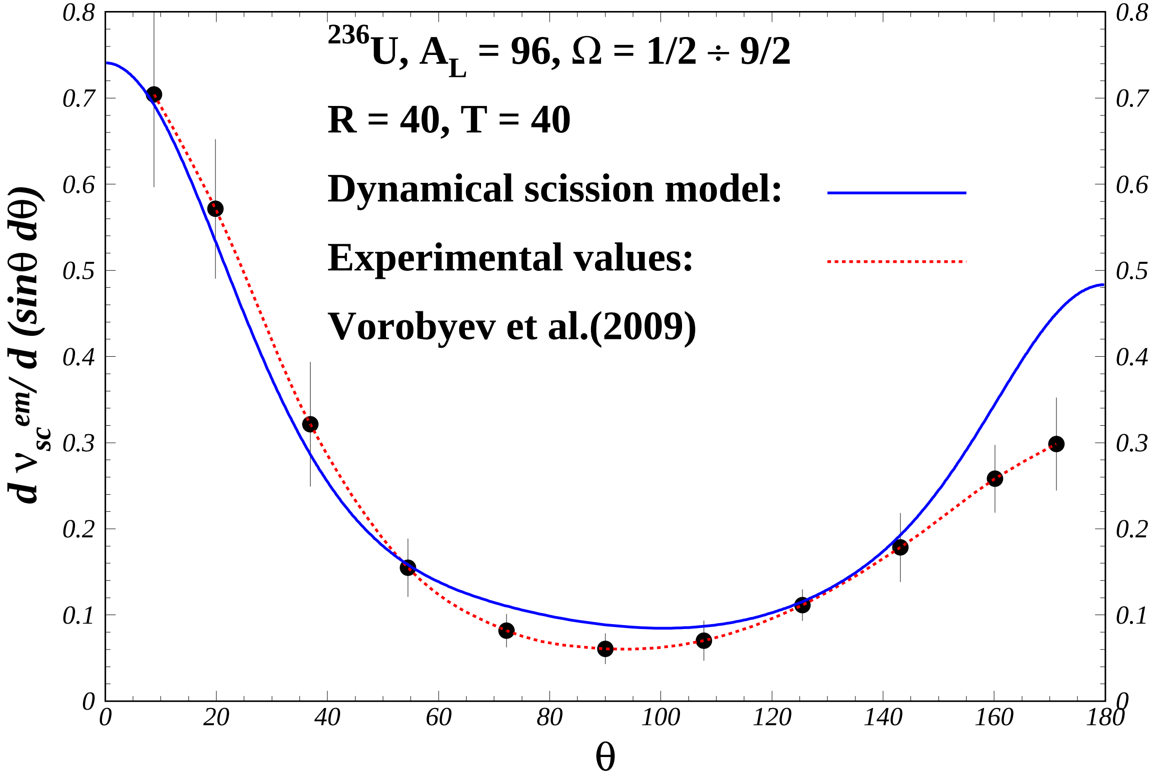
Characteristics of the scission process (best guesses)

The lower limit of ΔT should be about 5×10^{-23} sec, the time required for a Fermi level nucleon to cross a 4 fm thick neck. We consider ΔT = 1×10^{-22} and 2×10^{-22} to be realistic values. The minimum neck radius r_{min} of a fissioning nucleus is predicted to be ≈ 2 fm (by optimal scission shape method or from general considerations). We took a slightly lower value 1.6 fm; α_i=0.985 . There is no indication about the minimum distance between the surfaces of the two fragments d_{min} at the end of the scission process. We took 0.6 fm; α_f=1.001.

Next we will see that this dynamical scission model can also explain the main properties of the prompt fission neutrons. ^{236}U calculations vs $^{235}U(n_{th}, f)$ data.

N. Carjan, M. Rizea, Phys. Lett. B **747** (2015) 178.

Calculated (SN) and experimental (PFN) ang. distribution



Note the agreement of prompt neutron data with dynamical scission model calculations (except the last 2 points on the heavy fragment side).

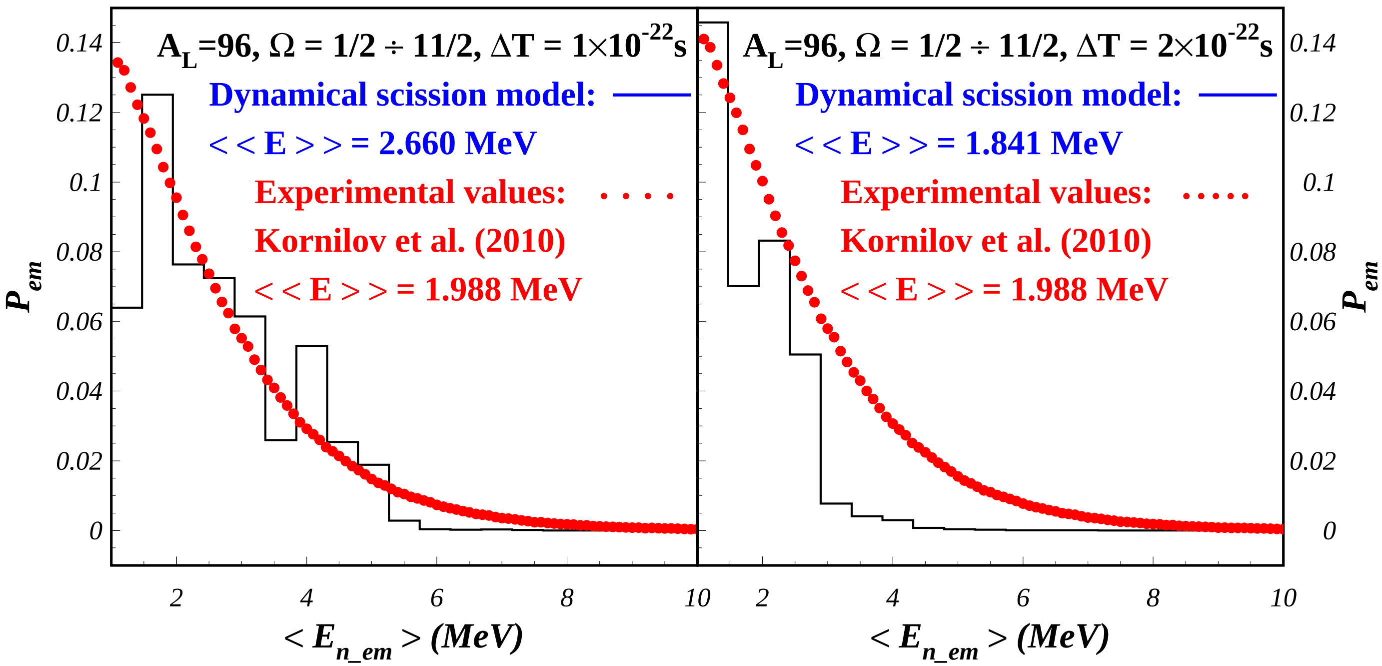
Table 1: Total number of scission neutrons released ν_{sc} for A_L=96 and ΔT=1 and number of neutrons ν_{sc}^{em} that crossed the spheres of R = 30 and 40 fm at succesive time intervals T. All times are in 10^{-22} sec.

A$_L$ / Δ **T**	$96(R=30)$		$96(R=40)$	
	ν_{sc}	ν_L/ν_H	ν_{sc}	ν_L/ν_H
1	0.561	1.075	0.551	1.123
T	ν_{sc}^{em}	ν_L^{em}/ν_H^{em}	ν_{sc}^{em}	ν_L^{em}/ν_H^{em}
10	0.118	1.424	0.043	1.562
20	0.258	1.348	0.152	1.256
30	0.363	1.402	0.247	1.281
40	0.429	1.414	0.320	1.407

The ratio ν_L^{em}/ν_H^{em} corresponds to two regions of the interval $(0, 180)$. The separation point is the center of the interval (90^0) corresponding to the point $z = 0$ of the laboratory system. They represent neutrons which move left and right with respect to a plane perpendicular to the fission axis. All states with $\Omega = 1/2, ..., 9/2$ have been taken into account. The calculated ratio at T=40 is in agreement with the experimental value 1.41. No adjustment necessary.

Histogram of the average scission-neutron energy

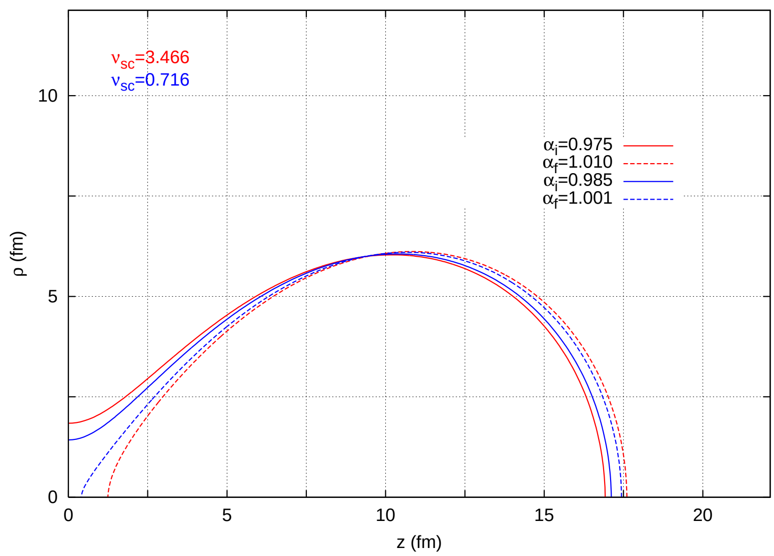
immediately after scission (α=1.001) for two transition times ΔT. Each average energy belongs to a spectrum (the calculation of which will be presented later).



The experimental prompt-neutron spectrum is also shown to compare the trends: the slope, the range and the average value.

Table 2: Dependence of ν_{sc} (for A_L=96) on r_{min} and on ΔT.

ΔT \ r_{min}	1.6 fm	1.9 fm
1×10^{-22} sec	0.551	2.538
2×10^{-22} sec	0.385	1.887



In conclusion one can say that the agreement is not only surprising but it is also better than expected for a model that uses little-known scission shapes and scission times. The values chosen are first-guess (not adjusted).

If this alternative explanation had existed in the 50's the field of fission neutrons would have probably known a different development.

Dynamical scission model: formalism

Let $|\Psi^i\rangle$, $|\Psi^f\rangle$ be the eigenfunctions corresponding to the just-before-scission and immediately-after-scission configurations respectively. In the non-adiabatic regime, the propagated wave functions $|\Psi^i(t)\rangle$ are wave packets with some positive-energy components.

The probability amplitude that a neutron occupying the state $|\Psi^i\rangle$ before scission populates a state $|\Psi^f\rangle$ after scission is

$$a_{if} = \langle\Psi^i(T)|\Psi^f\rangle = 2\pi\int\int(g_1^i(T)g_1^f + g_2^i(T)g_2^f)d\rho dz.$$

The result strongly depends on the duration T of the scission process.

