Lecture 10: Fission
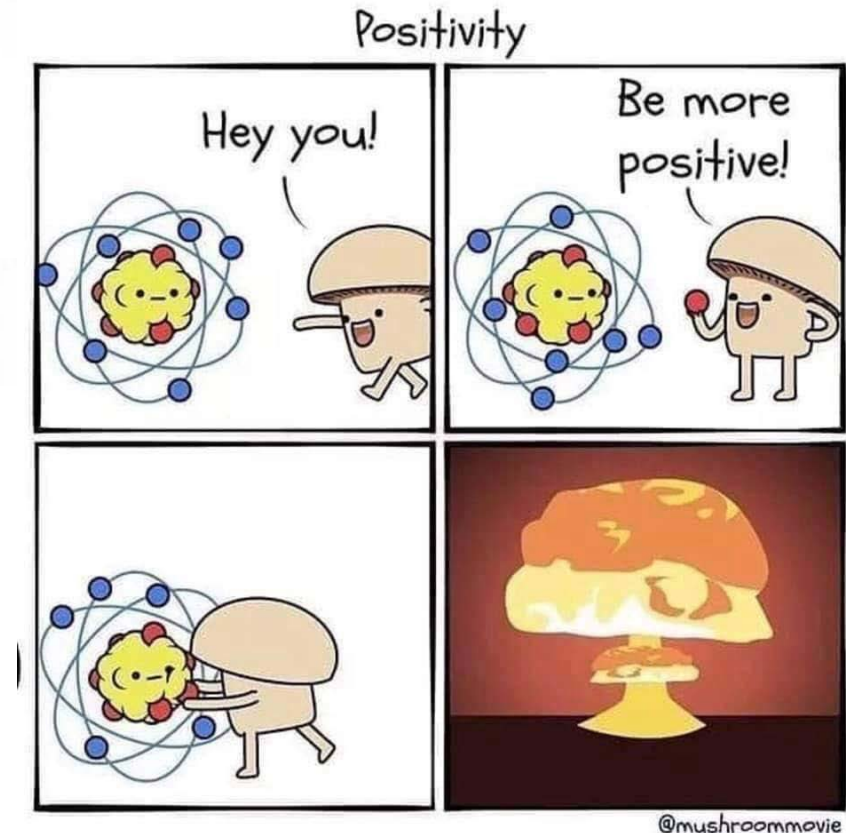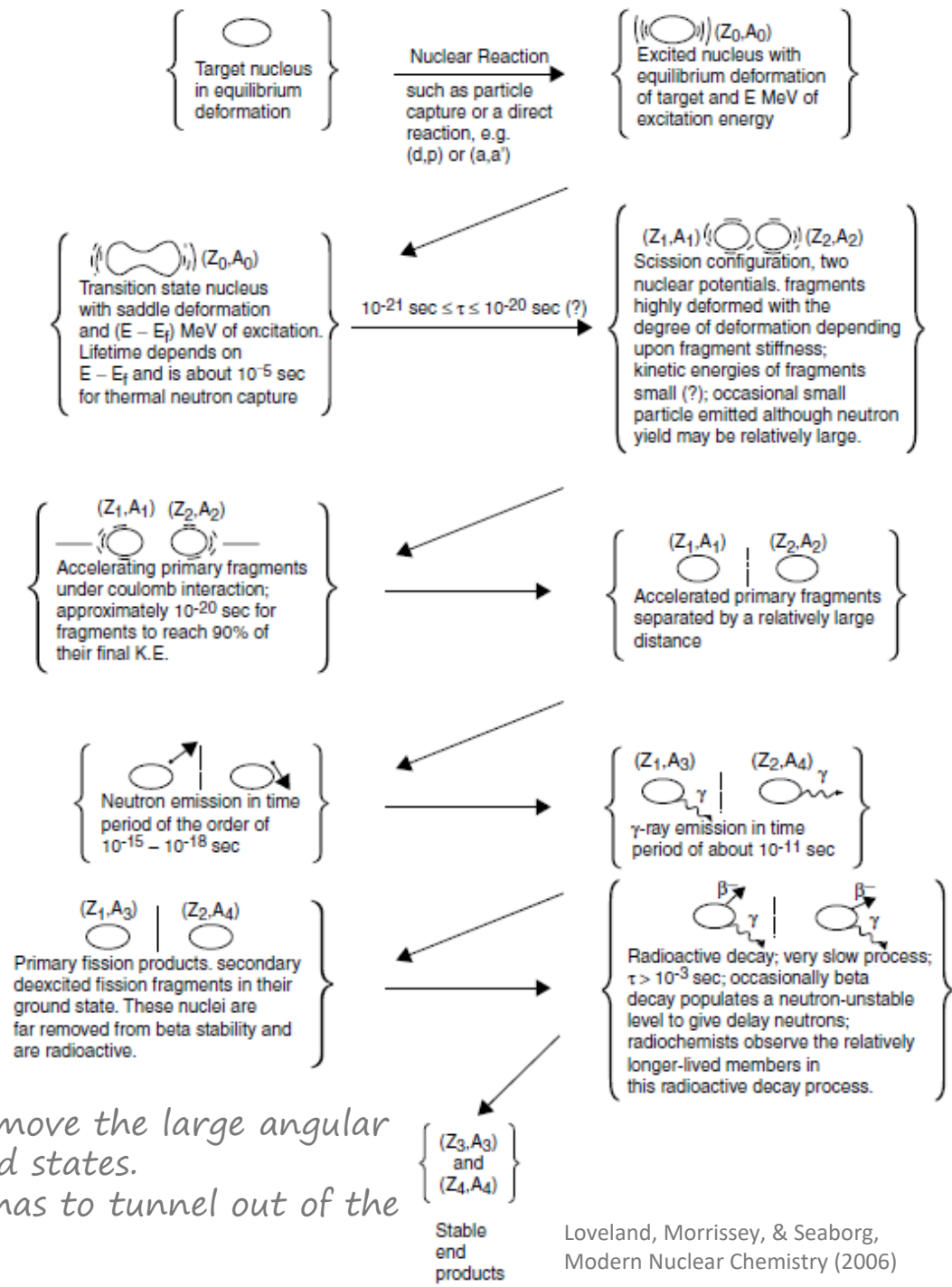
- Conceptual process
- Fissionability
- Decay rate
- Decay branching
- Mass distribution
- Kinetic energy
- Neutrons

Steps of fission

1. A nucleus becomes deformed either due to an external perturbation that brings in energy or an internal cluster rattling around within the potential well

2. The energy is absorbed as a collective excitation that manifests itself as a drastic shape change, elongating the nucleus into a peanut shape

3. The separation of the two lobes of the peanut becomes great enough that the two repel each other, splitting apart at the scission point

4. The coulomb repulsion accelerates the two fragments apart

5. The two fragments are each highly excited and de-excite initially via neutron emission, followed by γ emission
   (meaning prompt neutrons will be emitted along the direction of the fragments)

6. The neutron-rich fragments will then β decay back to stability, possibly emitting delayed neutrons via β-delayed neutron emission

Target nucleus in equilibrium deformation

Nuclear Reaction such as particle capture or a direct reaction, e.g. (d,p) or (a,a')

(Z_0,A_0) Excited nucleus with equilibrium deformation of target and E MeV of excitation energy

(Z_0,A_0) Transition state nucleus with saddle deformation and $(E - E_f)$ MeV of excitation. Lifetime depends on $E - E_f$ and is about 10^{-5} sec for thermal neutron capture

10^{-21} sec $\leq \tau \leq 10^{-20}$ sec (?)

(Z_1,A_1) (Z_2,A_2) Scission configuration, two nuclear potentials. fragments highly deformed with the degree of deformation depending upon fragment stiffness; kinetic energies of fragments small (?); occasional small particle emitted although neutron yield may be relatively large.

(Z_1,A_1) (Z_2,A_2) Accelerating primary fragments under coulomb interaction; approximately 10^{-20} sec for fragments to reach 90% of their final K.E.

(Z_1,A_1) (Z_2,A_2) Accelerated primary fragments separated by a relatively large distance

Neutron emission in time period of the order of $10^{-15} - 10^{-18}$ sec

(Z_1,A_3) (Z_2,A_4) γ-ray emission in time period of about 10^{-11} sec

(Z_1,A_3) (Z_2,A_4) Primary fission products. secondary deexcited fission fragments in their ground state. These nuclei are far removed from beta stability and are radioactive.

Radioactive decay; very slow process; $\tau > 10^{-3}$ sec; occasionally beta decay populates a neutron-unstable level to give delay neutrons; radiochemists observe the relatively longer-lived members in this radioactive decay process.

(Z_3,A_3) and (Z_4,A_4)

Stable end products

Why do you figure there is neutron emission at first and finally only γ emission?

A massive particle is better suited to remove the large angular momentum present in high-lying excited states.
At lower excitation energies, a particle has to tunnel out of the nucleus, while the γ doesn't.

