Shells, Anti-shells and Modes

in fission of pre-actinides to actinides

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Review on old and new ideas and experiments related to the notion of modes in nuclear fission

Shells and Anti-Shells

Nuclear Masses are parameterized in the

Liquid Drop Model (LDM) by

$$M(A,Z) = a_V A + a_S A^{2/3} + a_C Z^2/A^{1/3} + a_I (N-Z)^2/A - \delta(A)$$

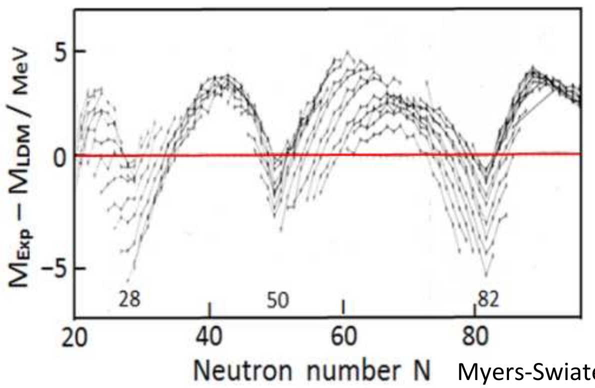
volume, surface, Coulomb, symmetry, pairing

Compare experimental mass with LDM mass

$$\boxed{\delta W = M_{exp} - M_{LDM}}$$

LDM with $\delta W = 0$ averages over N ranges where nuclei are

stronger or lesser bound and hence more or less stable:



Myers-Swiatecki 1966

Periodic fluctuations of nuclear stability are explained by the **shell model:**

in a central nuclear potential the density of energy levels to be occupied by nucleons is fluctuating: regions of nucleon numbers with high and low density of levels are alternating

Shells and Anti-Shells

For nuclei with bunched occupation levels the total energy (mass) is lower than in the LDM and stability is higher than in LDM

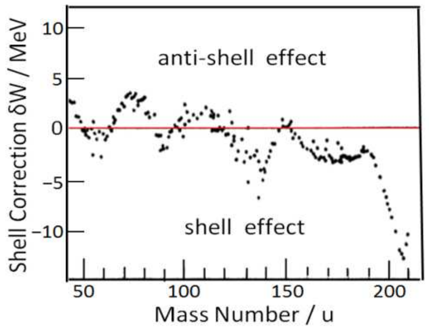
Shell Effect for $\delta W < 0$

For nuclei with low density of occupation states the total energy (mass) is higher than in the LDM and stability is lower than in LDM.

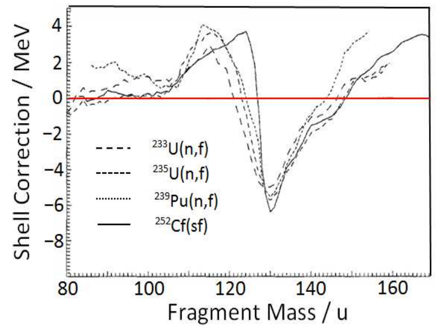
Anti-Shell effect for $\delta W > 0$

Shell corrections

for stable nuclei and fission fragments



Ignatyuk 1975

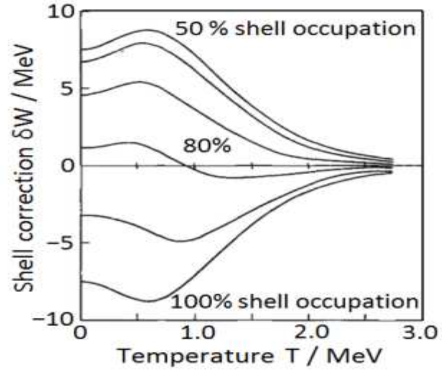


Ruben 1991

Shell corrections *vs* temperature

Shell corrections δW fade away at increasing temperature.

For decreasing occupation of a shell the correction δW turns from Shell into Anti-Shell effect.

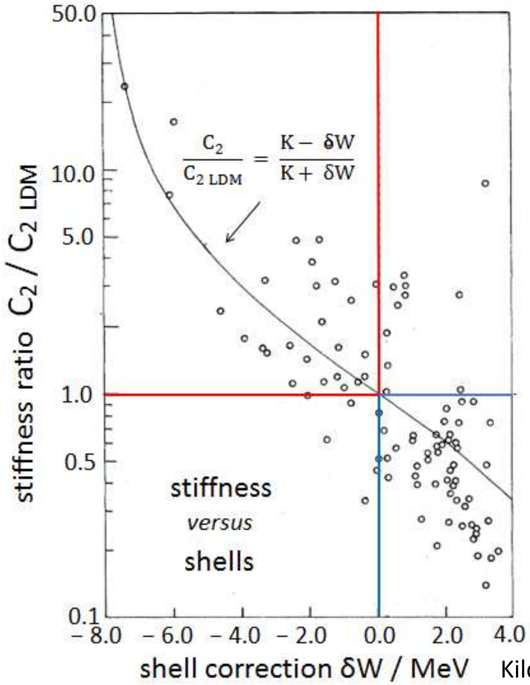


Jensen-Damgaard 1973

Stiffness of Nuclei

Shell effects not only affect mass correction δW but also stiffness (parameter α or $C_2 = 5\alpha R_0^2/2\pi$):

$\boxed{E_{def} = \alpha\,(D - R_o)^2}$ with D = major semi-axis of spheroid



Kildir-Aras 1982

Stiffness C_2 found in Coulomb excitation of collective vibrations in e-e spherical nuclei (Alder, Bohr et al).

correlation
stiffness ⟷ δW

Shell nuclei are stiff: $\alpha > \alpha_{LDM}$

Anti-Shell nuclei are soft: $\alpha < \alpha_{LDM}$

Parameterization in Fig. with K = 8 MeV

Scission Point Model (SPM)

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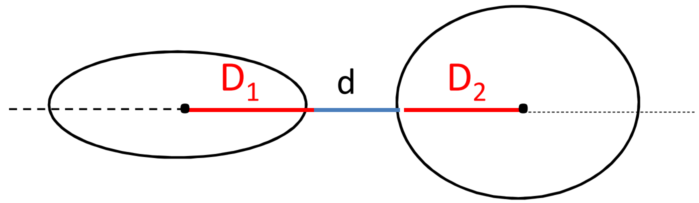
Scission Point is visualized by two aligned spheroids:



In SPMs the energy bound as potential energy V is

$$V = V_{Coul} + V_{Def} = \frac{Z_1 Z_2}{D_1+D_2+d} + \alpha_1(D_1 - R_{01})^2 + \alpha_2(D_2 - R_{02})^2$$

The disposable energy is the Q-value for the mass split:

$$Q = TKE + TXE = (V_{Coul} + E_{Kpre}) + (V_{Def} + E_{int}^*)$$

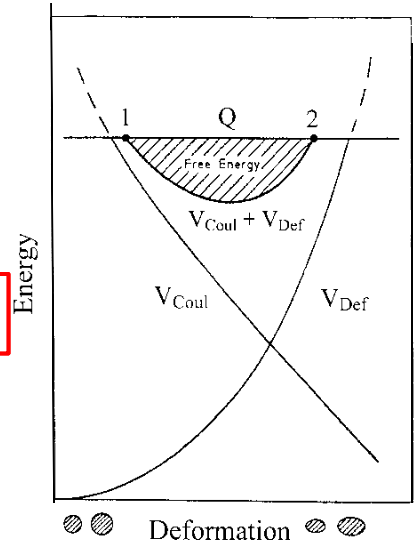
The energy available for $(E_{Kpre} + E_{int}^*)$ is **F = Q – V.**

Quasi-static configuration is attained for F at minimum :

$$\partial F/\partial D_1 = 0$$
$$\partial F/\partial D_2 = 0 .$$

Calculate

$$\boxed{E_{def1} / E_{def2} = \alpha_2 / \alpha_1}$$



In the combination

soft α_1 ⟷ stiff α_2

the soft FF gets the larger deformation energy

Note: $\frac{\alpha}{\alpha_{LDM}} = \frac{K - \delta W}{K + \delta W}$ with K = 8 MeV

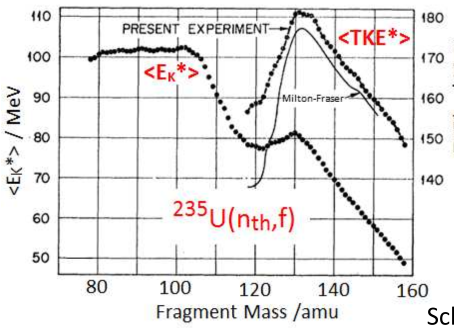
$\alpha_{LDM} = 2.896 - 0.0630\,(Z^2/A)$ MeV/fm² BW 1939

Total Kinetic Energy

Shells and anti-shells in the TKE of fragments for **^{235}U(n,f)**

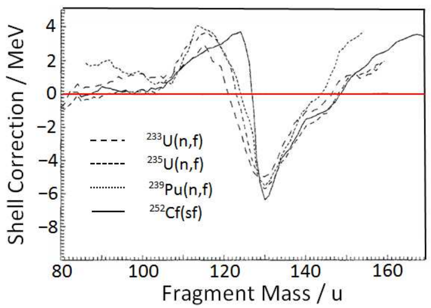
Total Kinetic Energy *vs* Fragment Mass

In low energy fission of all actinides the dip in total kinetic energy TKE near mass symmetry is spectacular:



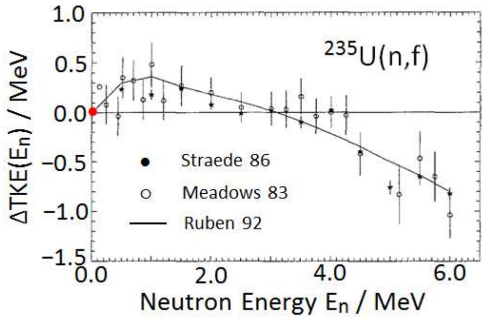
Schmitt 1966

It is understood in terms of shell and anti-shell effects : for near-symmetric fission two fragments A ≈ 120 with δW > 0 appear. They are particularly soft leading to elongated scission configurations with small V_{Coul} and hence small TKE. Neighboring asymmetric events with A_H ≈ 132 have δW < 0. Strong shell effects lead to compact scission configuration with large TKE.

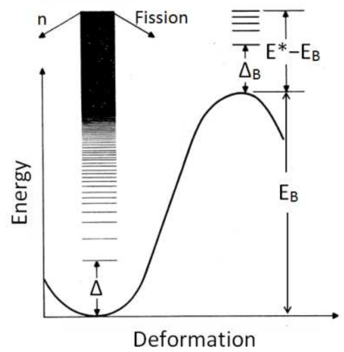


Ruben 1991

Total Kinetic Energy *vs* incoming neutron energy E_n



In ^{235}U(n,f) TKE ↘ for E_n ↗ is attributed to fading shell near A ≈ 132. Up to A ≈ 145 nuclei become softer and the scission configurations more elongated and hence TKE ↘ .

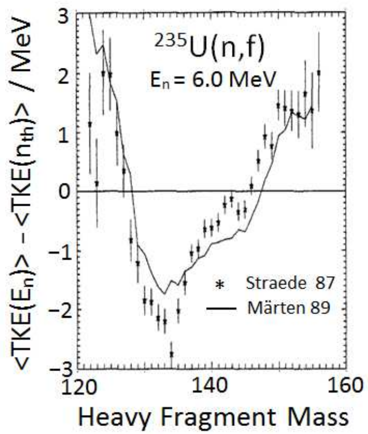


Increase of TKE for E_n ≤ 1 MeV?

^{236}U is fissile:

B_F = 5.62 MeV ; B_n = 6.8 MeV.