Excitation energy of the fission fragments

The total occupation probability of a given final eigenstate is:

$$V_f^2 = \sum_{bound} v_i^2 |a_{if}|^2$$

where v_i^2 is the ground-state occupation probability of a given initial eigenstate. Since V_f^2 is different from v_f^2 (the ground-state value), the fragments are left in an excited state. The corresponding excitation energy at scission is:

$$E^*_{sc} = 2 \sum_{bound\ states} (V_f^2 - v_f^2) e_f .$$

The factor of 2 is due to the spin degeneracy.

Neutrons emitted at scission

One can also calculate the multiplicity of the neutrons released during scission:

$$\nu_{sc} = 2 \sum_{bound} v_i^2 (\sum_{unbound} |a_{if}|^2).$$

A quantity that can clarify the emission mechanism of the scission neutrons is the probability density i.e., the spatial distribution of the emission points at t=T

$$S_{em}(\rho, z) = 2 * \sum_{bound} v_i^2 |\Psi^i_{em}(\rho, z, T)|^2,$$

$$|\Psi^i_{em}\rangle = |\Psi^i(T)\rangle - \sum_{bound\ states} a_{if}|\Psi^f\rangle$$

is the unbound part of the wave packet that will leave the nucleus and asymptotically will describe the emitted scission neutron.
Similarly, the current density

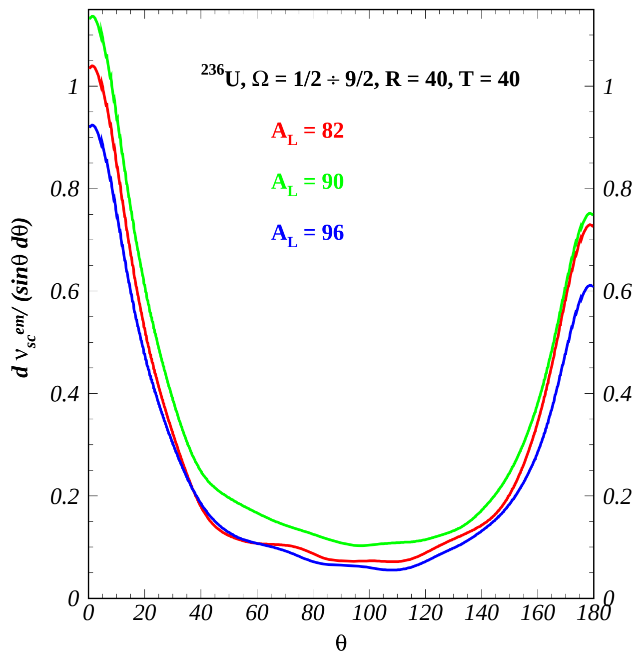
$$\bar{D}_{em}(\rho, z) = \frac{i\hbar}{\mu}\sum_i v_i^2(f^i\bar{\nabla}f^{i*} - f^{i*}\bar{\nabla}f^i), \tag{1}$$

with $f^i = |\Psi^i_{em}\rangle$, provides the distribution of the average directions of motion of the unbound neutrons at t=T.
These two quantities influence the amount of neutrons that are reabsorbed, scattered or left unaffected by the fragments.

Angular distribution - formula

- The number of neutrons that leave a sphere of radius R (around the fissioning nucleus) in a solid angle $d\Omega$ and in a time interval dt is:
$d\nu_{sc}^{em} = \bar{J}_{em}(R, \theta, t)\bar{n}(R, \theta, t)R^2 dt d\Omega.$
- The angular distribution is given by the integral with respect to t of the above quantity. The upper limit should in principle be ∞. In practice we can reach only a finite value t_{max}.
- The total number of emitted neutrons ν_{sc}^{em} at t_{max} is obtained by a further integration with respect to θ ($d\Omega = sin\theta d\theta$).
A factor of 4π also appears due to the integration over the angle ϕ and to the spin degeneracy.

Dynamical scission model: typical angular distributions



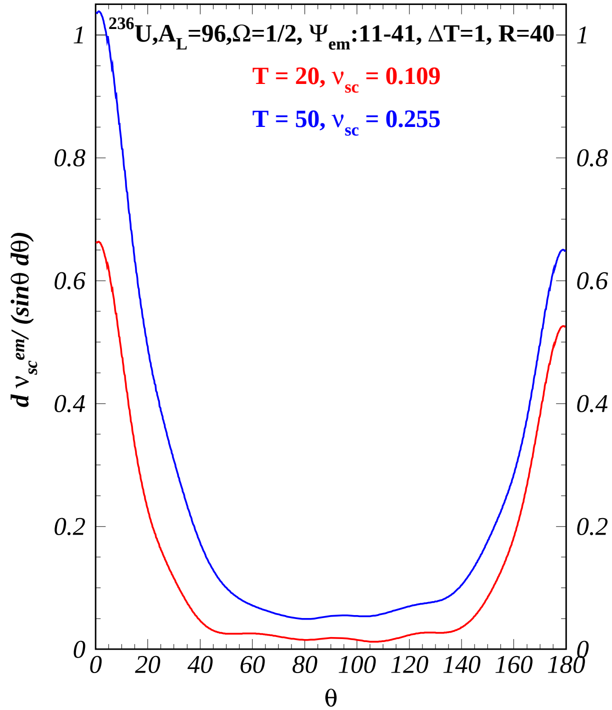
The angular distributions for all mass asymmetries (82/154, 90/146, 96/140) display weak oscillations (from 50° to 150°).

They are most probable due to scattering of the neutrons on the just born fragments. The maxima and minima are typical of a non-monotonic deflection function (rainbow effect).

The amplitudes of the oscillations are small partly because the initial neutron source has tails inside the fragments, i.e., it is not totally localized between the fragments.

Angular distribution corresponding to Ω=1/2 states

at 2 and 5 $\times 10^{-21}$ sec.

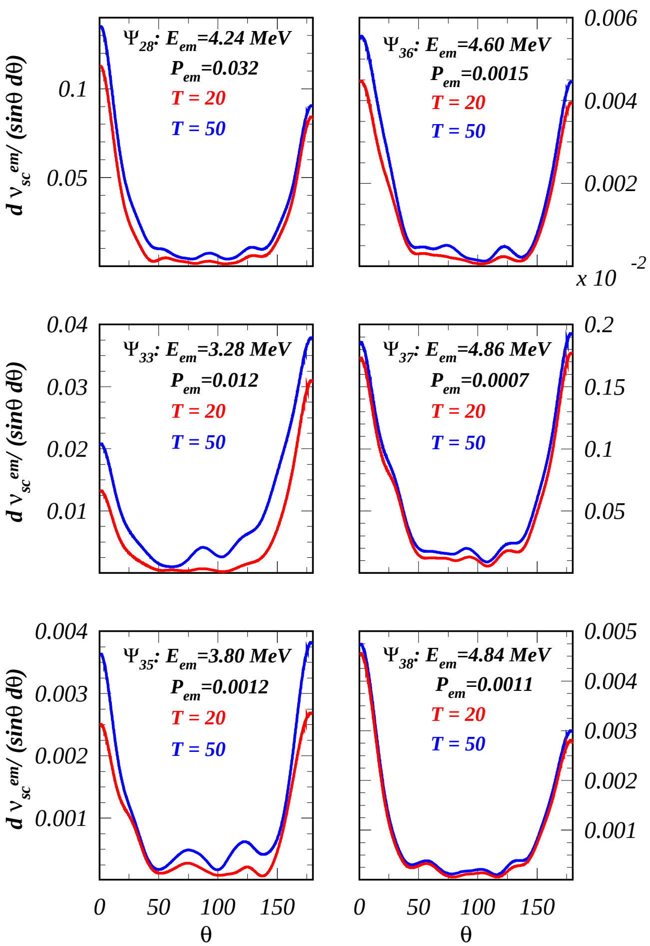


30 states with Ω=1/2 contribute. The oscillations are small.

Due to the diabaticity of the scission process, each neutron of the fissioning nucleus is more or less emitted and therefore each contributes to the total angular and energy distributions.

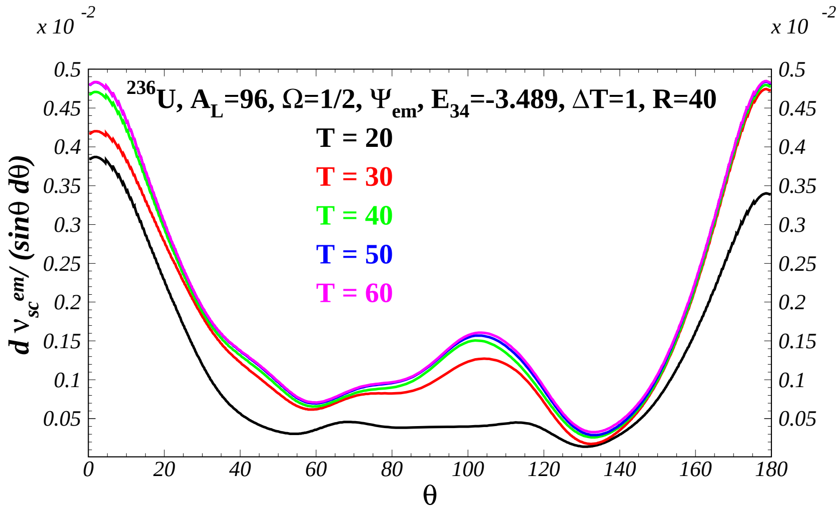
To underline the microscopic nature of the process, we will show in the next slide some of these contributions.

Angular distributions for single neutron states



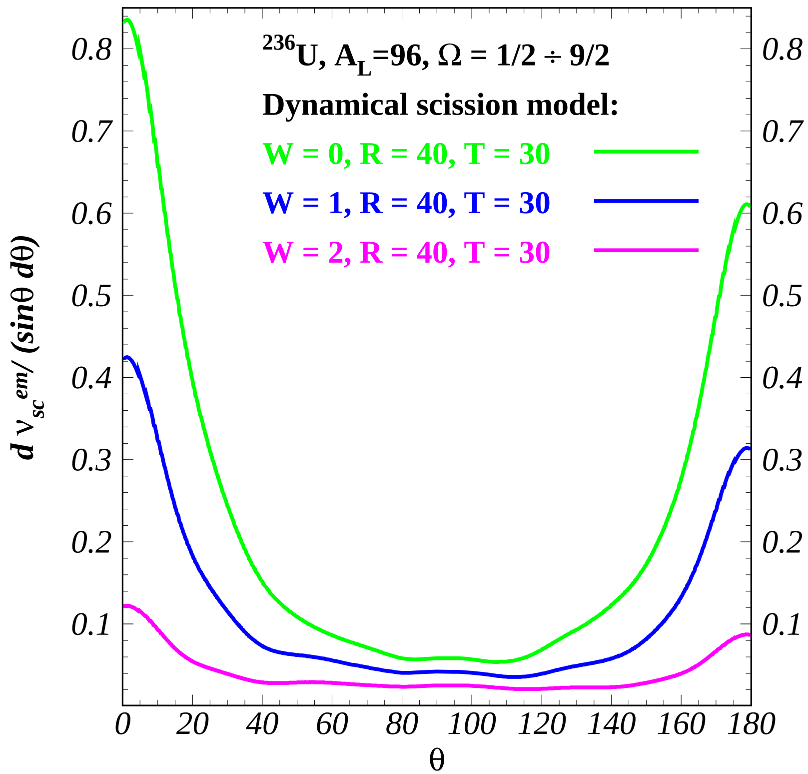
The angular distributions are very diferent from one state to the other. Most of them are peaked in the direction of the L-fragment but some prefer the H-fragment and some move with equal probability in both directions. When summing over all states the oscillations are reduced.