Loveland, Morrissey, & Seaborg, Modern Nuclear Chemistry (2006)

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Energetics of shape change: *Liquid drop picture*

- Recall the contributions to the binding energy from the semi-empirical mass formula

- What happens if we deform the nucleus?
  - Volume will be conserved, A-Z stays the same, paired nucleons stays the same...
  - But, the surface will be enlarged and the charges within the nucleus will spread apart
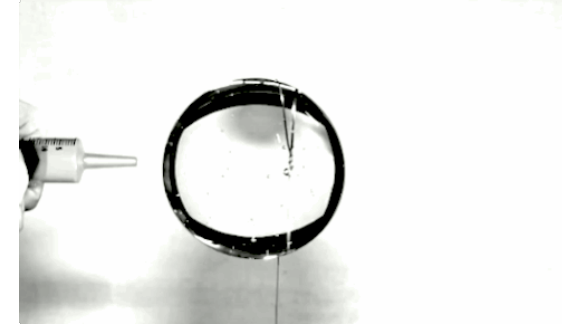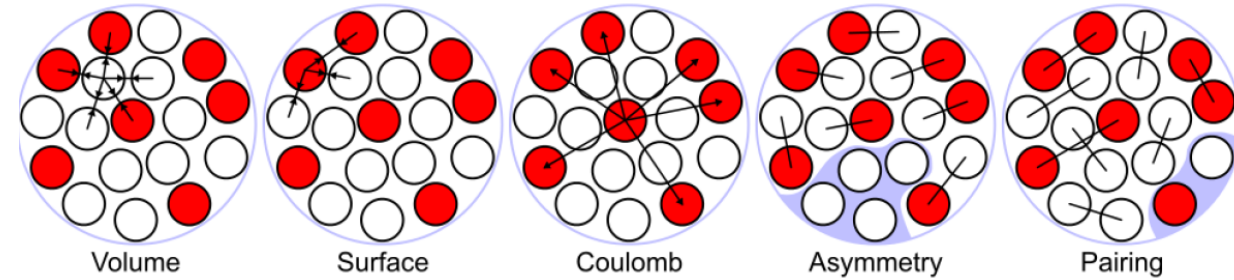
- In this picture, deformation will increase the binding energy penalty from the surface term, but decrease the binding energy penalty from the coulomb term

- So to understand what happens to the overall binding energy when the nucleus is deformed, we need to understand how these two terms evolve under deformation

- The change in binding energy will be $\Delta E = BE_{final} - BE_{initial} = (E_c' + E_s') - (E_c + E_s)$, where the primed terms include the modification to coulomb and surface terms under deformation, i.e. transforming from a sphere to an ellipsoid

Energetics of shape change: *Liquid drop picture*

- For the deformed shape, which is an ellipsoid in this picture, the nuclear radius can be parameterized as an expansion in terms of Legendre polynomials
  [which for axial symmetry will only keep the $l = 2$ term]

B.R. Martin, Nuclear and Particle Physics (2009)

  - $R(\theta) = R_0[1 + \alpha_2 P_2(\cos\theta)]$

  - α_2 is the quadrupole distortion parameter, which is related to the quadrupole deformation by

    $\alpha_2 = \sqrt{\frac{5}{4}\pi\beta_2}$, and the ellipsoid axes by: $a = R_0(1 + \alpha_2)$, $b = R_0\frac{1}{\sqrt{1+\alpha_2}}$

- It turns out, expanding the Coulomb and surface energy terms as a power series in α_2 yields

  - $E_C' \approx a_c \frac{Z^2}{A^{1/3}}\left(1 - \frac{1}{5}\alpha_2^2\right)$ and $E_s' \approx a_s A^{2/3}\left(1 + \frac{2}{5}\alpha_2^2\right)$

- Meaning the energy cost for deformation is $\Delta E = \frac{\alpha_2^2}{5}\left(2 a_s A^{2/3} - a_c \frac{Z^2}{A^{1/3}}\right)$ *which we determined back in lecture 4*

- So, when the non-deformed Coulomb energy is twice the non-deformed surface energy or greater, there is zero energetic cost (or even an energetic gain) to deform (and ultimately fission!)

- The fissionability parameter $x = \frac{E_C}{2E_s} = \frac{a_c}{2a_s}\frac{Z^2}{A} \equiv \frac{Z^2/A}{\left(Z^2/A\right)_{critical}}$ is a measure of fission favorability

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Energetics of shape change: *Liquid drop picture*

- What is the Z,A combination prone to fission? i.e. what is $\left(Z^2/A\right)_{critical}$?
  - For the liquid drop, we just defined it as $\frac{2a_s}{a_c}$ ...which, from our fit to masses is ≈ 48

    [that's Z>116,A≥270]

  - Taking Loveland, Morrissey, & Seaborg's word for it "a more sophisticated treatment" yields $\left(Z^2/A\right)_{critical} = 50.8333\left[1 - 1.7826\left(\frac{N-Z}{A}\right)^2\right]$

- Note larger fissionability (i.e. closer to $\left(Z^2/A\right)_{critical}$) means a nucleus is more prone to fission
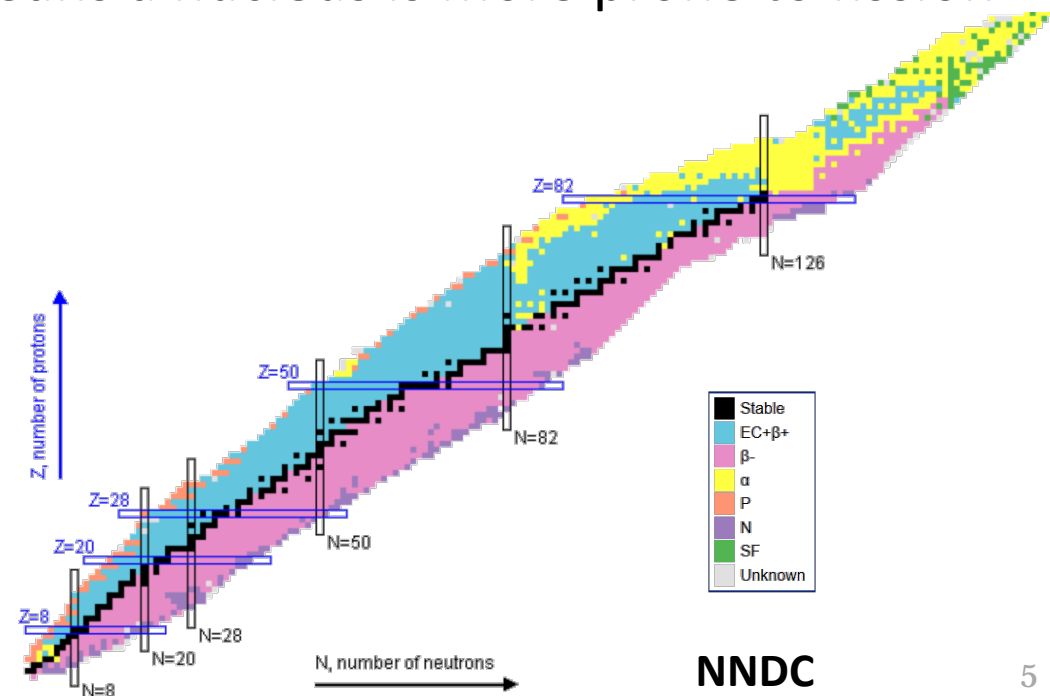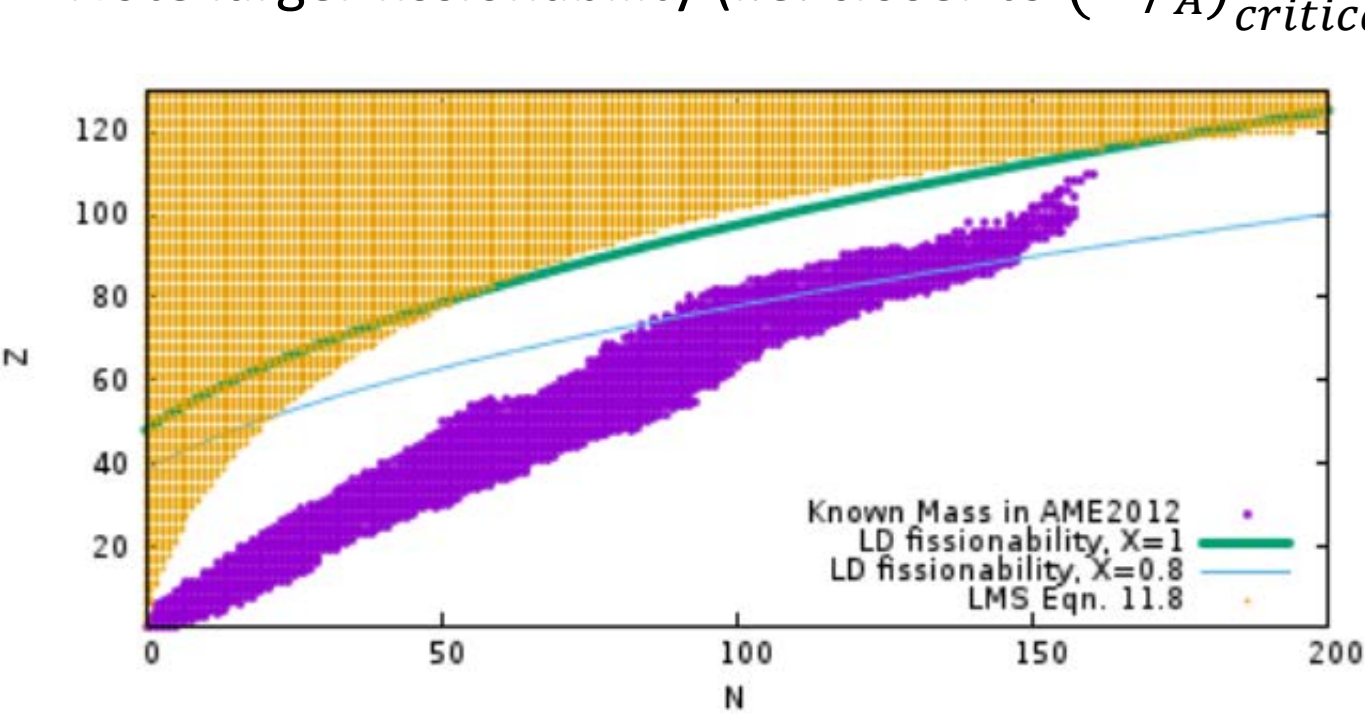
Fission barrier

- To quantify the probability of fission occurring, we can consider the potential energy associated with a particular shape of the nucleus

- This potential energy is dominated by liquid drop energetics,

  ...but with a major correction for enhanced binding near closed shells

  ...the magnitude of which is altered by the nuclear deformation

- So, the peaks and valleys in potential energy space are associated by the competition between liquid-drop and shell-model modified by deformation

- The likelihood of transitioning from one shape to another depends on the energetic cost associated with overcoming *(or tunneling through)* the barrier between them