For thermal neutron fi the transition state is in the level gap and for E_n ↗ the energy goes in TKE = V_{Coul} + E_{kpre} to pre-scission kinetic energy: TKE ↗



Straede 1987

Surprise:

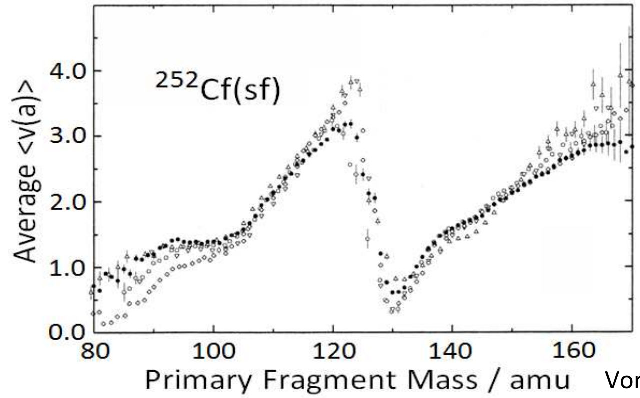
For heavy fragment A_H ≥ 150 u TKE increases for E_n = 6 MeV relative to E_n thermal.

Anti-shell effect fades at higher excitation, nuclei become stiffer leading to smaller D at scission and larger V_{Coul} → TKE

Neutron Multiplicity

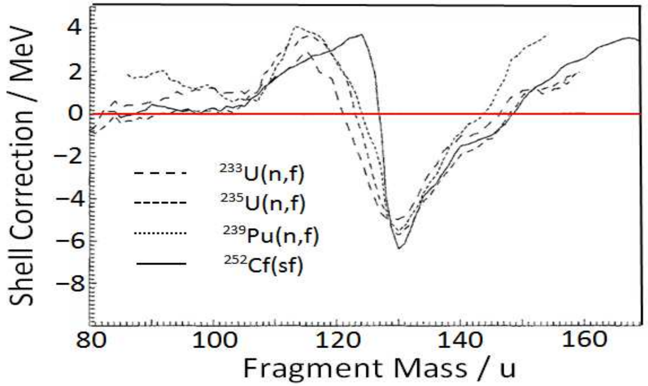
Shells and anti-shells in neutron evaporation from fragments

Neutron multiplicity



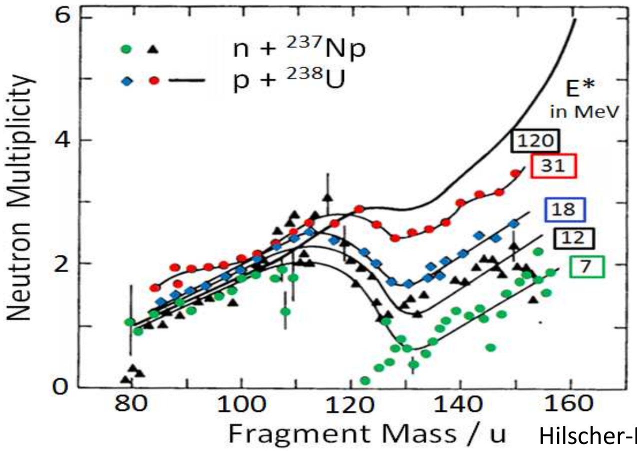
Vorobyev 2001

Shell correction δW



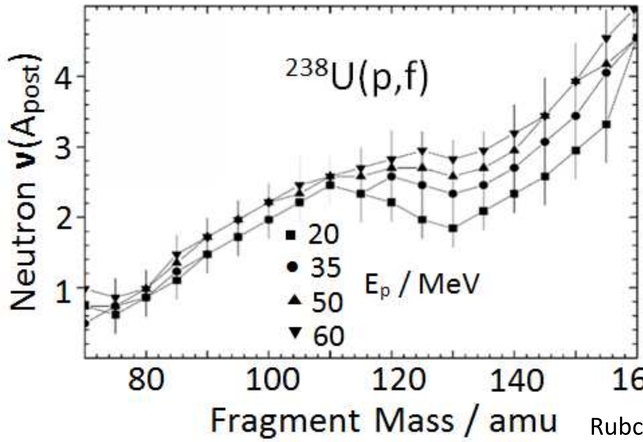
The sawtooth ν(A) of neutron multiplicity reflects the combination of stiff shell nuclei near A ≈ 132 and soft anti-shell nuclei near A ≈ 120. The Scission Point Model explains the relative deformation energies and hence n-multiplicities. Note that even finer structures in the shell correction are mirrored in the n-multiplicity.

With increasing excitation energy both, shell and anti-shell effects are fading. Shell nuclei become softer and anti-shell nuclei become stiffer. This is reflected as the smoothing of the neutron sawtooth ν(a).



Hilscher-Rossner 1992

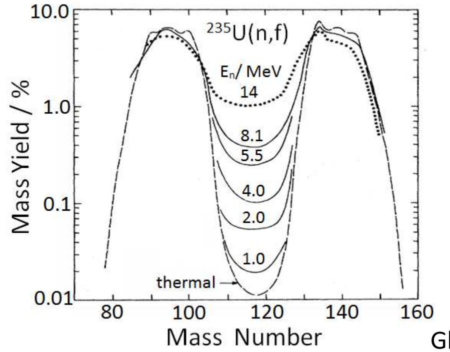
With excitation increasing the neutron multiplicity ν(A) approaches the expectation from LDM: ν(A) ~ A. In the LDM there are no shell nor anti-shell effects.



Rubchenya 2001

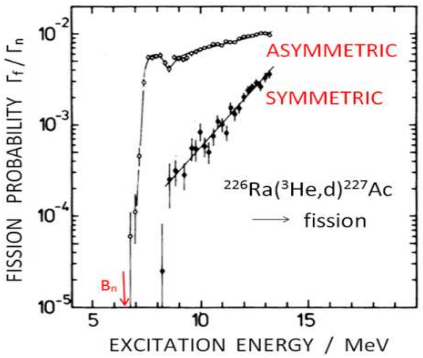
Symmetric – Asymmetric Fission in the Actinides

Turkevich-Niday Modes



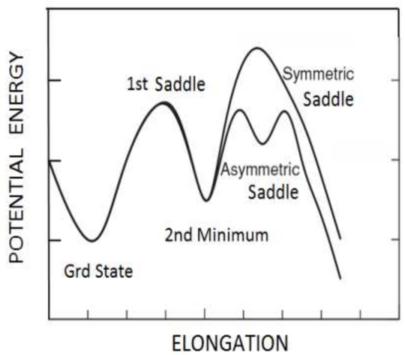
Glendenin 1981

The mass yield Y(A) in low energy fission of actinides is dominantly asymmetric. The position of the heavy group is fixed by shell effects for N = 82 and ≈ 88, respectively).