Time evolution of the structures



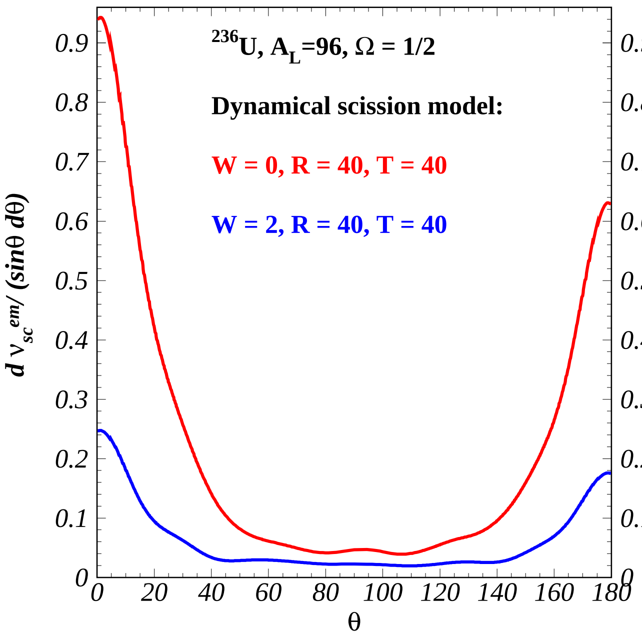
At T=6$\times 10^{-21}$ sec saturation is attained. Very large grids are however necessary to reach so long times.

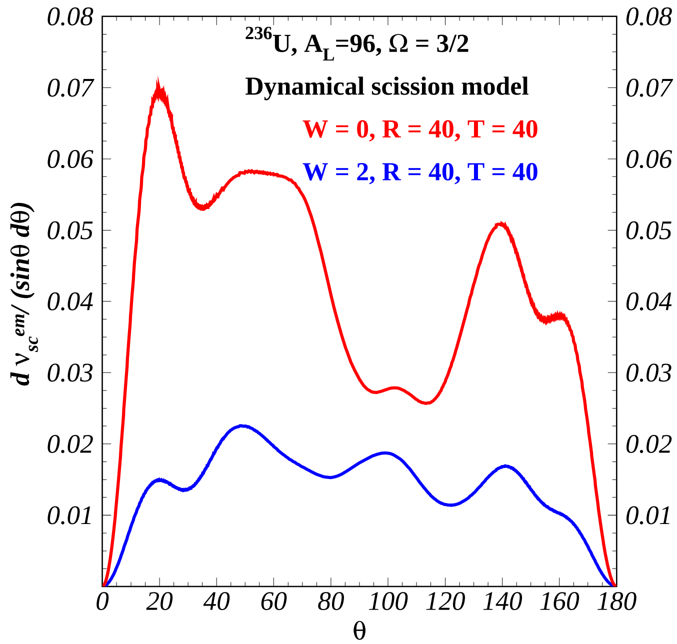
Role of the imaginary potential (W_0=0,1 and 2 MeV)



As expected: it reduces the scission neutron flux in the direction of the fragments. The neutrons moving perpendicular are less affected. The oscillations are slightly attenuated.

Ω has a strong influence on the angular distribution

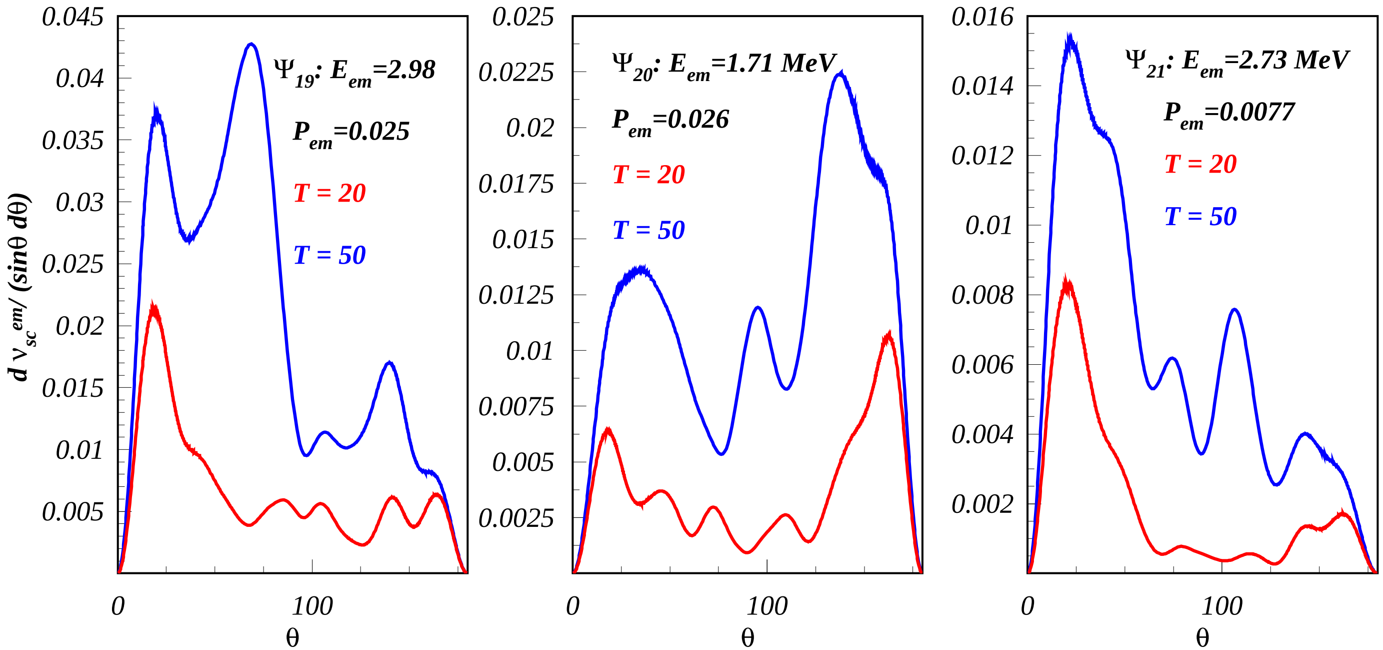




Neutrons with Ω=1/2 represent the majority (65 %). Their angular momentum being perpendicular to the fission axis, they mainly move along this axis.

Neutrons with Ω=3/2 represent 25 % of the scission neutrons. They cover all angles (except 0° and 180°). They exhibit intense structures.

Angular distributions for individual states Ω=3/2

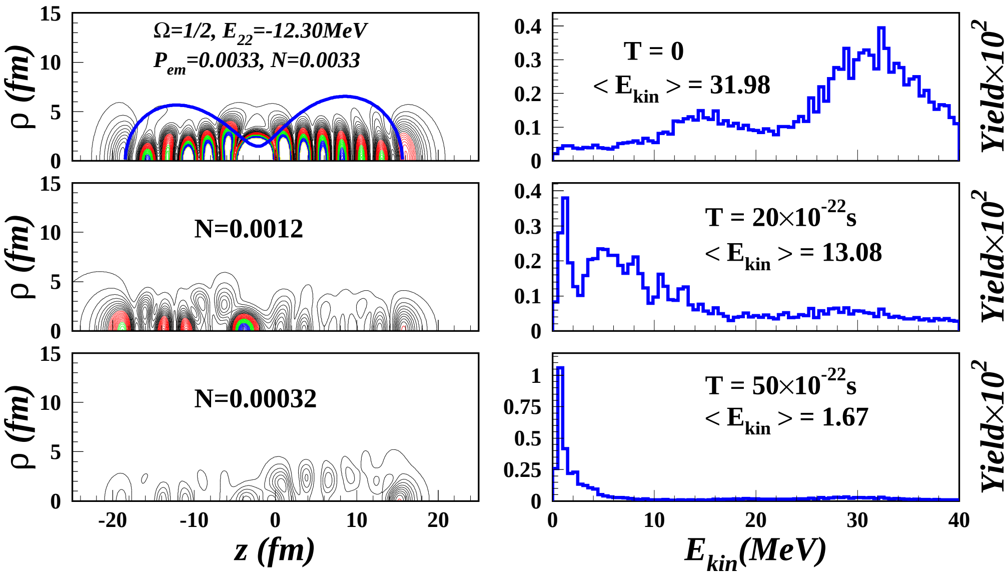


They are again very different from one state to the other

Energy spectrum of the scission neutrons

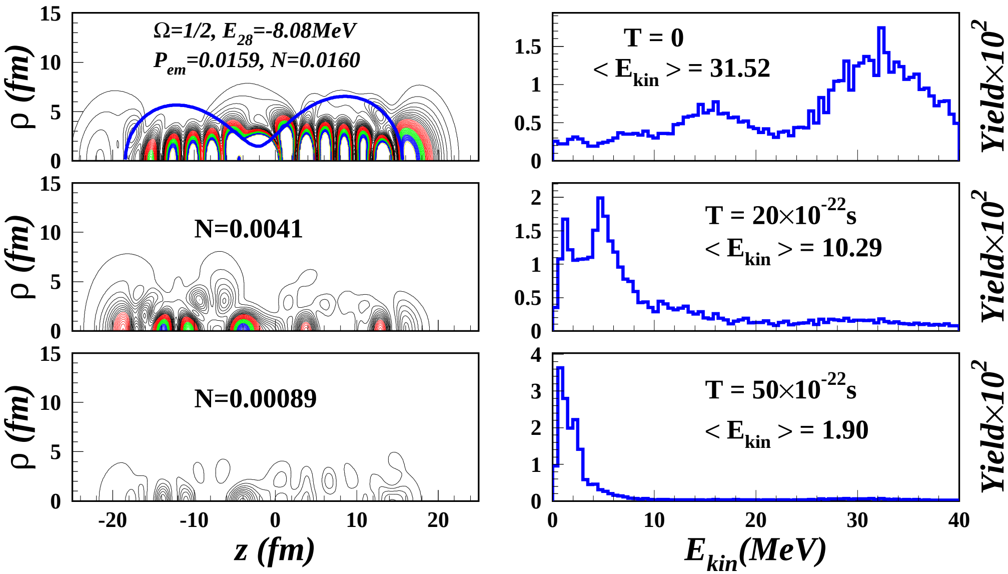
Main difficulty: it is necessary to propagate in time the unbound part $|\Psi^i_{em}\rangle$ of each neutron wave packet until it completely leaves the fission fragments. It is a hard numerical task that requires very large (ρ,z) grids and very long CPU times. We were able to go until $\Delta T + T_{max}$ with $T_{max} = 50 \times 10^{-22}$ sec. Since the separation of the fragments is slower than the neutron emission, for the sake of simplicity, we keep the fragments in their configuration at ΔT.
The Fourier transforms of these wave packets are calculated in order to get the corresponding momentum distributions which lead to the kinetic energy distributions. To obtain the whole kinetic energy spectrum for a fixed mass asymmetry, one has to sum the single spectra over all occupied states and all Ω values.