Hill & Wheeler, Phys. Rev. 1953
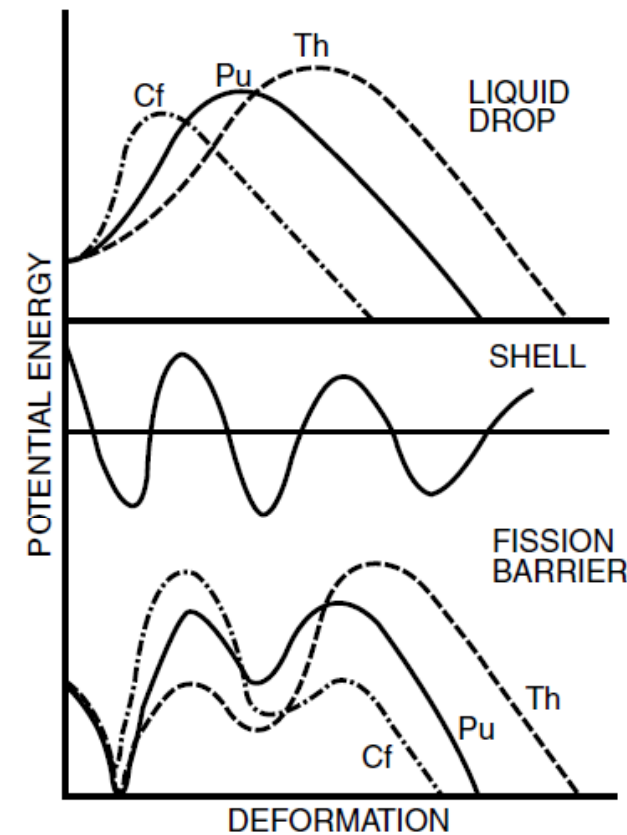
Loveland, Morrissey, & Seaborg, Modern Nuclear Chemistry (2006)
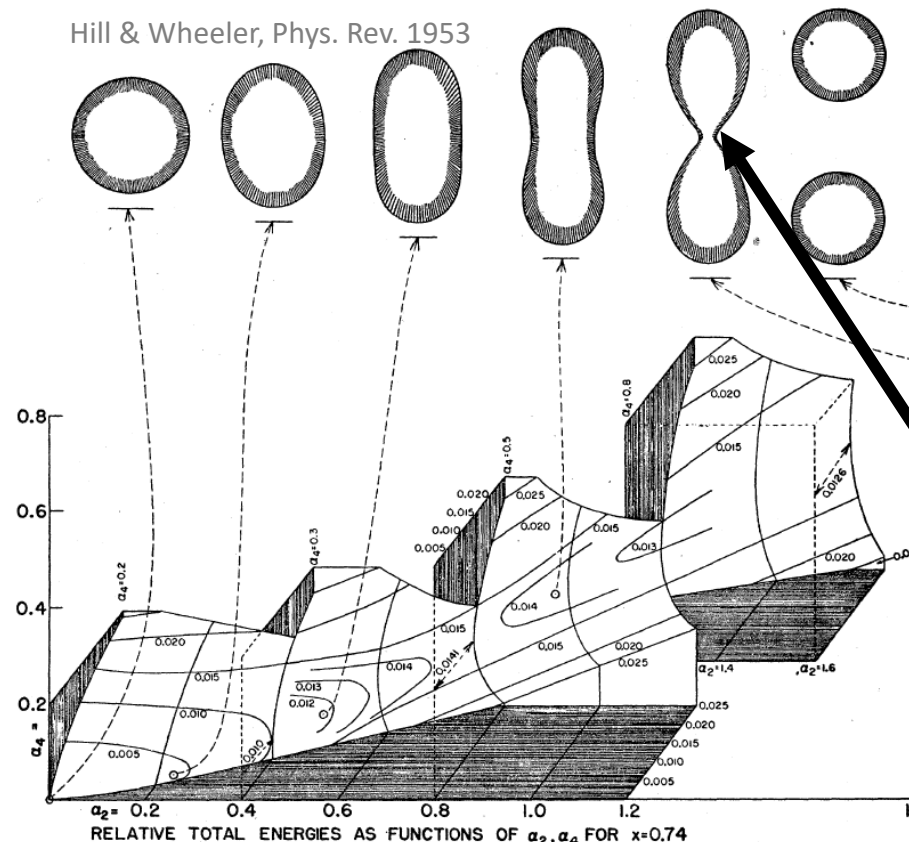
scission occurs at the saddle point in the potential

RELATIVE TOTAL ENERGIES AS FUNCTIONS OF α_2, α_4 FOR x=0.74

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Fission barrier height / Activation energy

- Quantitatively, the energy that must be paid *(or at least approached [tunneling])* is an activation energy

- The more common parlance is to refer to the fission barrier height (E_f in the picture), which is measured relative to the ground-state of the original system, as quantified as a harmonic oscillator $\frac{1}{2}\hbar\omega$

- Rather than overcome the barrier, it is also possible to tunnel through it, in which case the relevant quantity is the barrier width

- This width will be related to the barrier curvature $\hbar\omega$, where a large curvature means a thin barrier (easier to tunnel through) and a small curvature means a thick barrier (harder to tunnel through)

- Overcoming the barrier (or overcoming part of it and tunneling through the top) corresponds to *induced* fission, while tunneling through the barrier corresponds to *spontaneous* fission
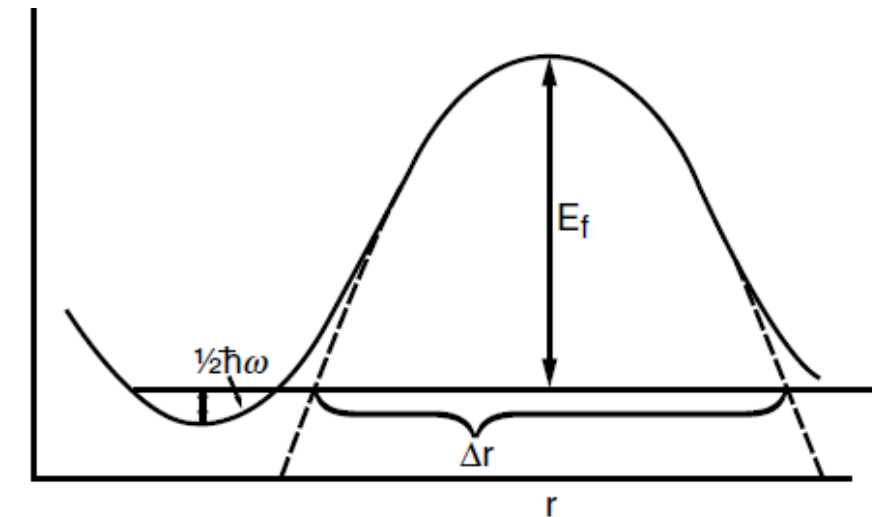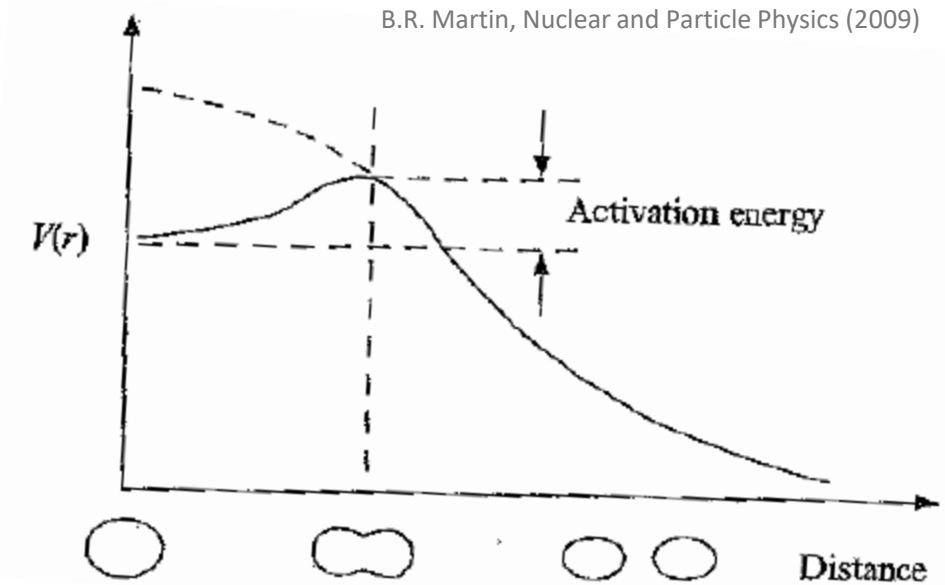
Fission barrier height

- In a liquid drop estimate, the fission barrier height is the difference between the ground-state binding energy of a system and the energy of two spherical nuclei made by splitting that system

- Near the longest-lived isotopes of Uranium and Thorium, $E_f \sim 5 - 7 MeV$

- Note this simple method yields one useful prediction already:
  - ^{235}U has a much shorter fission barrier height than ^{238}U and so the $A = 235$ isotope is more useful when a lot of fission is what you're looking for