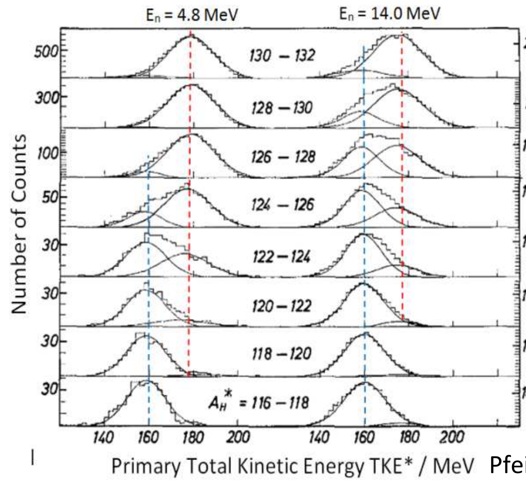
Konecny 1974

In symmetric fission anti-shell effects prevail. The two distinct modes, symmetric and asymmetric, have different thresholds. In actinides:
thr. Symm. > thr. Asymm.



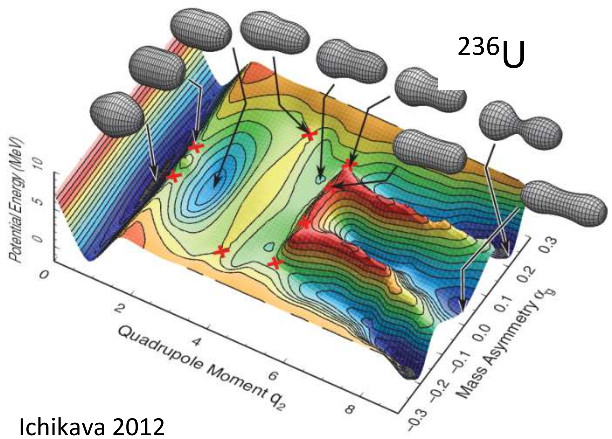
The double-humped PES has near saddle two outer barriers of different height steering the symmetric and asymmetric distributions Y(A) of mass. For increasing excitation energy the symmetric mode catches up with the asymmetric mode.

As postulated by Turkevich–Niday symm. and asymm. fission evolve independently



Pfeiffer 1971

In theory of the PES the two valleys of modes are separated by a high ridge preventing cross talk between modes.



Ichikava 2012

BROSA modes in the Actinides

Bimodal Asymmetric Fission

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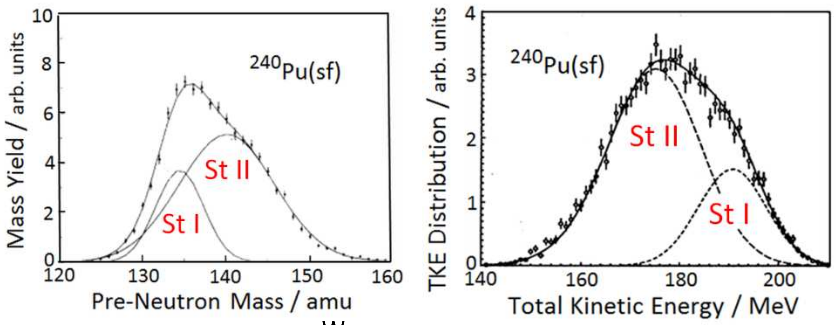
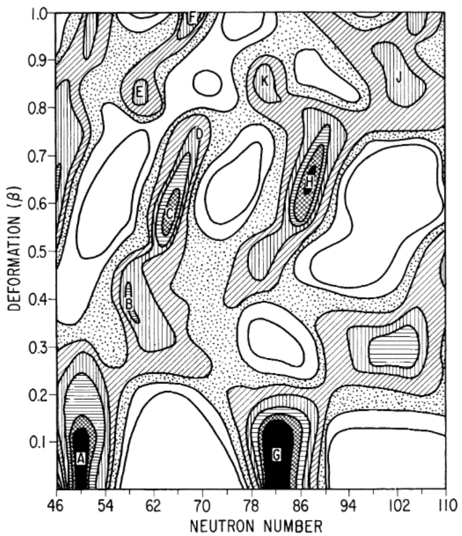
Structure in fragment mass and energy distributions of asymmetric fission are described by Brosa as the superposition of Standard I and Standard II modes.



Wagemans 1989

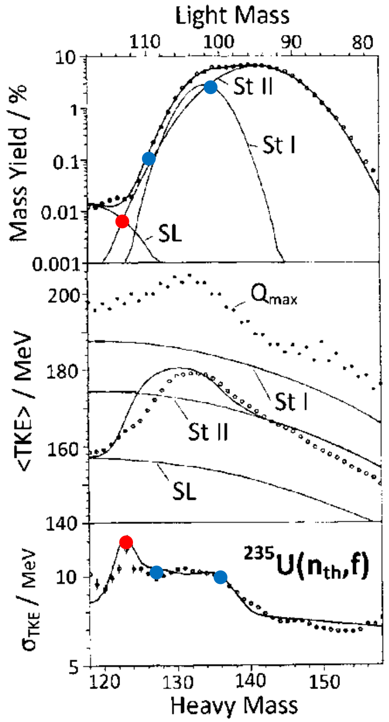
They are ascribed to shell effects in heavy fragments:



Standard I :
Spherical Shells
Z = 50 , N = 82

Standard II :
Deformed Shells
N = 88

Wilkins 1976



^{235}U(n$_{th}$,f)