Post-Scission Evolution of the Unbound Neutrons



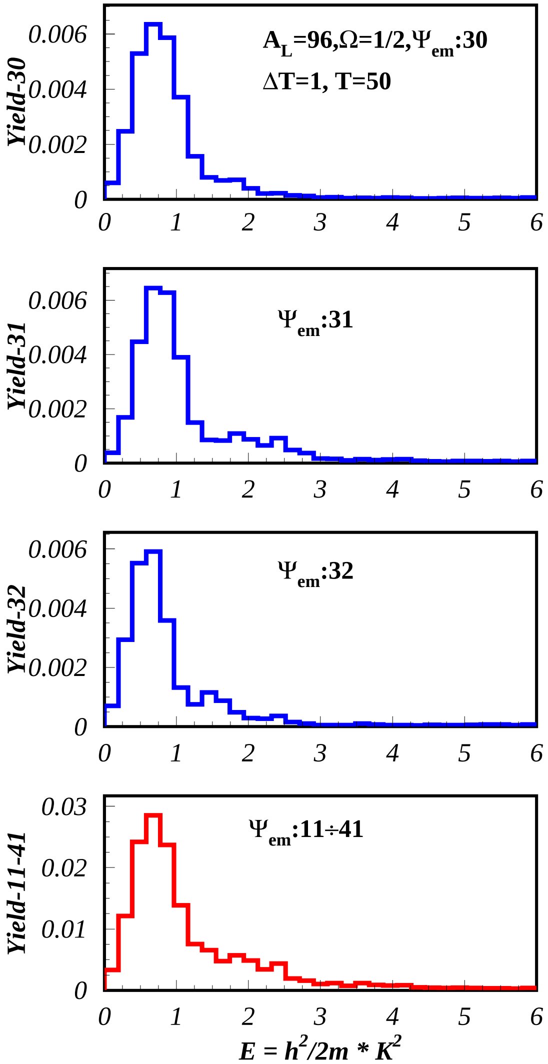
At T=0 the released neutron is mainly localized in the neck region but it covers the whole fissioning system. Its energy can reach values of the order of the potential depth V_0. With increasing time (T = 20 and 50×10^{-22} sec) the amplitude of the wavefunctions diminishes and the E_{kin}-distribution is shifted to lower values showing that the neutron is leaving the nucleus.

Post-Scission Evolution of the Unbound Neutrons (2)



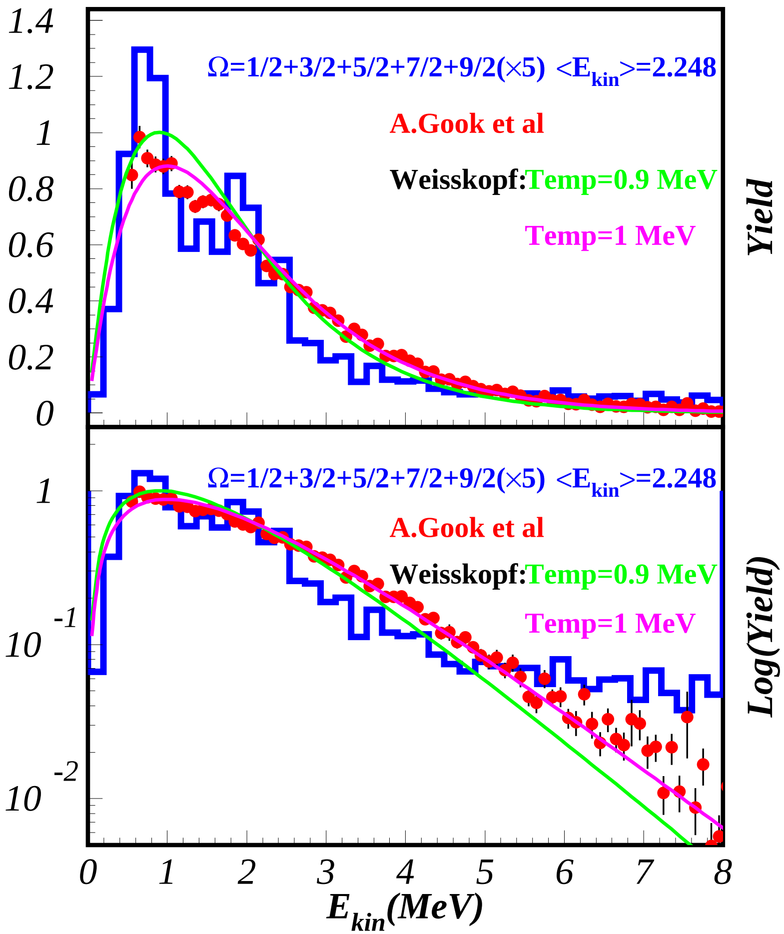
These single spectra are characterized by a peak at low energies (below 2 MeV) plus a short tail towards higher energies. At very large times the neutron should be completely emitted. Due to numerical limitations, we cannot reach this situation: at T_{max} the neutron still has 10% probability of being inside the fragments.

Kinetic energy distribution for Ω=1/2 neutron states



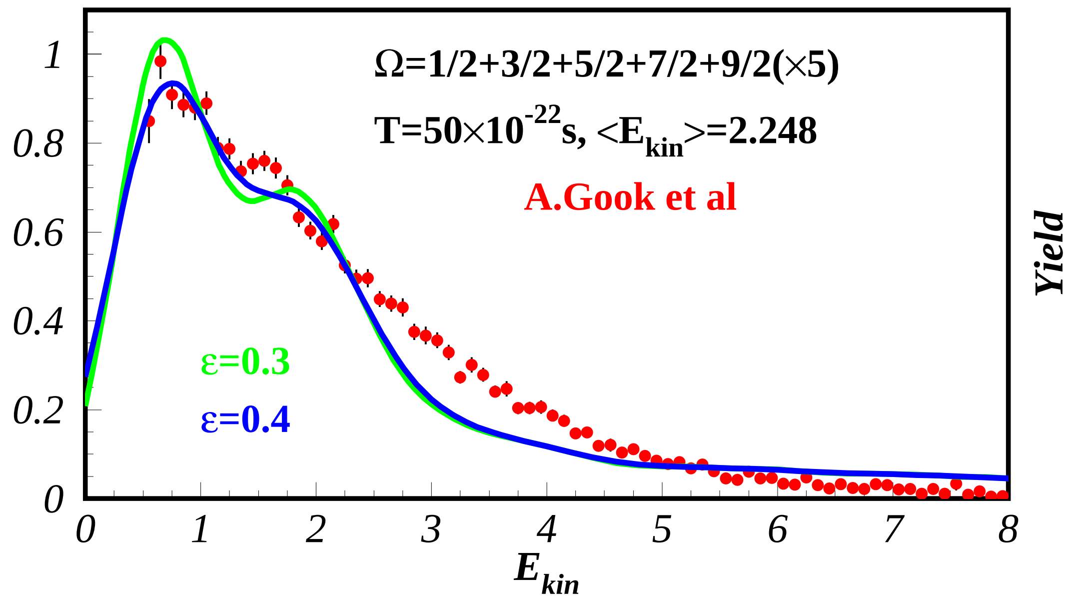
The total distribution (red/bottom) is a finite weighted sum (30) of individual quasi-gaussian distributions with different mean values and widths. Three such examples are plotted (blue/above). For this reason it cannot be smooth .

Scission-Neutron Spectrum



To obtain the whole kinetic energy spectrum for a fixed mass asymmetry, one has to sum the single spectra over all occupied states and all Ω values. The result is compared with recent experimental data from the reaction $^{235}U(n_{th}, f)$. Two typical evaporation spectra characterized by nuclear temperatures $Temp = 1.0$ and 0.9 MeV are also shown.

Scission Neutron Spectrum Convoluted



Both the data and the calculation are not smooth. However the data do not oscillate as much as the calculations. One reason is that the data are affected by a finite energy resolution. A resolution between 0.3 and 0.4 MeV brings the amplitudes of the oscillations into better agreement.

Experimental angular resolution

To compare with the experimental data one has to fold with the angular resolution function:

$$\frac{d\sigma}{d\theta}|_{\theta=\theta_0} = \int_{-\infty}^{\infty} \frac{d\sigma}{d\theta} r(\theta, \theta_0) d\sigma$$

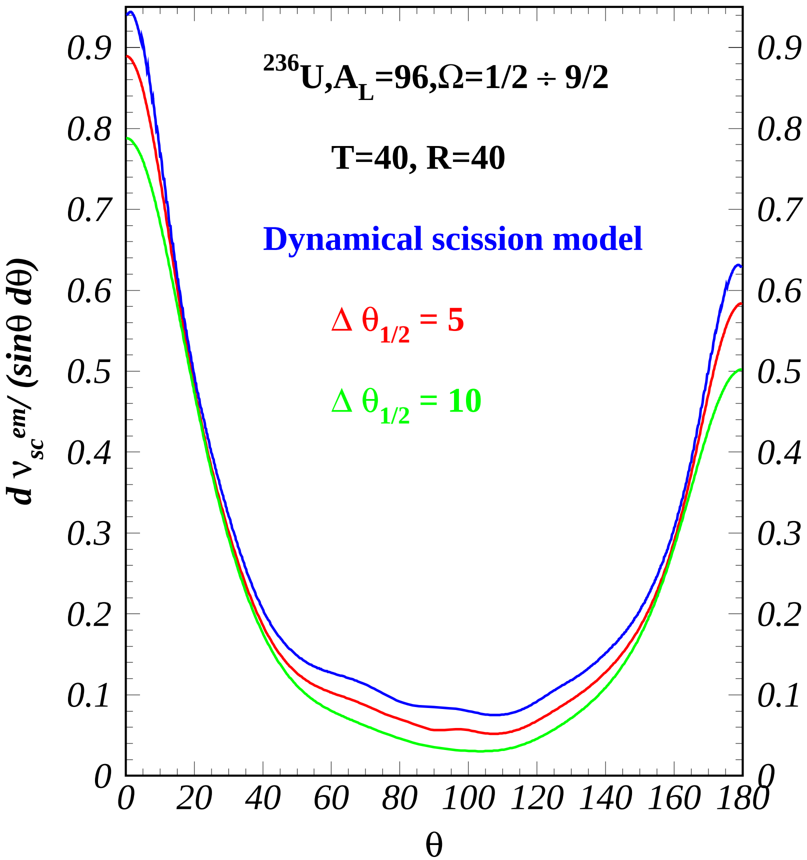
where

$$r(\theta, \theta_0) = \frac{1}{\sqrt{2\pi\epsilon}} \exp\left[-\frac{(\theta - \theta_0)^2}{2\epsilon^2}\right]$$

and $\frac{d\sigma}{d\theta}$ is the angular distribution. The value of ϵ is obtained via the half width:

$$\epsilon\sqrt{2\ln 2} = \Delta\theta_{1/2}.$$

Convolution with the angular resolution function

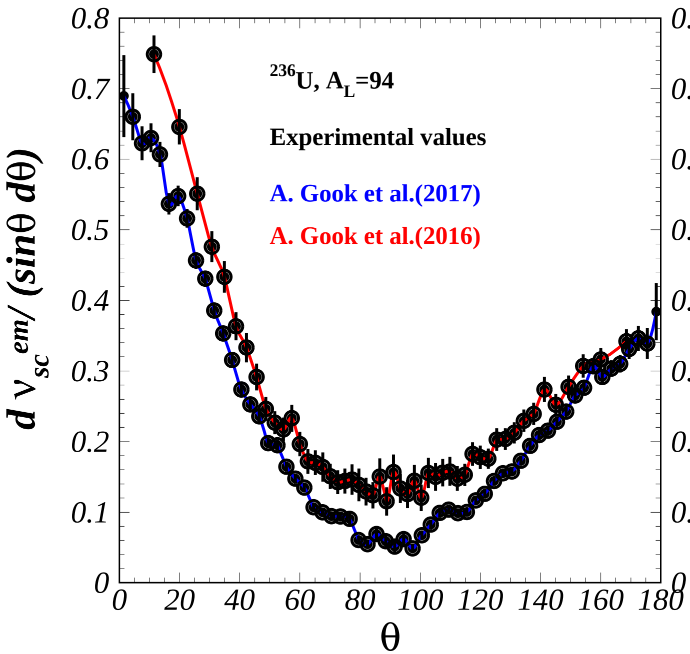


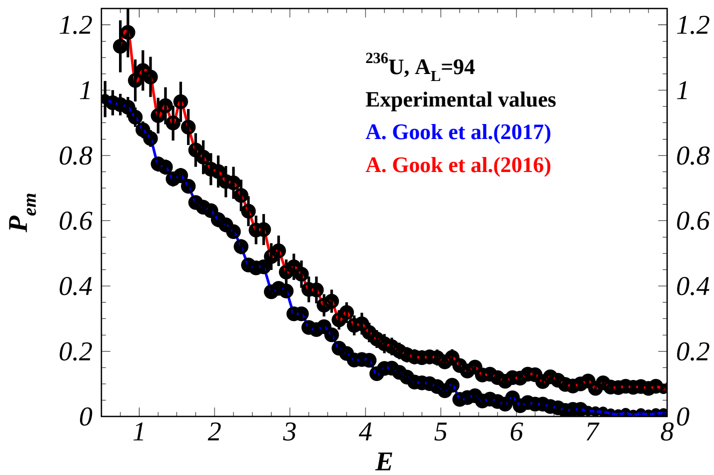
Confrontation with experimental data

Phenomenological approaches to the scission process are always applied to a given scission configuration at a time. Our results are not an exception. They correspond to a given Cassini oval that describes a pre-scission shape with a given left-right asymmetry and a given D_{cm}. So, in principle, one cannot directly compare them with experiment. The best one can do is to select data for a given fragment mass ratio and a given TKE and assume that the range of D_{cm} that contribute to a fixed TKE is narrow.

Recent data obtained with improved angular (7 deg) and fragment-mass (3 amu) resolutions and good statistics can be used for this purpose: A. Gook, F.-J. Hambsch, et al., private communication

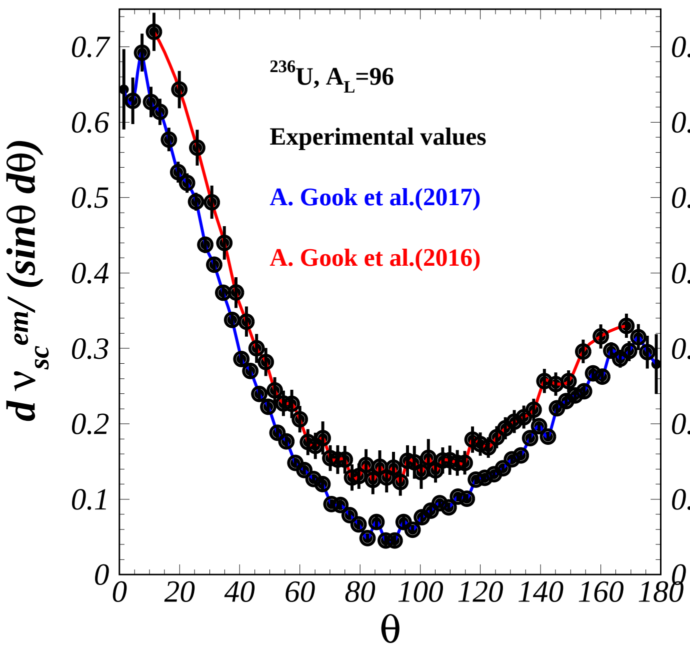
New data: ang and en distr for a given mass (A_L=94)

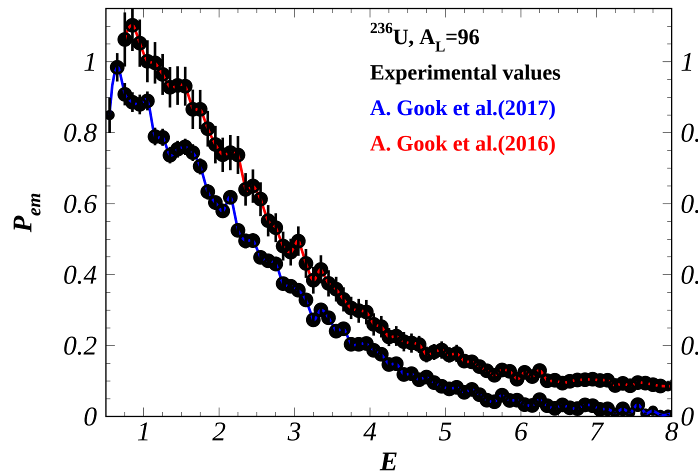




If the oscillations are real they can only be due to the proximity of the fragments at the moment of emission (left) and to the finite number of states that contributes (right).

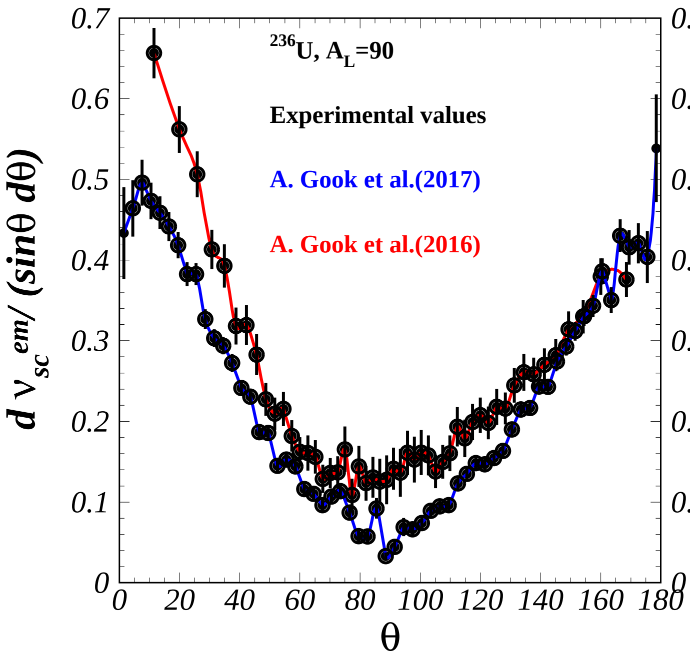
New data: ang and en distr for a given mass (A_L=96)

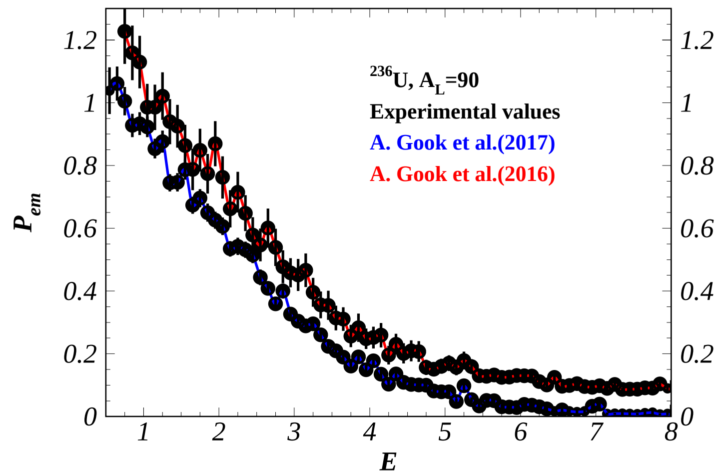




If the oscillations are real they can only be due to the proximity of the fragments at the moment of emission (left) and to the finite number of states that contributes (right).

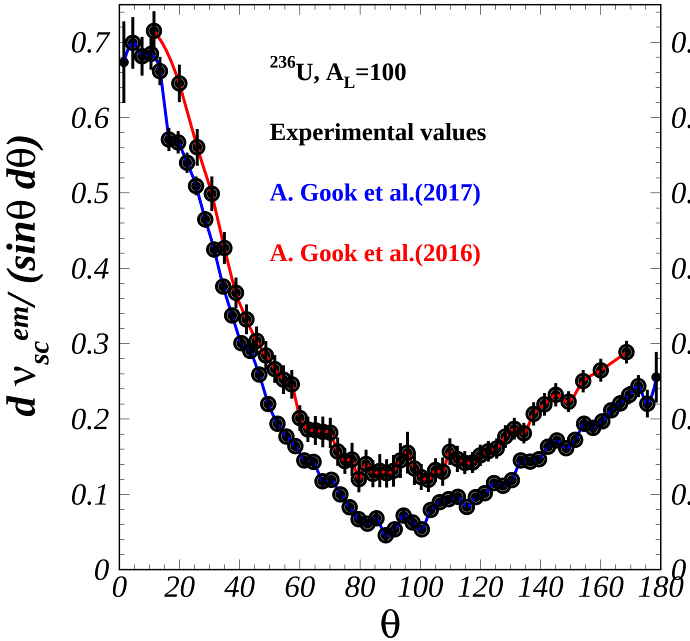
New data: ang and en distr for a given mass (A_L=90)

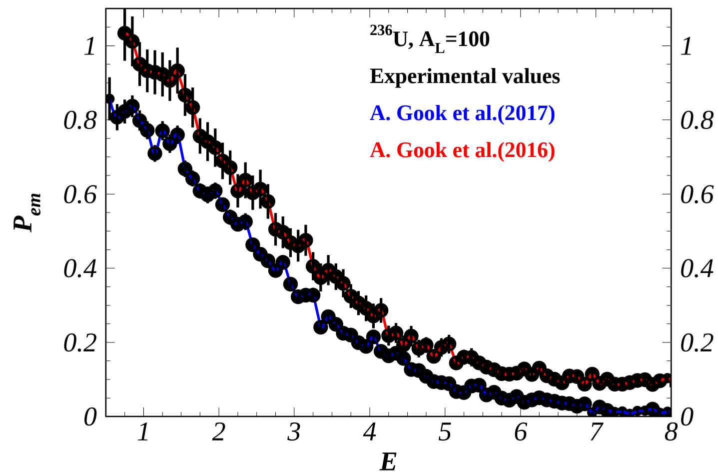




Increasing the number of events by a factor of 3 doesn't wash out the stuctures; on the contrary: they become more pronounced. This proves that they are real.