N. Bohr & J. Wheeler, Phys. Rev. 1939

$f(x) = E_f / 4\pi R^2 O$

$x = \left(\frac{Z^2}{A}\right) / \left(\frac{Z^2}{A}\right)_{limiting}$

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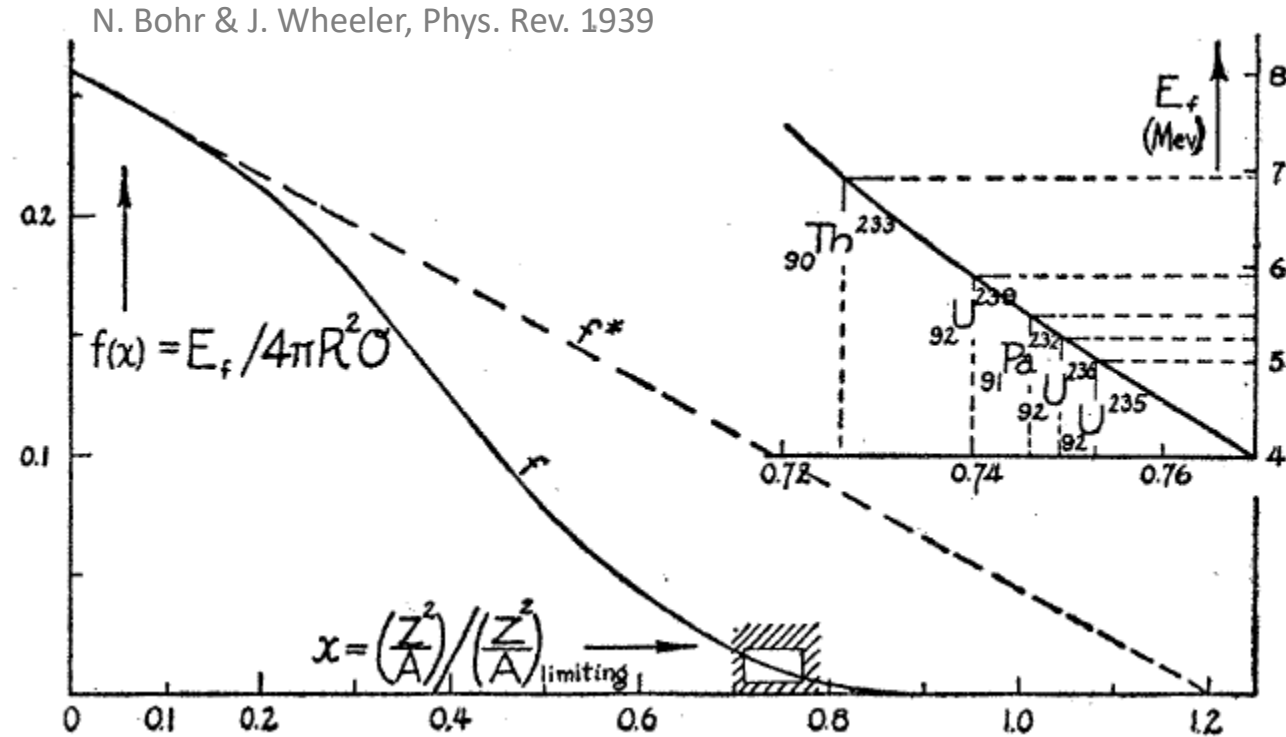
Calculating the fission barrier height ...is really hard

- The rough estimate is $B_f = E_{saddle} - E_{gs}$, where E_{gs} is the ground-state mass of the fissioning nucleus and E_{saddle} is the energy of the system when it is at the saddle-point in the potential energy landscape (i.e. the scission point)

- ...so we would need to calculate energy of the system versus deformation for lots of cases, which is a pain.

- Luckily Myers & Swiatecki (Phys.Rev.C 1999) performed fits for nuclei with $30.0 < x < 48.5$:

  - $B_f(N,Z) = S(N,Z)F(x)\ [MeV]$,

    where $S(N,Z) = (N+Z)^{2/3}\left(1 - k\frac{(N-Z)^2}{A^2}\right)$, with $k = 1.9 + (Z-80)/75$

      is clearly inspired by the surface energy with some regard for asymmetry

    and $F(x) = \begin{cases} 0.000199749(x_0 - x)^3 & if\ x_1 \le x \le x_0 \\ 0.595553 - 0.124136(x - x_1) & if\ 30 \le x \le x_1 \end{cases}$ with $x_1 = 34.15, x_0 = 48.5428$

      is clearly from an act of desperation

- E.g. $B_f(^{234}U)$=4.4 MeV, $B_f(^{236}U)$=4.5 MeV, $B_f(^{233}Th)$=6.2 MeV

- From FRDM (Moller et al. PRL 2004, PRC 2009): 4.9, 5.0, 5.5
  data quoted in FRDM papers: 5.5, 5.7, 6.2

Shell corrections aren't included here and can be ~1MeV order corrections

Fission barrier heights are difficult to pin-down

Theoretical estimates are sensitive to the adopted nuclear mass model

Experimental results differ because the height is not something that can be directly measured
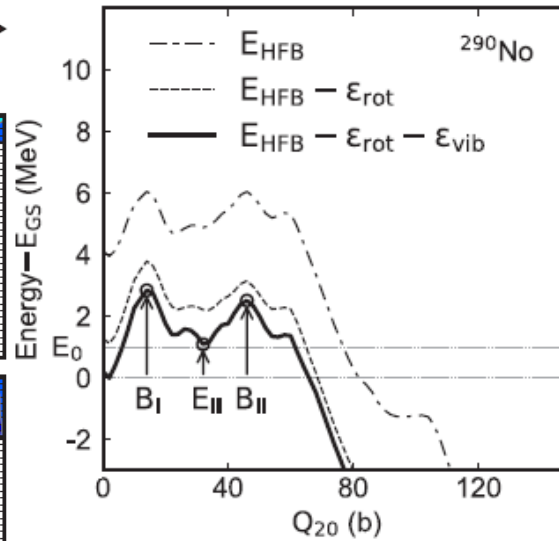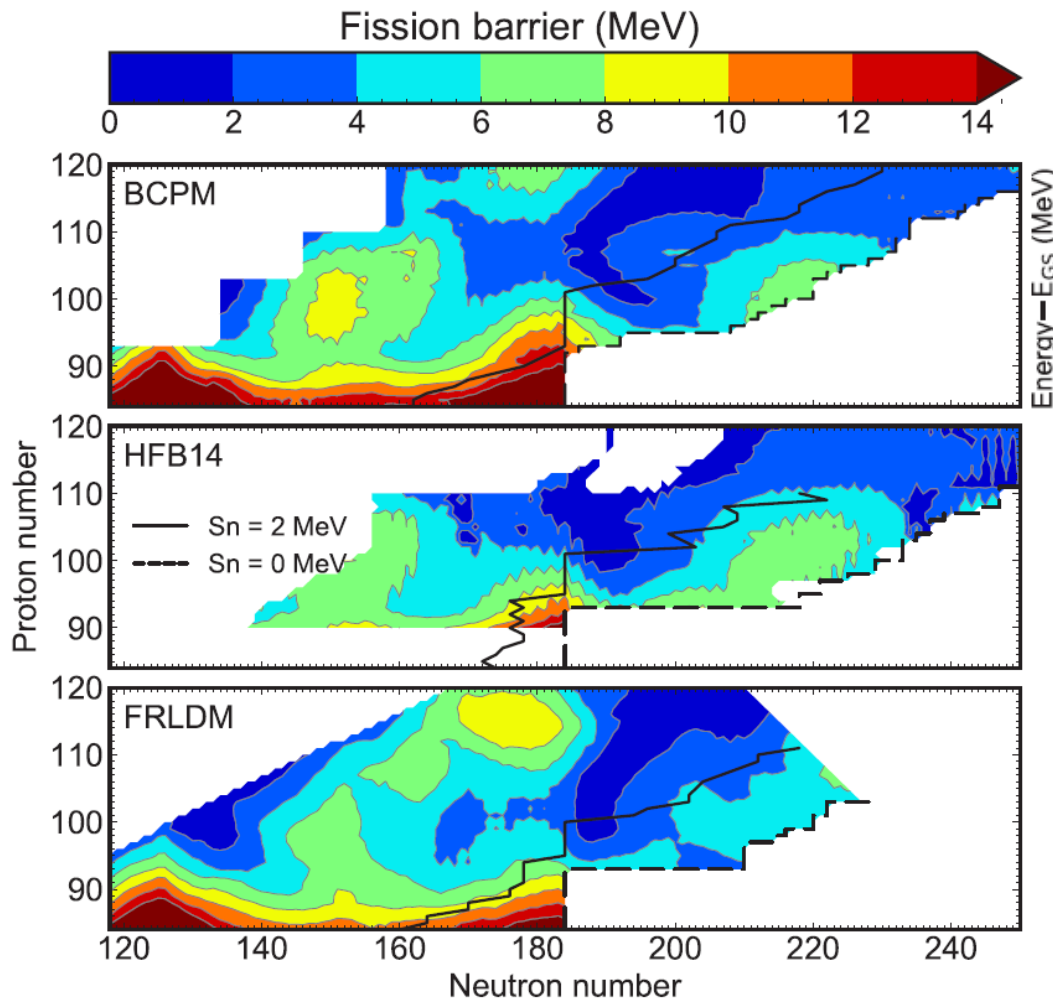
B_I: inner barrier
E_{II}: isomer
B_{II}: outer-barrier
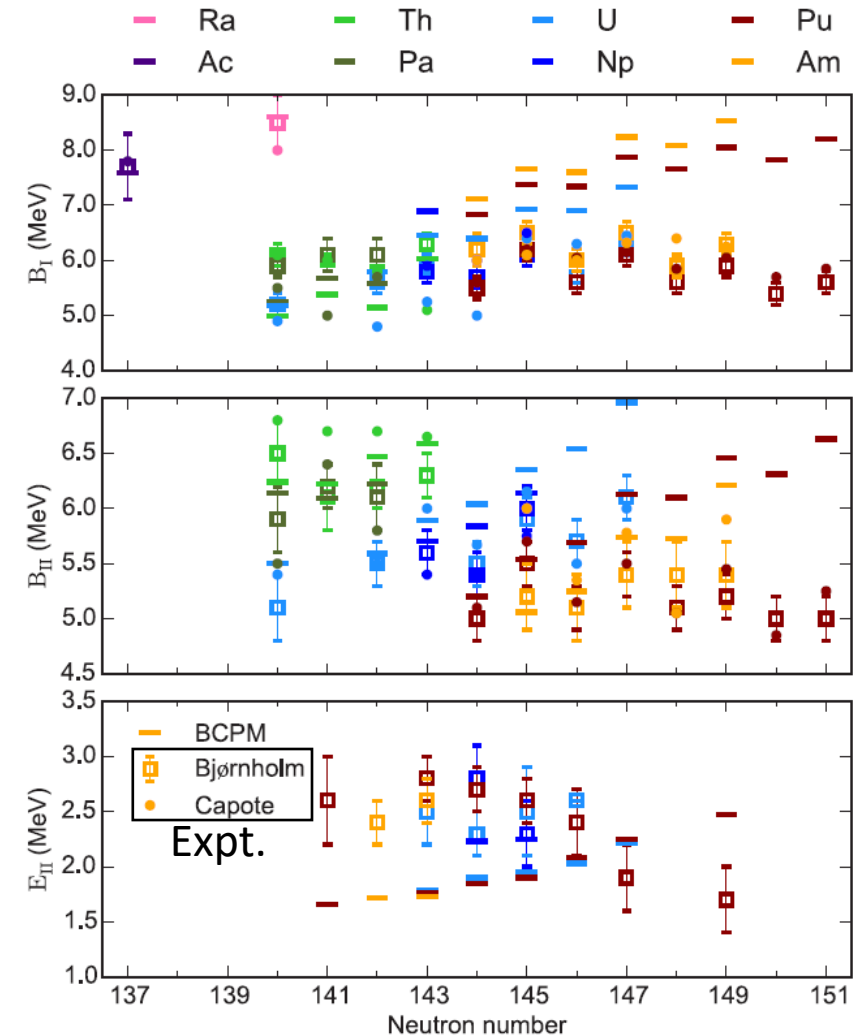
Spontaneous fission rate