Superlong SL:
anti-shells

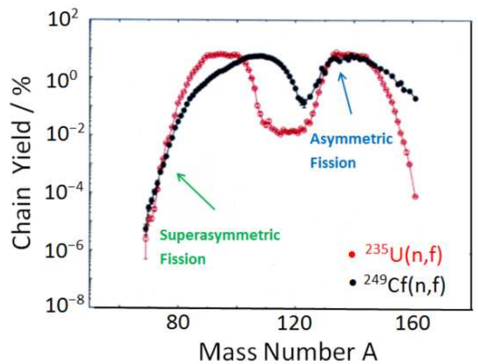
Standard I :
spherical shells

Standard II :
deformed shells

<A_{HF}>	118	134	141
<TKE>	157	187	167

σ_{TKE} large in overlap

Knitter 1987



Shell effects in
the light fragment
lead to

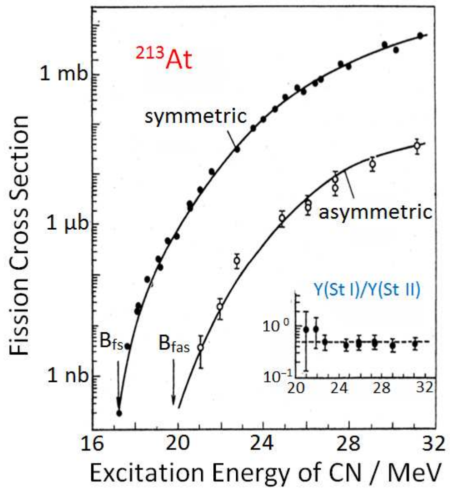
Super-asymmetric
Fission
(Standard III)

Gönnenwein 1999

ITKIS modes in the Pre-Actinides

Bimodal Asymmetric Fission

Symmetric-asymmetric fission in pre-actinides

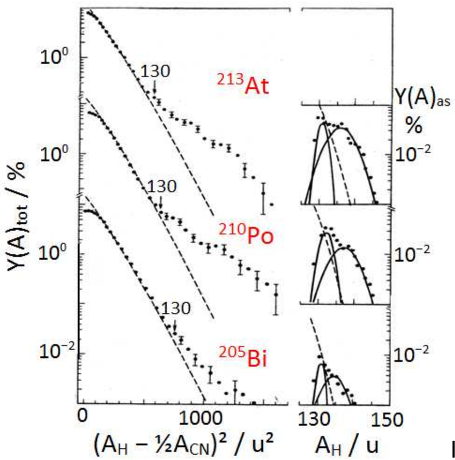


Itkis 1988

In contrast to actinides:
From ^{201}Tl to ^{213}At symmetric fission is dominant in the pre-actinides

Thresholds:
$B_f^{symm} < B_f^{asymm}$

ITKIS modes in bimodal asymmetric fission

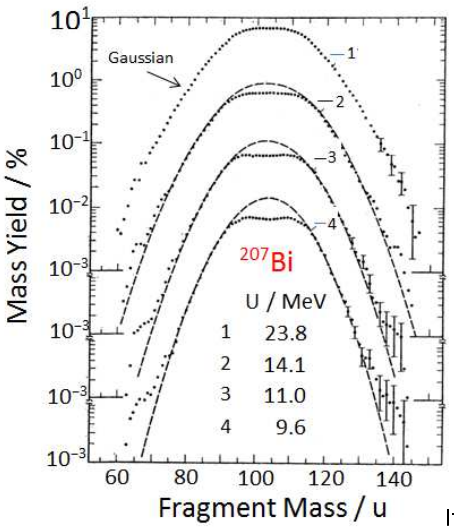


Itkis 1988

In the wings of Y(A)
Bimodal Asymmetric Fission

Standard I: $<A_H> \approx 132$
Standard II: $<A_H> \approx 139$

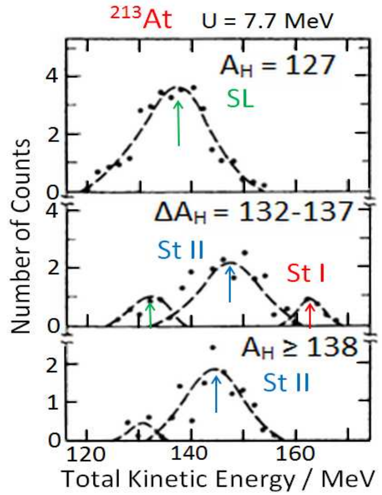
In figure excitation energies at saddle are $E^* = 9.0 \pm 0.5$ MeV



Itkis 1985

Mass Distributions of fi fragments in the pre-actinides are Gaussians
$Y(A) \sim \exp[-(A-A_{CN}/2)^2/2\sigma_A^2$

Deviations:
1) asymmetry in the wings
2) dent right at symmetry : anti-shell effect for 2 FF with $N \approx 62$



Itkis 1985

Like in actinides also in Total Kinetic Energy 3 modes are observed : one symmetric SL and two asymmetric modes.

Like in actinides :

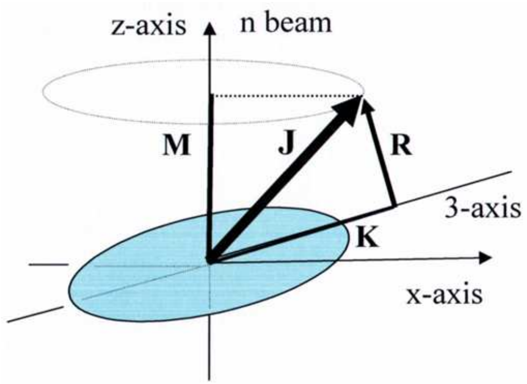
TKE(SL) < TKE(St II) < TKE(St I)

Itkis modes ≡ Brosa modes

Itkis: no asymmetric fission for compound nuclei with $A_{CN} \leq 200$ u
However: asymmetric fi of ^{180}Hg newly discovered Andreyev 2010

Angular Distributions of Fission Fragments (FF)

in (n,f) reactions with (e,e) targets near barrier



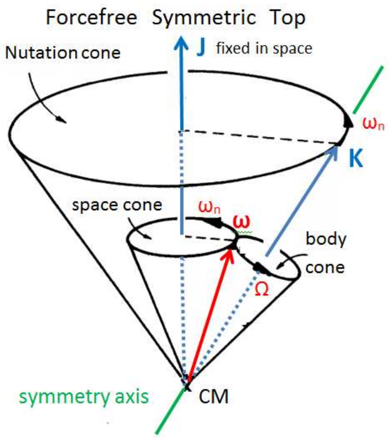
- Fission prone nucleus near saddle = spheroid
- good quantum numbers are J, M and K
- FF are ejected along axis of elongation: fission axis
- Angular distribution of FF ≡ orientation of fission axis