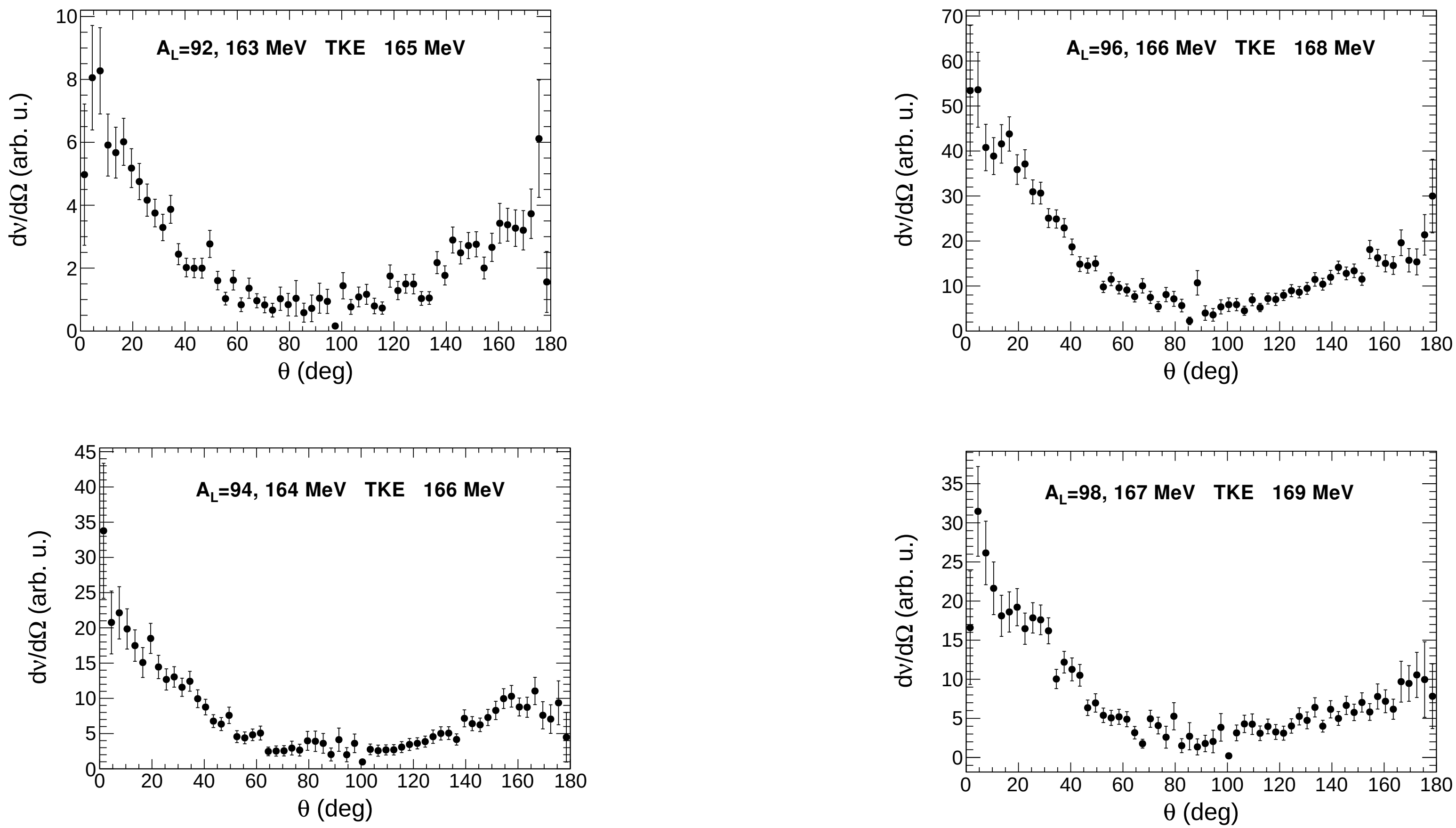
New data: ang and en distr for a given mass (A_L=100)





Increasing the number of events by a factor of 3 doesn't wash out the stuctures; on the contrary: they become more pronounced. This proves that they are real.

Unique data: PFN ang distr for a given mass and TKE



These distributions contain about 2000 events

Synopsis

There is nothing wrong with the evaporation except that it is put first. When trying to understand the origin of PFN one should think in chronological order. First comes scission. If this process is diabatic, there is no doubt that neutrons will be released (as are protons and α particles). The amount of scission neutrons depends on the speed and on the length of the scission jump. Some will be reabsorbed when they cross the fragments but the rest will leave the fissioning system during the acceleration stage.

Synopsis (2)

Our calculations showed that the emission takes time: after 4$\times 10^{-21}$ sec only 65% of the released neutrons left the sphere of radius R=40 fm. However by the time the fragments reach 90% of their TKE (i.e., 10^{-20} sec) almost all scission neutrons left. Now the fragments are well apart, somewhat excited and fully equilibrated; they will evaporate neutrons and γ rays.
$\Rightarrow$ the condition that all PFN are evaporated (as commonly believed) is an adiabatic scission process that leaves the primary fragments well deformed. We estimated that the corresponding transition time should be 5$\times 10^{-22}$ i.e., one order of magnitude larger than the shortest possible transition time. N. Carjan, M. Rizea, Int. J. Mod. Phys. E **21** (2012) 1250031.

Synopsis (3)

There is *apriori* no reason to exclude such a long time but then we should figure out why scission p's and α's are emitted.

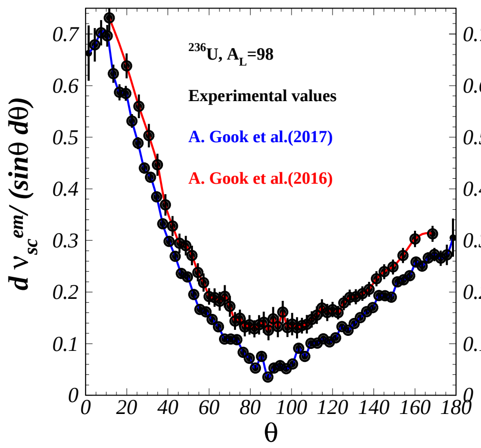
It is also possible that the large majority of PFN are released at scission. For this to happen we need the scission process: (a) to be diabatic (as in the present study) and (b) to leave the primary fragments close to their ground-state deformation (longer jump than in the present study). Even in this case the primary fragments are left excited due to the same diabatic coupling that is responsible for releasing scission neutrons. This energy will be used to emit prompt γ rays. Some excitation can also be generated through re-absorption of released neutrons by the fragments at the beginning of their acceleration. In principle this energy will be used to evaporate n's and γ's

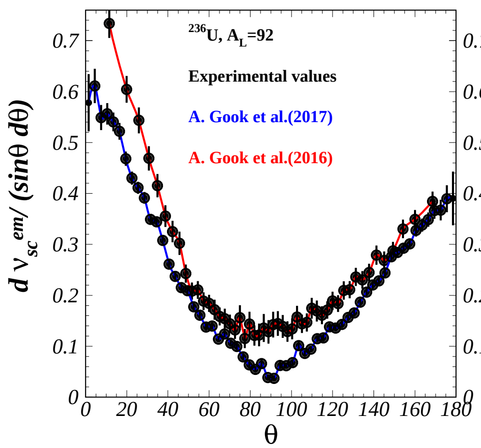
Final remark

Clarifying the role of scission neutrons inside prompt fission neutrons has a win-win outcome. Irrespective of the answer we will deduce exclusive information about the last fission stage such as: its degree of adiabaticity and the nuclear configurations involved.

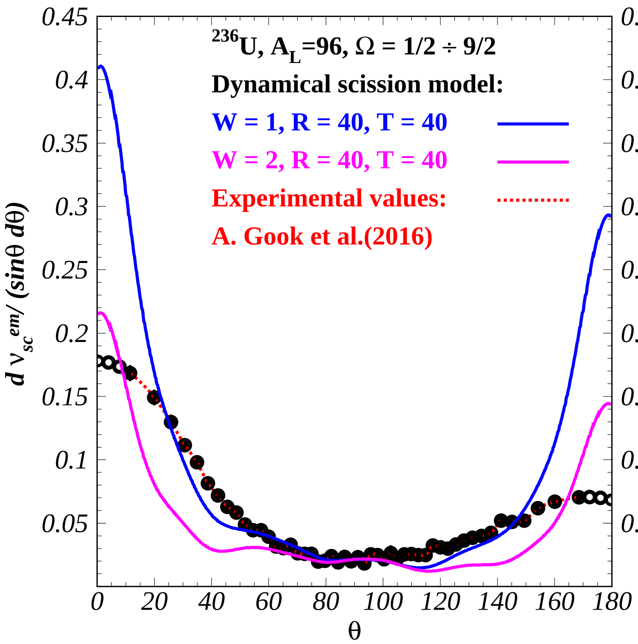
Gross structures in the angular and energy distributions are caracteristic patterns of the scission neutrons. They are due to the proximity of the fragments at the moment of their emission and to the finite number of neutrons that contibute. They should definitely be further investigated.

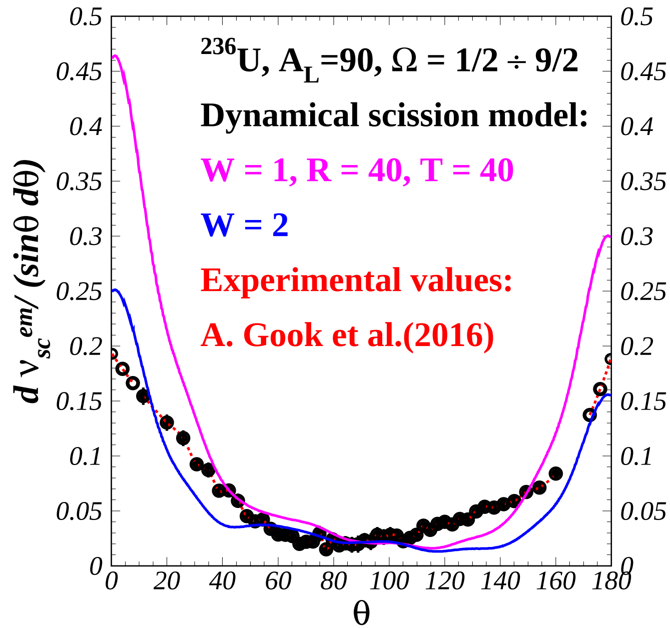
Recent data: PFN ang and energy distr for a given mass