- Spontaneous fission is akin to α decay (or proton or cluster decay, for that matter), where a barrier is "assaulted" at some rate and there is probability for tunneling through the barrier

- Here, the difference is that the potential is not from the nucleus, but rather the potential energy surface for the landscape of possible shapes.
The nucleus itself is tunneling through the barrier.

- $t_{1/2} = \dfrac{\ln(2)}{fP}$, where f is the assault frequency and P is the tunneling probability

- f corresponds to the rate at which the nuclear shape is changing, i.e. the frequency of surface oscillations, which is $\sim 10^{20} s^{-1}$ (Hill & Wheeler, Phys. Rev. 1953)

   (basically somewhere between a nucleon-orbit time and a full rotation time)

- This also gives us $\hbar\omega$ from $\Delta E \Delta t \geq \hbar$, with $\hbar\omega = \Delta E = \hbar/t \sim \hbar c/tc \sim 0.5 MeV$

- For the simplest case of a one-humped barrier, approximated as a inverted parabola, (due to the liquid-drop +single particle potential for deformation, See Lec. 4), the tunneling probability is (Hill & Wheeler, Phys. Rev. 1953)
$P = \dfrac{1}{1+\exp(2\pi E_f/\hbar\omega)}$

- So, "$t_{1/2} \approx 10^{-21}\exp(2\pi E_f / \hbar\omega)$"

Extremely sensitive to predictions of E_f and $\hbar\omega$ …so practically speaking this formula is useless
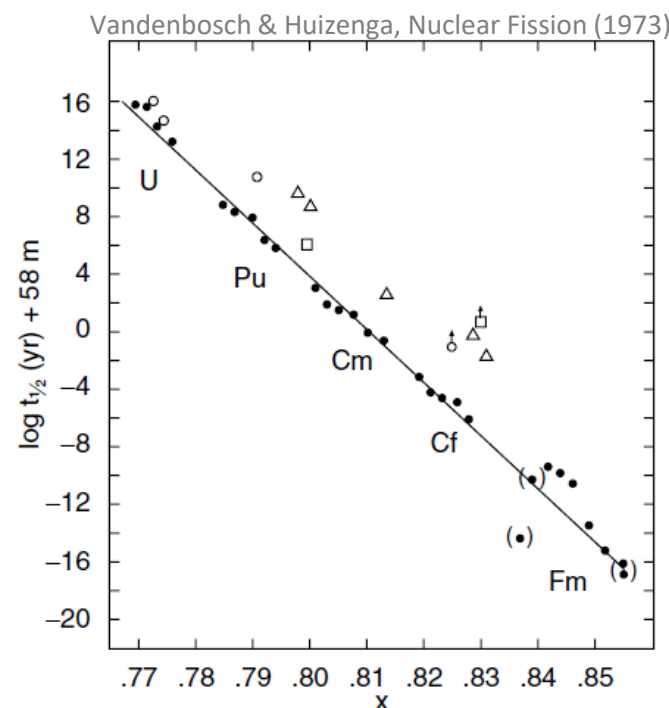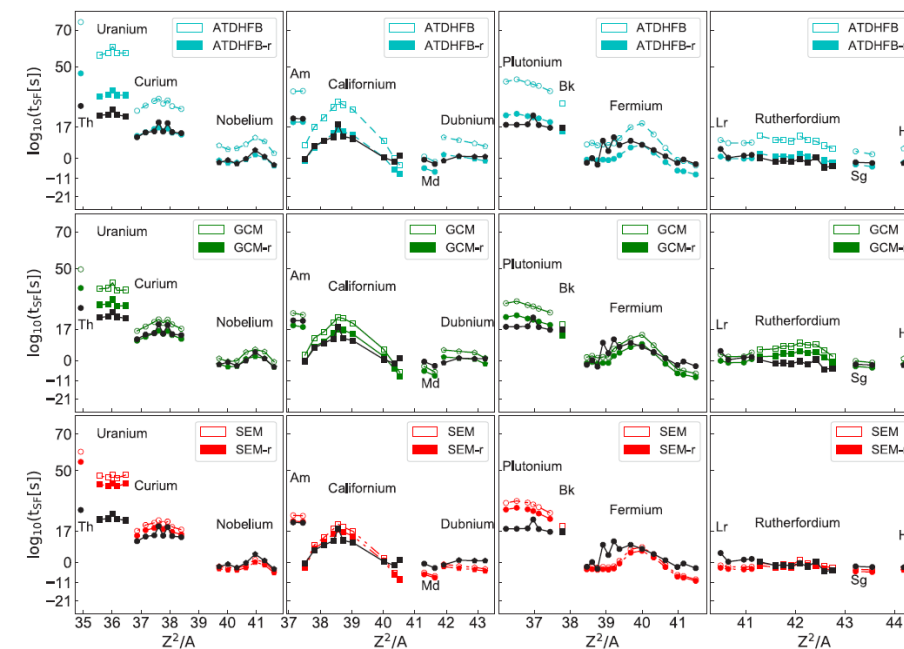
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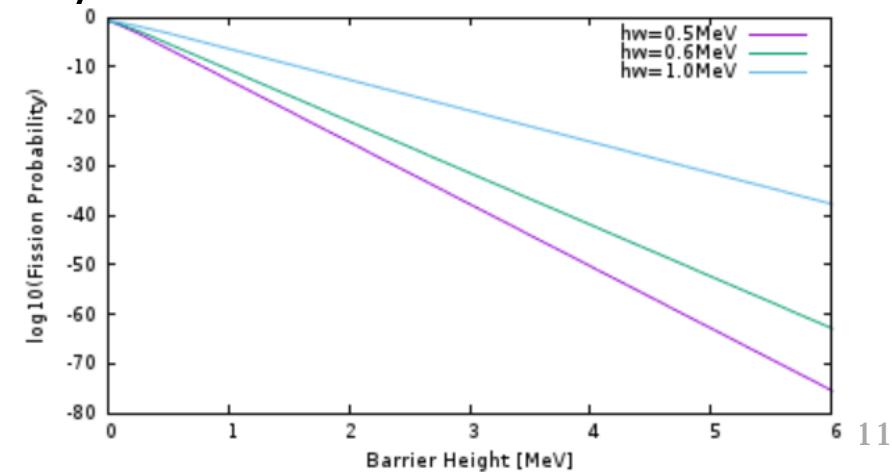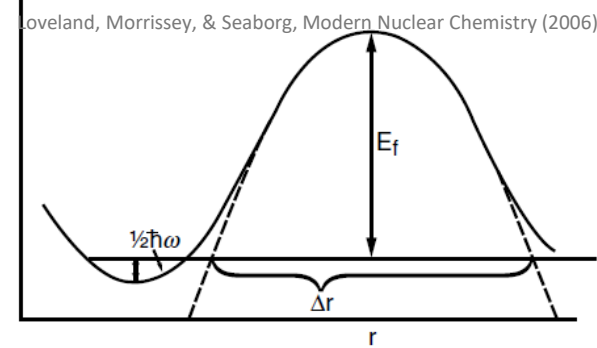
Spontaneous fission rate

- The height of the fission barrier is related to the fissionability parameter x *(recall x~1, i.e. Z^2/A~48, means fission immediately)*

- So without resorting to fancy calculations of E_f and $\hbar\omega$, the half-life for spontaneous fission can be estimated empirically