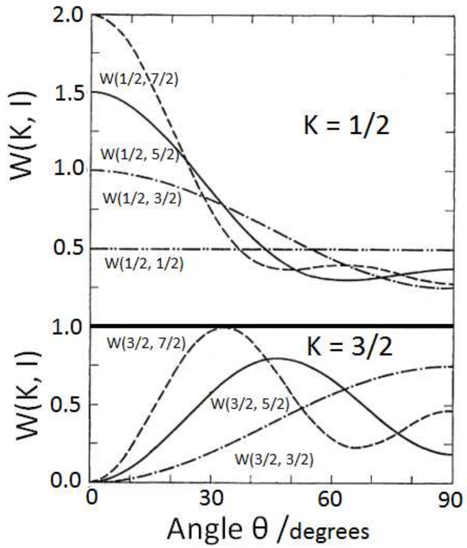
$$W^J_{MK}(\theta) = \tfrac{1}{4}\,(2J + 1)\,\{|\,d^J_{+\frac{1}{2}K}|^2 + |d^J_{-\frac{1}{2}K}|^2\}$$

with $\theta = \sphericalangle(n,FF)$ and d^J_{MK} = wavefctn of symmetric top

Symmetric top in classical mechanics:

nutation





K quantum numbers characterize W(θ).

For K = 1/2 the FF are ejected along fi axis

For K = 3/2 the FF are ejected sideways

Angular Distributions in Symmetric- Asymmetric Fission

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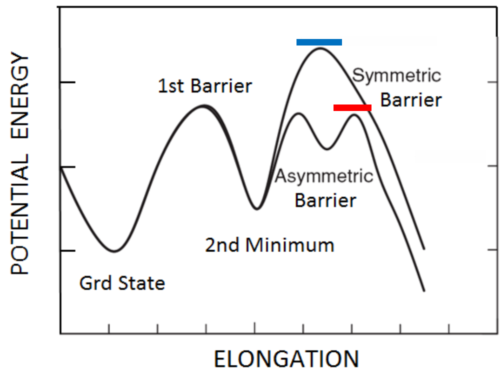
Turkevich-Niday modes 1951

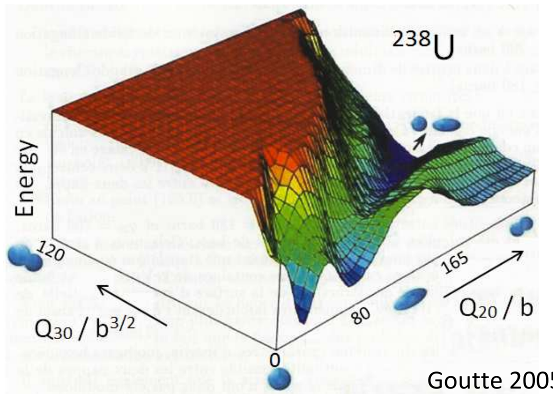
Symmetric ↔ asymmetric fission

Both, in pre-actinides and actinides the barriers differ for symmetric and asymmetric fission:

$\boxed{\text{symmetric } B_{fi} \neq \text{ asymmetric } B_{fi}}$

Actinides:





Goutte 2005

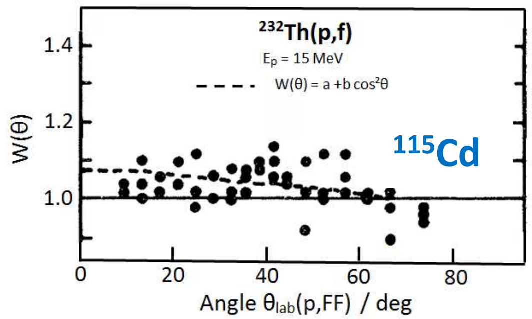
The experimentally found differences in barrier heights are well described by macroscopic and microscopic theories

Theory by A. Bohr of FF angular distributions

W(θ) of FF is steered by the quantum numbers (J,K) of transition states on top of barriers .

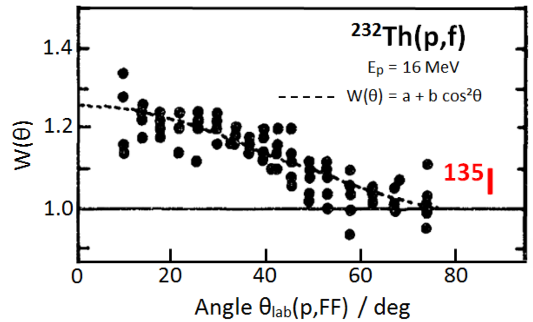
Since barriers B_{fi} differ for symm. ↔ asymm. fission, also transition states and quantum numbers (J,K) are different.

Hence W(θ) depends on FF mass A : $\boxed{W(\theta,A) = f(A)}$



Symmetric fission

Kudo 1982

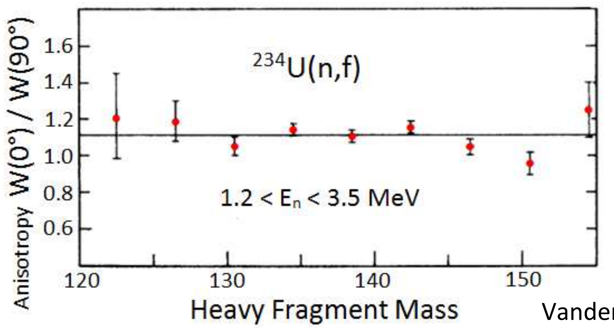


Asymmetric fission

However: the asymmetric St I and St II modes share the same W(θ).

In full mass range of asymmetric fission : $\boxed{W(\theta,A) \neq f(A)}$

Angular distributions for asymmetric St I and St II are identical as observed in experiment.