Calculated (SN) and measured (PFN) structures juxtaposed

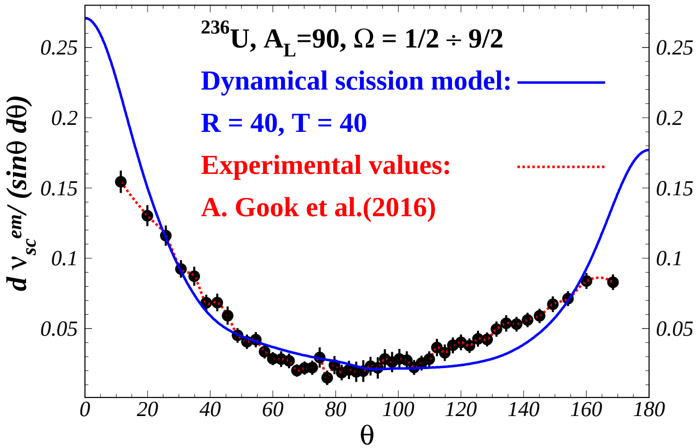


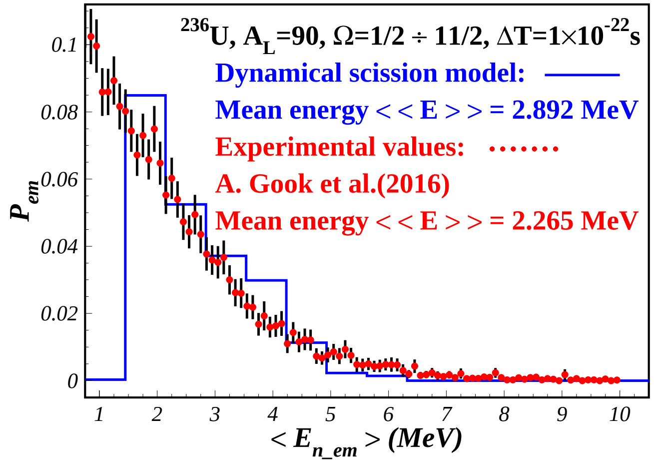


The amplitude of the oscillations between 50° and 150° are comparable. A stronger absorption ($W >$ 2 MeV) is required in the direction of the fragments.

Same qualitative agreement at A_L=90 as at A_L=96.

Dynamical scission model results for A_L/A_H = 90/146

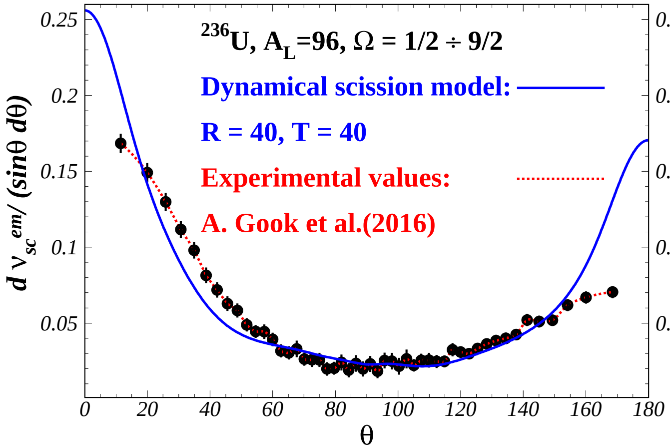


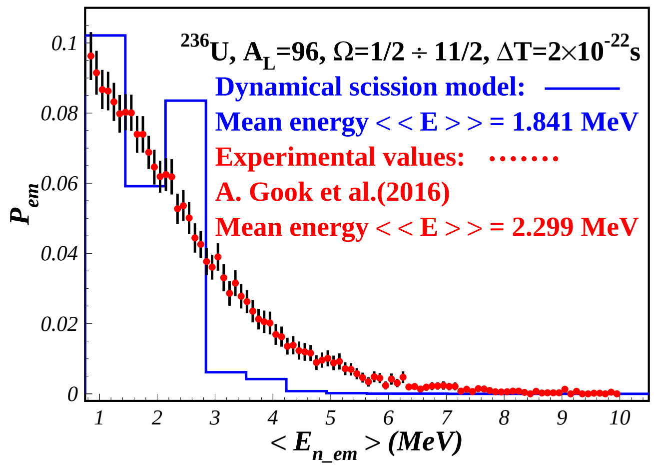


Recent data (with improved angular resolution and a given mass ratio) reveal structures. Statistics? Not sure!
In the direction of the fragments the data are rather flat (more on the HF side).

So far we could not estimate the energy spectrum but we could calculate the average kinetic energies of the neutrons emitted from each initial state i. $< E_n^i >= \langle \hat{\Psi}^i_{em}(\Delta T)|\hat{\mathcal{H}}(\alpha_f)|\hat{\Psi}^i_{em}(\Delta T)\rangle$.

DSM results for A_L/A_H = 96/140

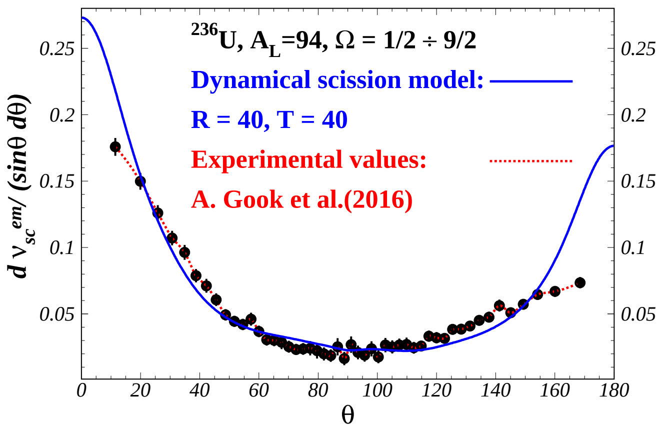


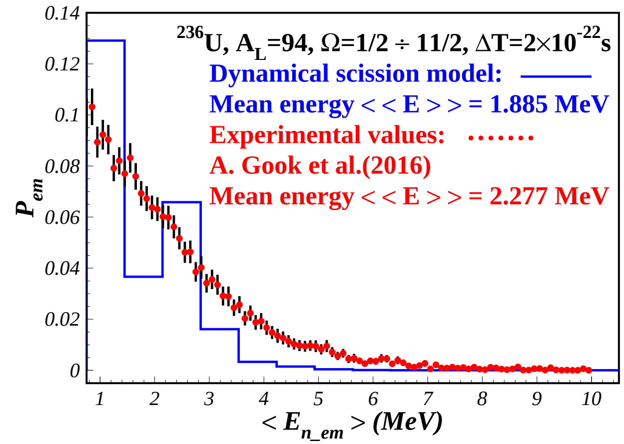


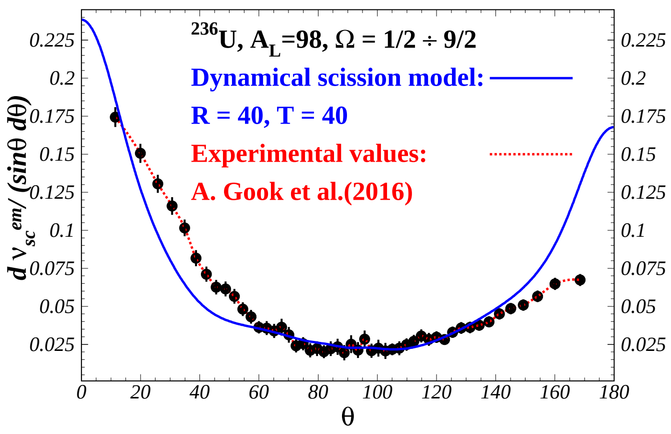
Same questions: are the oscillations real? if yes they can be due to scission-neutron scattering on fragments; the deviations around 0° and 180° can be signs of reabsorption.

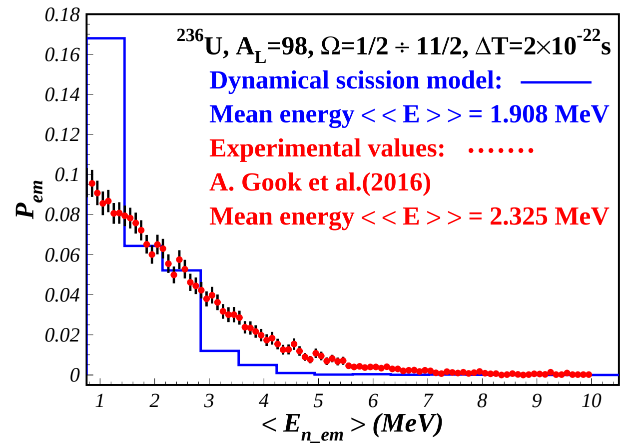
As in the previous slide the histogram represent the distribution of the average kinetic energies of the neutrons emitted from each initial state and not the full spectrum.

Results for A_L/A_H = 94/142 and 98/138



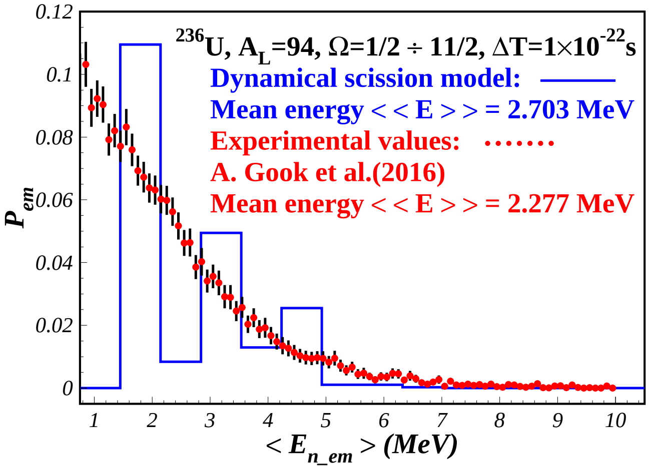


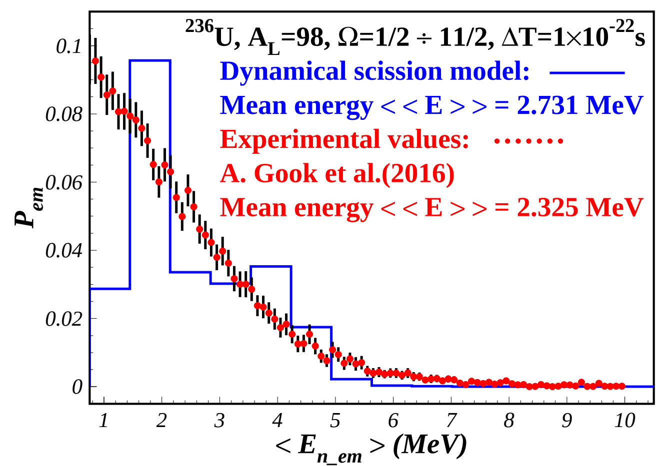


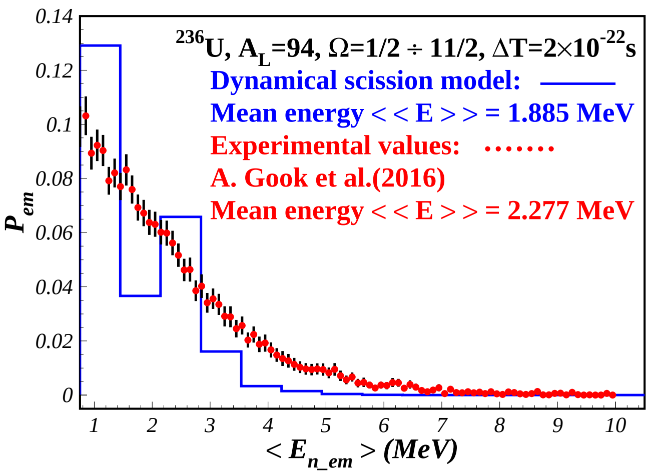


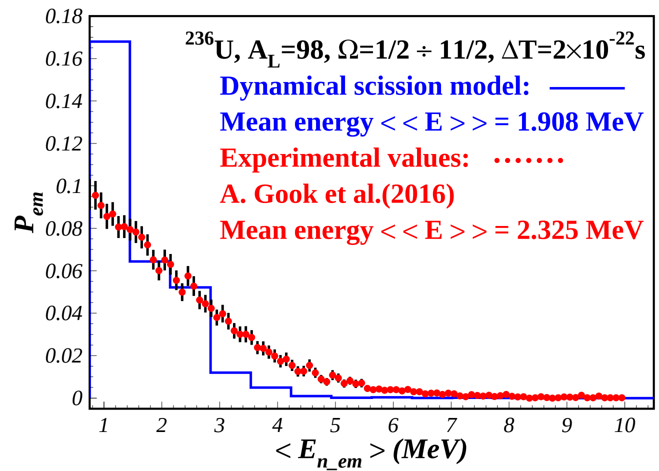
Effect of the transition time ΔT on the energy spectrum