- But this method is extremely rough
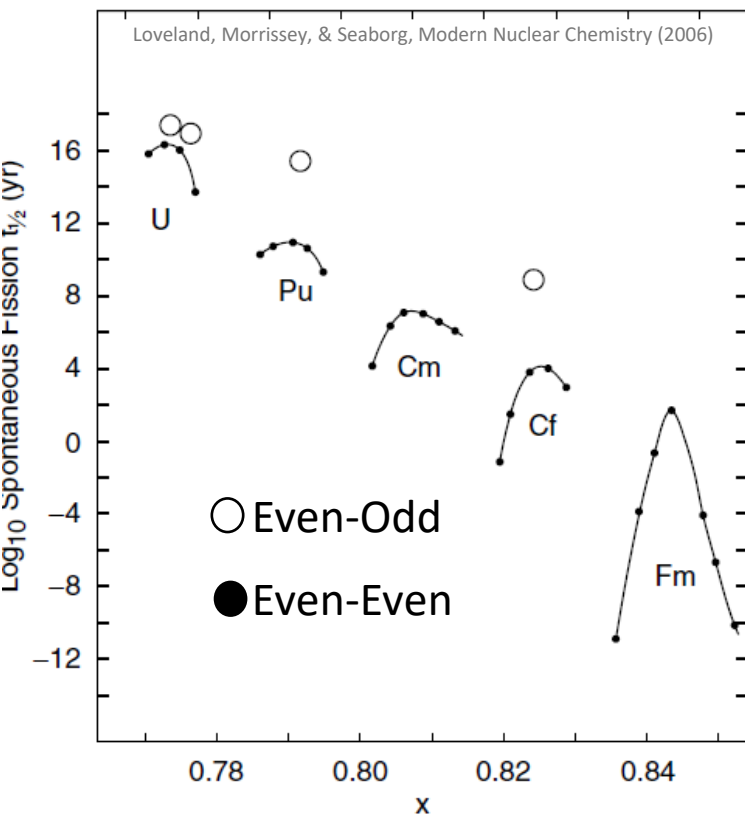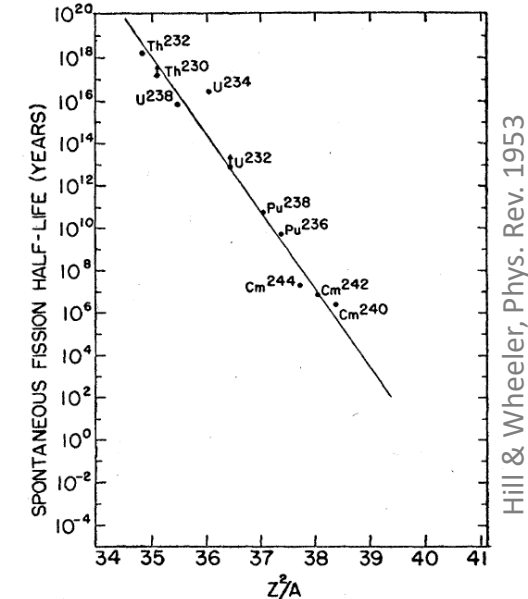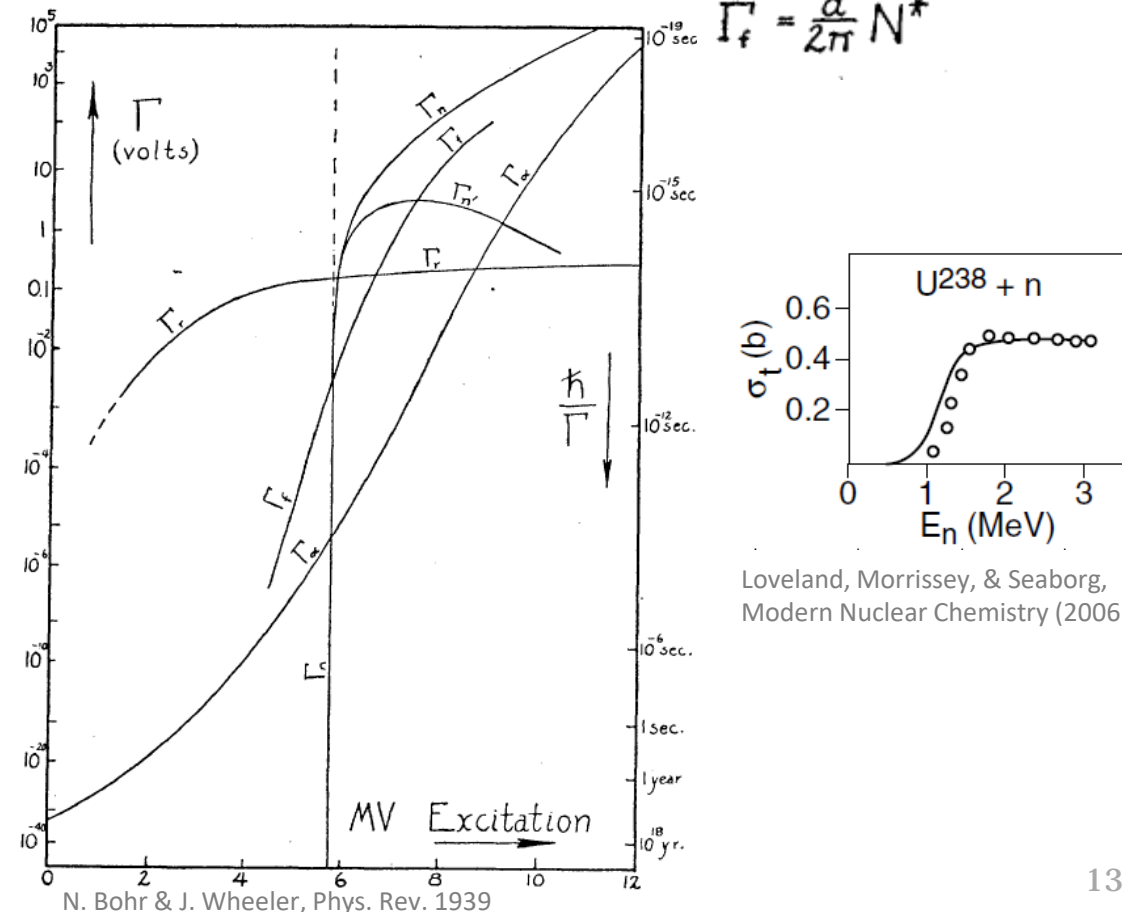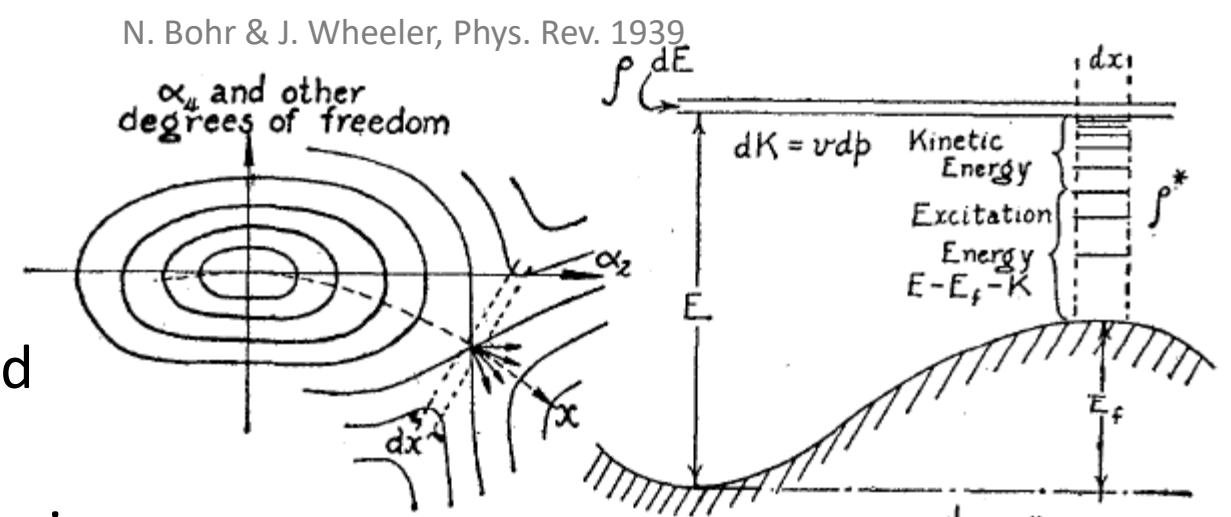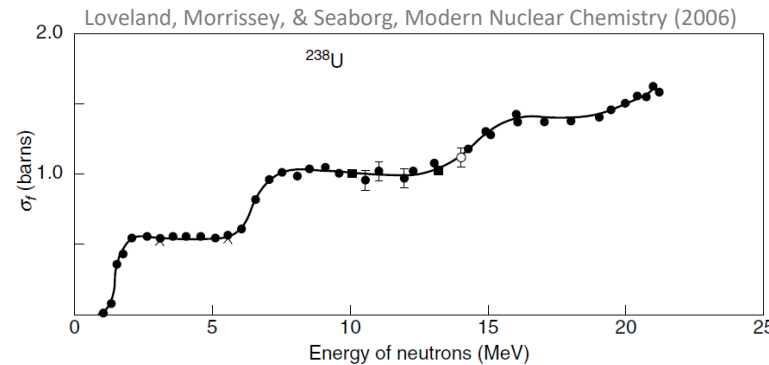
….using shell-corrected masses gives some improvement:

Even state-of-the-art models struggle to accurately reproduce data:

Loveland, Morrissey, & Seaborg, Modern Nuclear Chemistry (2006)