Vandenbosch 1965

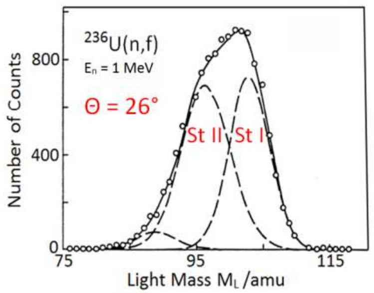
Within the full mass range of asymmetric fission the anisotropy W(0°)/W(90°) analyzed in terms of

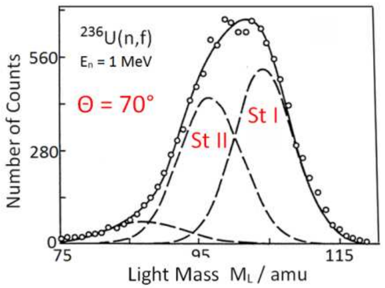
$$W(\theta) = A + B\cos^2\theta$$

is constant

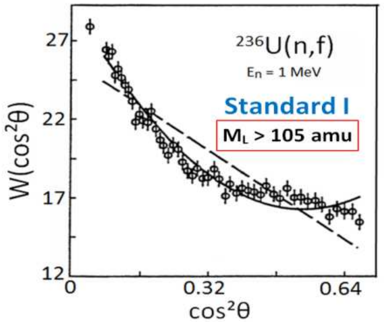
Bymodal Asymmetric Fission in Sub-barrier Resonances

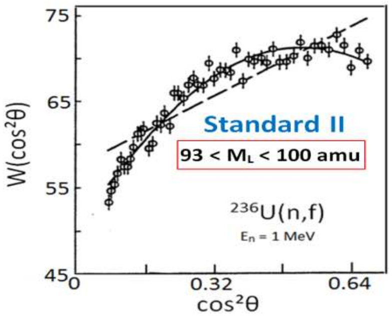
Brosa mode analysis of ^{236}(Un,f) at E_n = 1 MeV near **sub-barrier resonance** at E_n = 0.93 MeV. Goverdovski 93/94



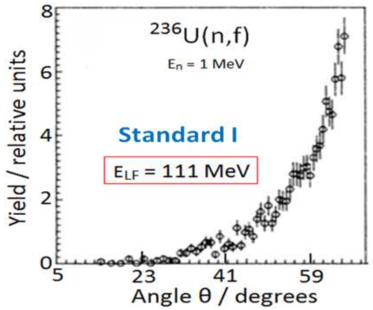


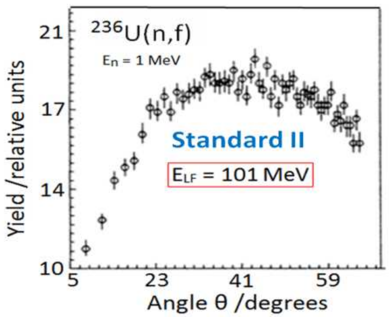
Mass distribution $Y(M_L)$ depends on emission angle θ





Angular distribution depends on LF mass M_L

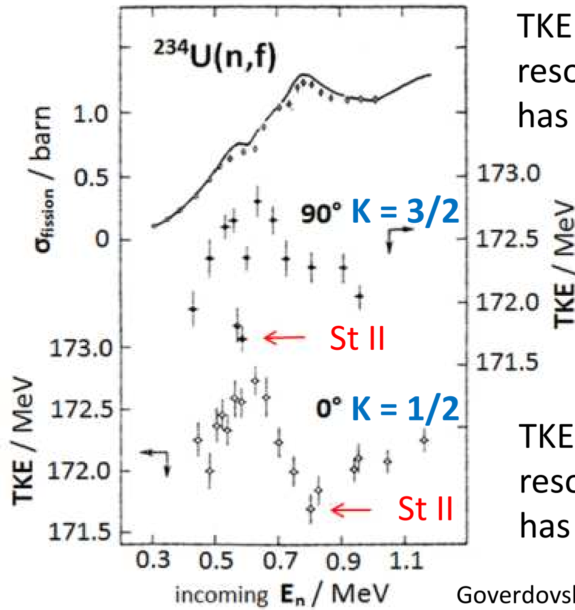




Angular distribution depends on LF energy E_{LF}

Angular distributions are different for Brosa St I and St II
⇒ St I and St II have different (J,K)

Sub-barrier fission in (n,f) of ^{234}U:
σ_{fi} exhibits pronounced resonances at E_n = 0.55 and E_n = 0.78 MeV



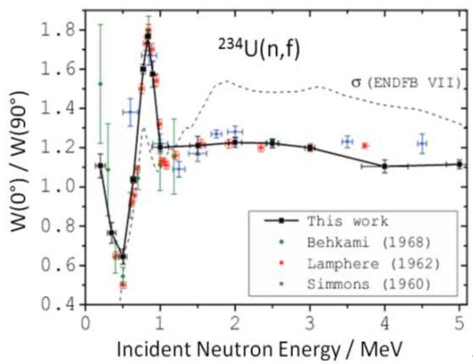
Goverdovski 1987

TKE data for θ = 90° : resonance 0.55 MeV has St II with K = 3/2

Dip in TKE is attributed to surge of St II

TKE data for θ = 0° : resonance 0.78 MeV has StII with K = 1/2

Since (J,K) differ for St I and St II
⟹ K quantum numbers for St I are complementary



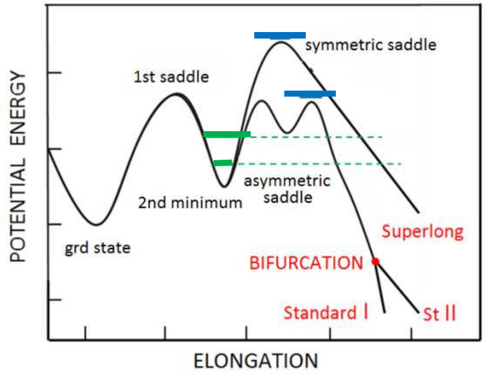
Al-Adili 2016

Anisotropy W(0°) / W(90°) <u>is smooth</u> above resonances <u>fluctuating</u> in resonances

Bimodal asymmetric fission

Brosa-Itkis modes

Where in the PES appear Brosa modes ?



Transition states

Transmission resonances

Why is only in sub-barrier fi the ang. distr. W(θ) dependent on mass and TKE of FF?

Model A: **Barriers**

StI and StII have DIFFERENT BARRIERS at saddle. W(θ, A,TKE) follows like for Turkevich-Niday modes.

BUT: 1) Saddle is under-tunnelled and not passed.