A_L/A_H = 94/142 A_L/A_H = 98/138





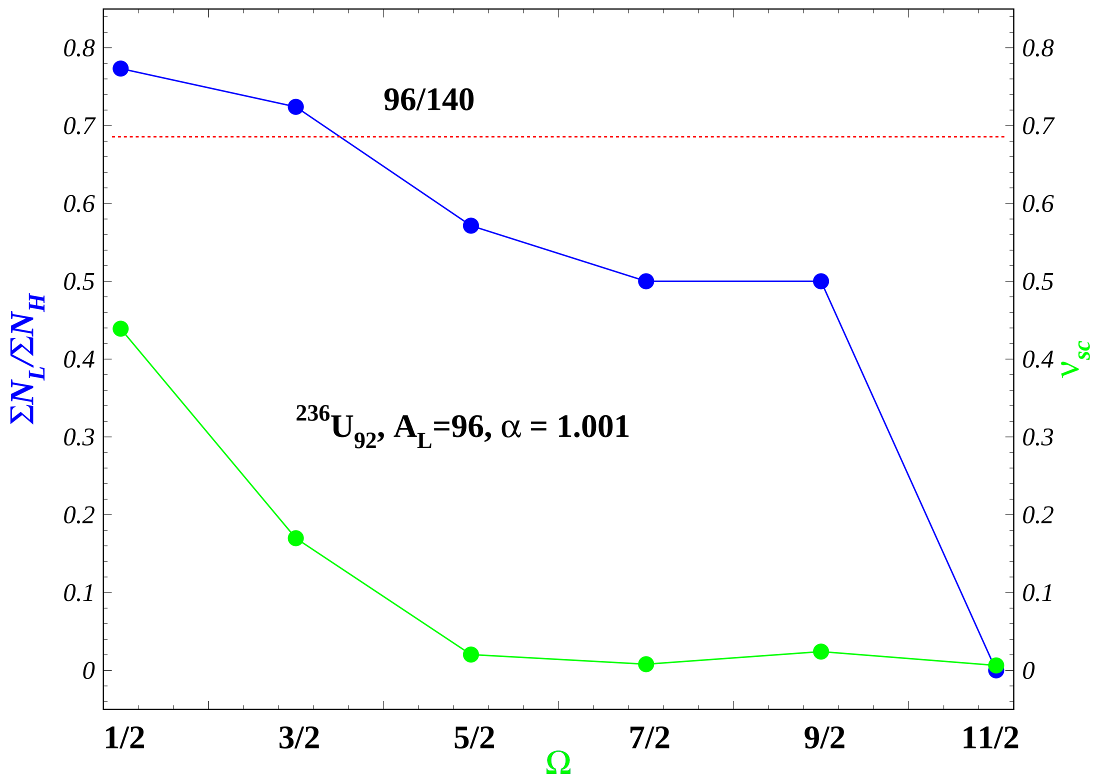




Faster is the neck rupture more energy is transfered to the neutrons

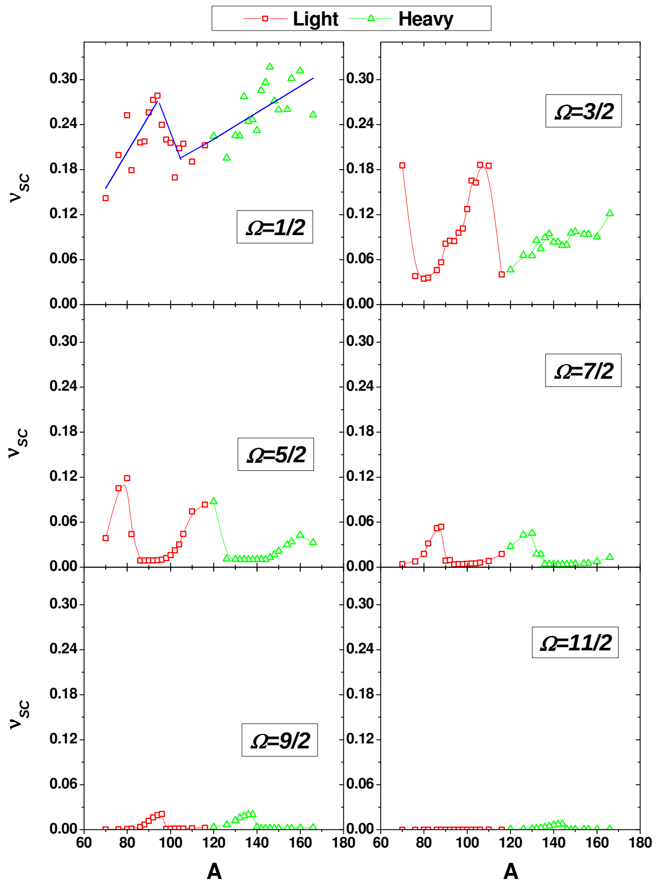
Distribution of neutrons with given Ω between L and H :

Uneven. It is a possible reason why the light fragment emits more neutrons. It is a trivial property of the Nilsson orbitals in asymmetric double-well potentials.



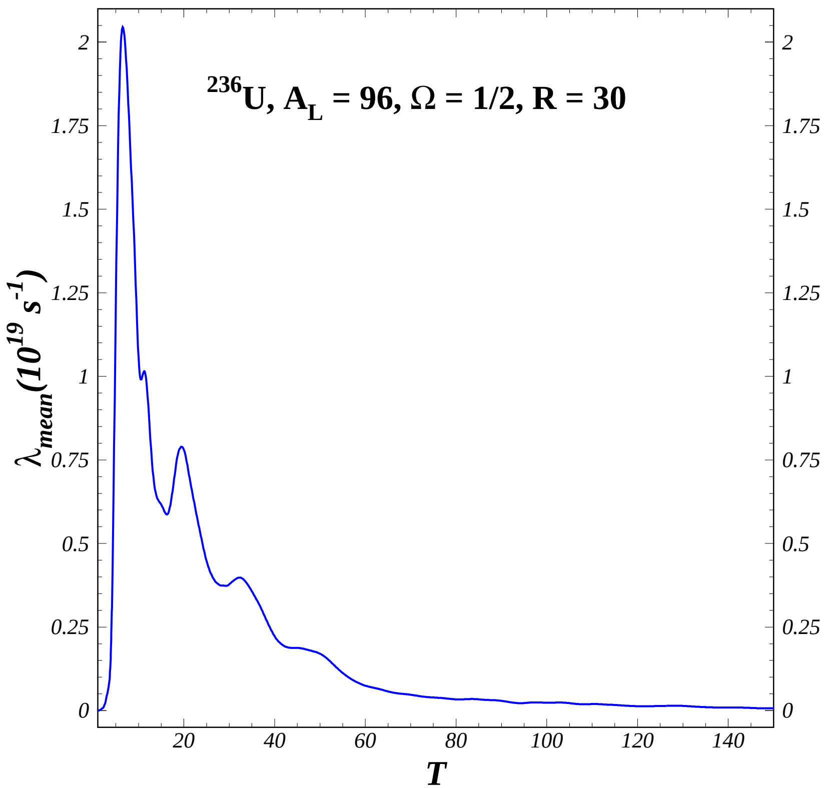
Nothing to do with deformation or temperature.

Neutron multiplicity function of fission-fragment mass



The 'saw-tooth' here is a nuclear structure effect not related to the partition of the excitation energy Calculation performed in the sudden approximation (ΔT=0): N.Carjan, F.-J.Hambsch, M.Rizea, O.Serot, Phys.Rev.C85(2012)044601

Scission-neutron time-dependent decay-rate



On notices oscillations that reflect a pulsed-emission. Most neutrons are emitted between 5 and 20 $\times 10^{-22} s$ after scission. For T $> 6 \times 10^{-21} s$ the decay rate is almost constant $\rightarrow$ tunnelling from a quasistationary state.

Remarks on angular distribution

The angular distribution of the neutrons emitted at scission is calculated starting with initial conditions given by a realistic scission model that is dynamical, microscopic and quantum mechanical. It uses nuclear configurations at scission that are appropriate for the main fission mode in the $^{235}U(n_{th}, f)$ reaction. Although the neutrons are mainly released in the interfragment region, they do not move perpendicular to the fission axis but are drained into the fragments (more into the light one) and finally leave the fissioning system through its tips. They therefore move along the fission axis with an average velocity larger than the velocity of the fully accelerated fragments. The ratio ν_L/ν_H calculated for A_L=96 is close to the experimental value (1.41) averaged over all fragment pairs.

Remarks on energy distribution and average multiplicity

Although all SN are emitted by the same mechanism, their energies can be very different depending on the single-particle state they originate from. The values of their average kinetic energies span a large interval from 1 MeV to 10 MeV with exponentially decreasing probabilities as in the PFN experimental spectrum. The measured average energy of the PFN spectrum (2.0 MeV) can be reproduced with a transition time a little shorter than 2×10^{-22} sec.

There is also no problem to explain the total average multiplicity since this quantity is very sensitive to the minimum neck radius r_{min} chosen and to a less extent also to the duration Δ T of the scission process.

General remarks

Unusual process : simultaneous partial emission of all neutrons present in a fissioning nucleus at scission.
Unusual approach: time-dependent shell-model.
Unexpected agreement: with measured properties of prompt fission-neutrons.
$\Rightarrow$ It is a viable alternative to the evaporation hypothesis.
Limitations: due to the complexity of the calculations we were not so far able to:
1)Use a larger numerical grid than: $\rho_{max} = z_{max} = 42 fm$; but TBC were implemented at the numerical boundary.
2)Propagate the wave packet of the unbound neutrons longer than: $4 \times 10^{-21} s$; however the majority of neutrons have left the system by then.