Vandenbosch & Huizenga, Nuclear Fission (1973)
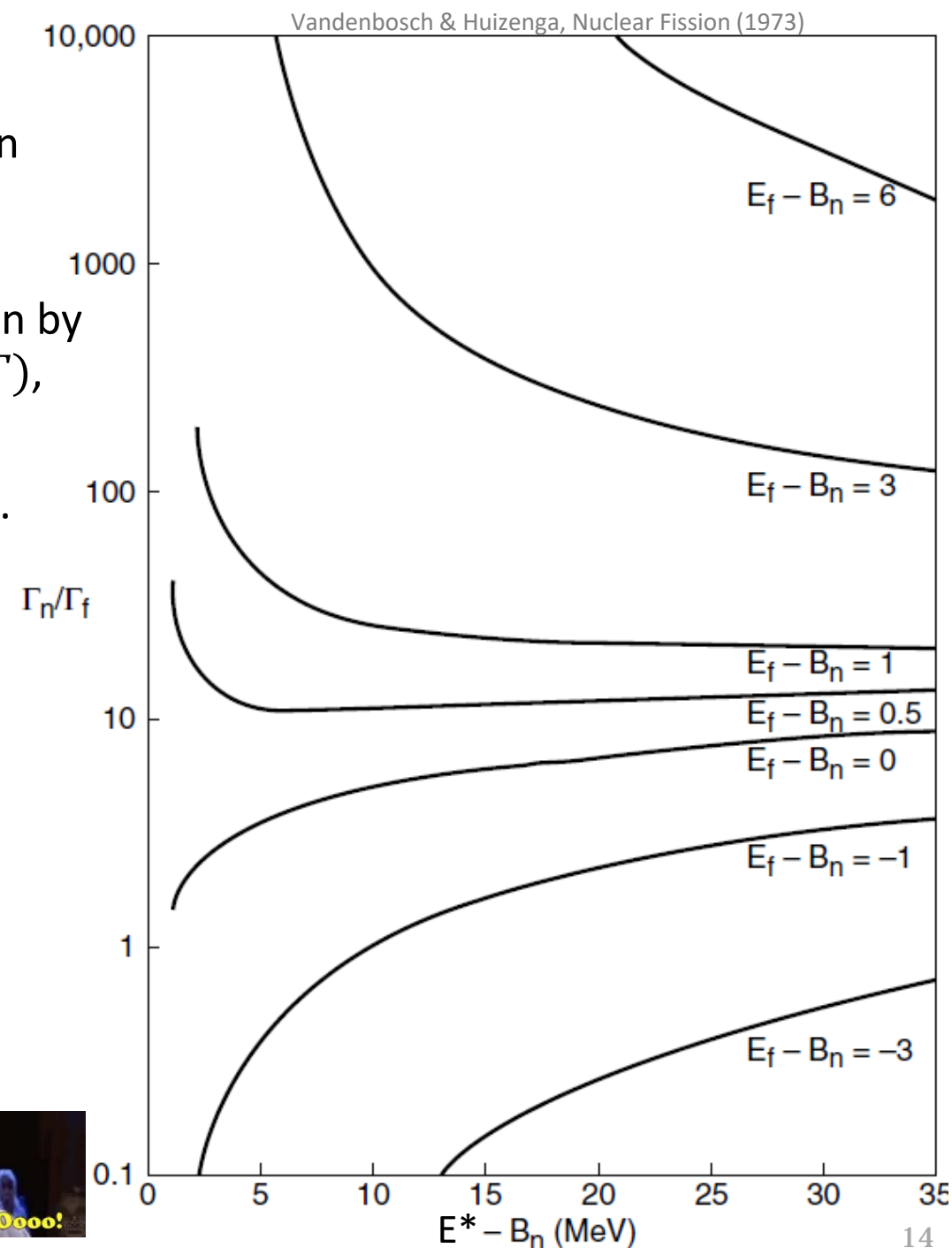
Fission decay branching ratio

- Fission is a means of de-excitation and, as such, competes with other decay modes

- Recall, the lifetime of an excited state can instead be specified in terms of a width $\Gamma = \hbar/\tau = \hbar\lambda$

- The fraction of decays proceeding through fission is
$$BR_f = \frac{\Gamma_f}{\sum_{modes}\Gamma_{mode}} = \frac{\Gamma_f}{\Gamma_f + \Gamma_n + \Gamma_\gamma + \Gamma_\alpha + \Gamma_{other\ c.p.}},$$
where the Γ_i are related to the density of final states available for that decay mode and the probability for transitioning from the initial to final state
(e.g. tunneling probability for charged particles. See Lectures 5,7,9.)

- When multi-neutron emission channels open, the fission cross section actually goes up!
[*2nd, 3rd, ...chance fission*]

N. Bohr & J. Wheeler, Phys. Rev. 1939

Loveland, Morrissey, & Seaborg, Modern Nuclear Chemistry (2006)

Loveland, Morrissey, & Seaborg, Modern Nuclear Chemistry (2006)

N. Bohr & J. Wheeler, Phys. Rev. 1939

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Fission decay branching ratio

- Since neutrons have no Coulomb barrier to tunnel through and can generally carry away more angular momentum than γ rays, the neutron decay branch is the primary competition for fission

- The probabilities for neutron emission and fission are roughly given by a Boltzmann distribution, $P_f \propto \exp(-B_f/T)$ and $P_n \propto \exp(-B_n/T)$, so the ratio of neutron emission to fission is $\propto \exp(B_f - B_n/T)$

- Higher excitation energies $E^* = E_{CM} + Q$ corresponds to higher T. At higher T, more of the phase space is sampled evening-out the competition between n-emission and fission. E.g. if n-emission is more likely, increasing E* makes fission more competitive

- Exact calculations involve detailed statistical model calculations, so the figure on the right can be used for a rough estimate

- E.g. ^{238}U bombarded by a 42MeV α

  - $E^* = 42\frac{238}{242} + (ME(U238) + ME(\alpha) - ME(Pu242))c^2 = 36.3 MeV$
  - $B_n(Pu242) = (ME(Pu241) + ME(n) - ME(Pu242))c^2 = 6.3 MeV$
  - $E_f(Pu242) = 3.3 MeV$ *[From Myers & Swiatecki PRC 1999 fit]*
  - $E^* - B_n = 30 MeV;\ E_f - B_n = -3 \quad \rightarrow \frac{\Gamma_n}{\Gamma_f} \sim 0.5$

- ... so fission occurs 2/3 of the time for this case

(actual answer [J. Wing et al. Phys Rev 1959] is ~80% of the time)

Γ_n/Γ_f plotted versus $E^* - B_n$ (MeV), with curves labeled $E_f - B_n = 6$, $E_f - B_n = 3$, $E_f - B_n = 1$, $E_f - B_n = 0.5$, $E_f - B_n = 0$, $E_f - B_n = -1$, $E_f - B_n = -3$.