2) Different barriers should be seen in above-barrier fi which is not the case

Model B: **Bifurcation**

Fission emerges from a transmission resonance into the PES below the barrier. Resonance ≡ β-vibration contributes to total barrier penetrability but is not a transition state subject to theory of A. Bohr :

St I and St II may be populated with different (J,K) !

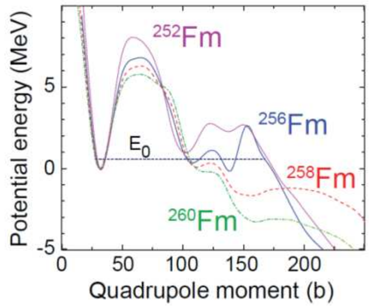
Modes ≡ BIFURCATION in downhill PES to scission

Example $^{234}U(n,f)$: resonances partially favor St II .

Bimodal symmetric fission

Hulet modes

Fm has Z = 100 = 2 x 50. For heavy isotopes with N ≥ 164 = 2 x 82, the second asymmetric barrier dives under the ground state. Therefore

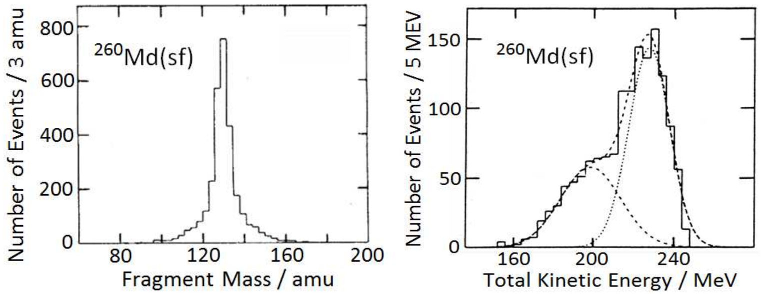


Staszcak 2009

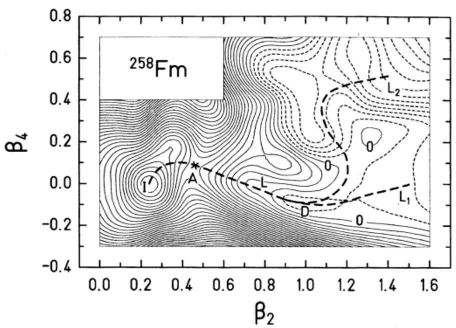
Effective barrier is symmetric →

Symmetric Fission

Both in Mass and TKE distributions discovery of fine structure : Hulet modes



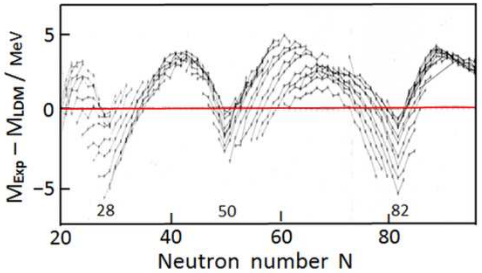
Hulet 1986

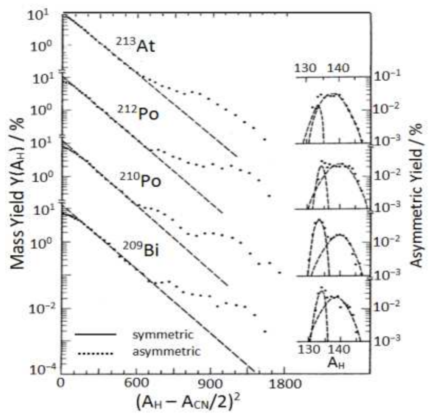


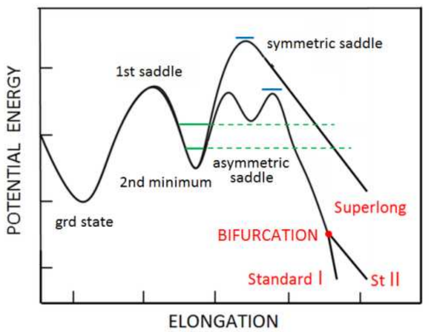
Cwiok 1989

Hulet modes show up in the PES once the symmetric saddle has been passed as a bifurcation

Summary







- Shell correction $\boxed{\delta W = M_{exp} - M_{LDM}}$ Myers-Swiatcki 1966

  Shell effect $\delta W < 0$ nuclei stiffer than in LDM $\longleftrightarrow$ Anti-shell effect $\delta W > 0$ nuclei softer than in LDM
- Two-Mode Hypothesis of Turkevich-Niday 1951

  Symmetric $\longleftrightarrow$ Asymmetric Fission

  Fission barriers differ in height for symmetric and asymmetric mode

  Pre-actinides $B_f^{symm} < B_f^{asymm}$

  Actinides: $B_f^{asymm} < B_f s^{ymm}$
- Symmetric fission: anti- shell effects $\longleftrightarrow$ Asymmetric fission: shell effects
- Fine structure in asymmetric fission:

  Bimodal asymmetric modes: Itkis in pre-actinides $\longleftrightarrow$ Brosa in actinides

  Standard I mode: shell effect for spherical nuclei with Z = 50 and N = 82

  Standard II mode: shell effect for deformed nuclei with N = 88
- For symmetric $\longleftrightarrow$ asymmetric fission angular distributions differ because the transition states (J,K) at the two barriers controlling W(θ) are different (A. Bohr theory)
- For bimodal asymmetric fission in nuclei excited above the barrier, the W(θ) is identical for both modes. Both modes hence share the same (J,K) imposed by one common transition state. Modes develop once barrier has been passed.
- In sub-barrier fission a pronounced mode dependence of W(θ) is observed near resonances of cross section σ_{fi}. The modes St I and St II have hence different K-values.
- Transition resonances through double-humped barrier are traced to β-vibrations in 2nd minimum of barrier. Fission emerges into PES below the barrier. There is no transition state. Resonance populates St I or St II with different weights.

  Modes ≡ BIFURCATION in downhill PES to scission
- In double-humped barrier 1st saddle is tri-axial and 2nd saddle asymmetric. In heavy Fm isotopes 2nd saddle < ground state $\longrightarrow$ symmetric fission.
- Bimodal symmetric fission with modes according to theory bifurcating once barrier is passed