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The fission decay branching ratio is of interest for r-process nucleosynthesis and "fission recycling"
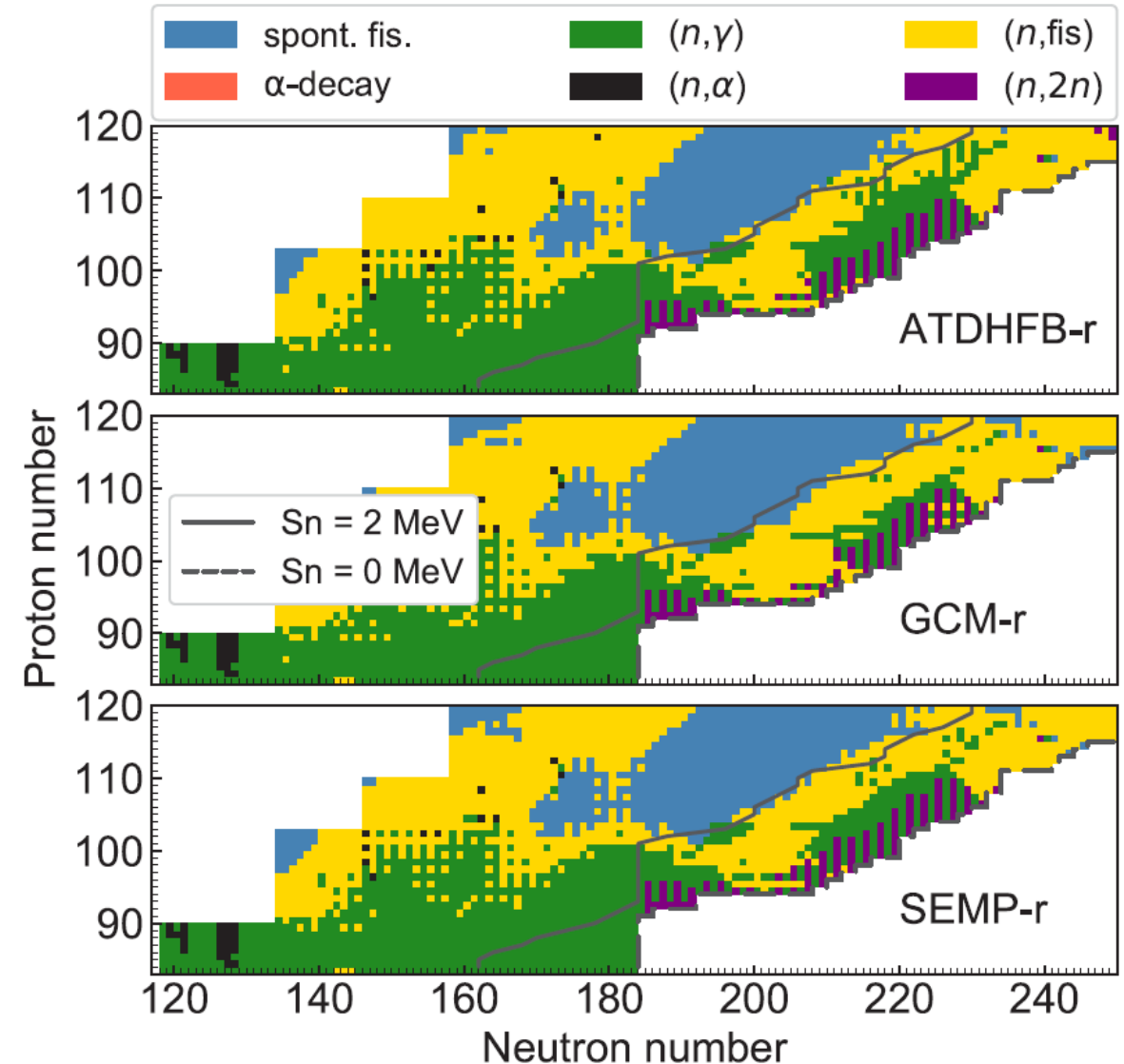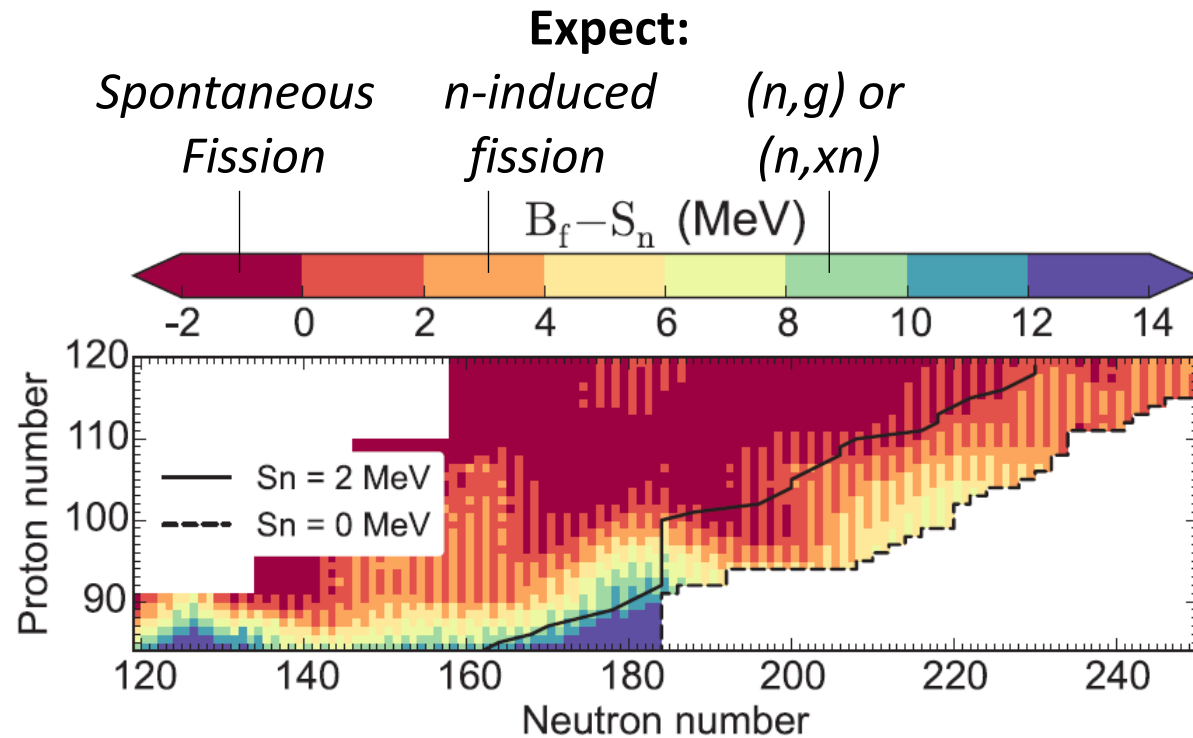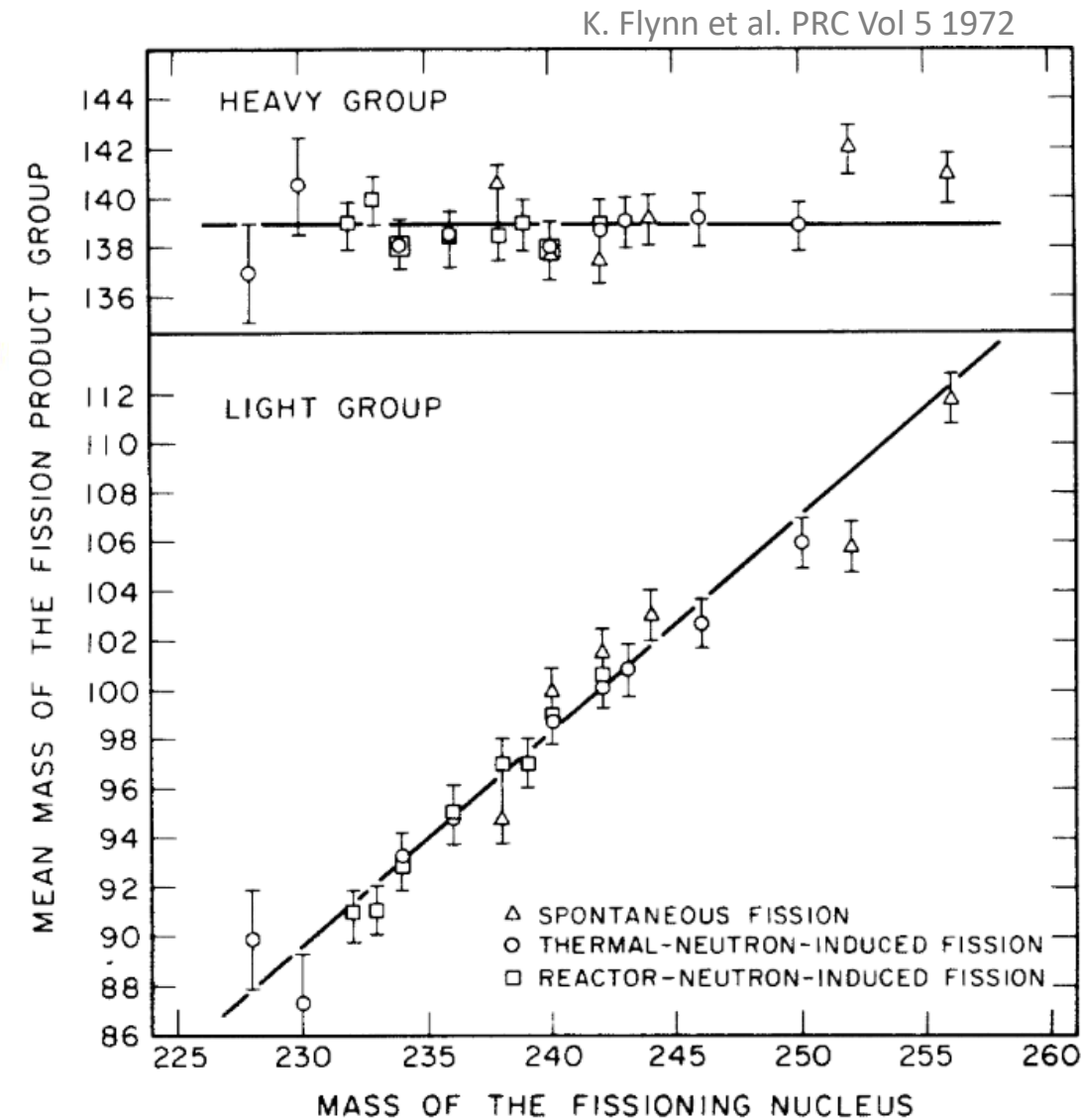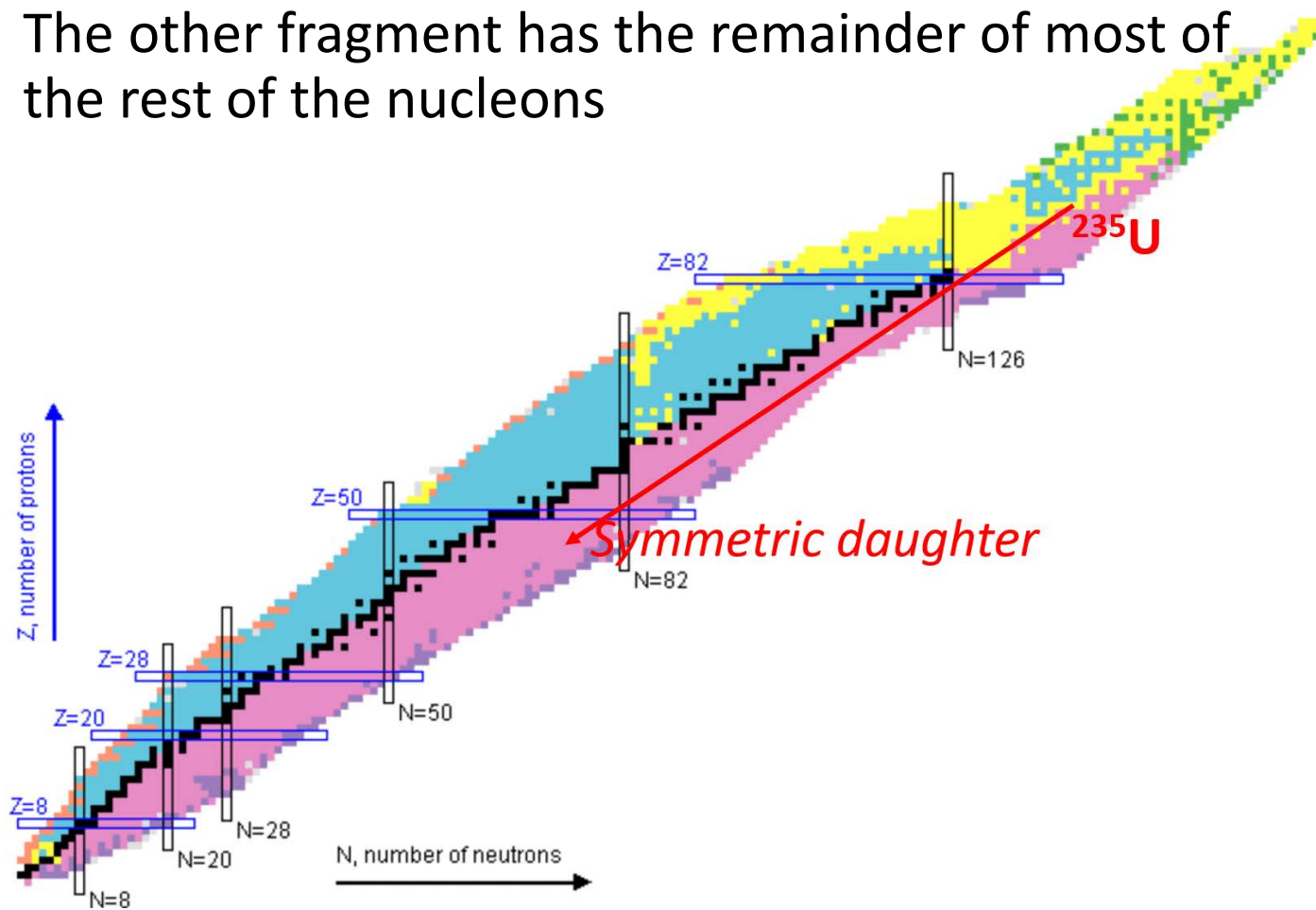
Fission fragment mass distribution

- To first order, the fission fragments will be those that maximize the total energy release
  [which can be estimated from the liquid drop model]

- This would always result in a case of a symmetric distribution of the matter

- However, often the observed mass distributions are instead quite asymmetric:

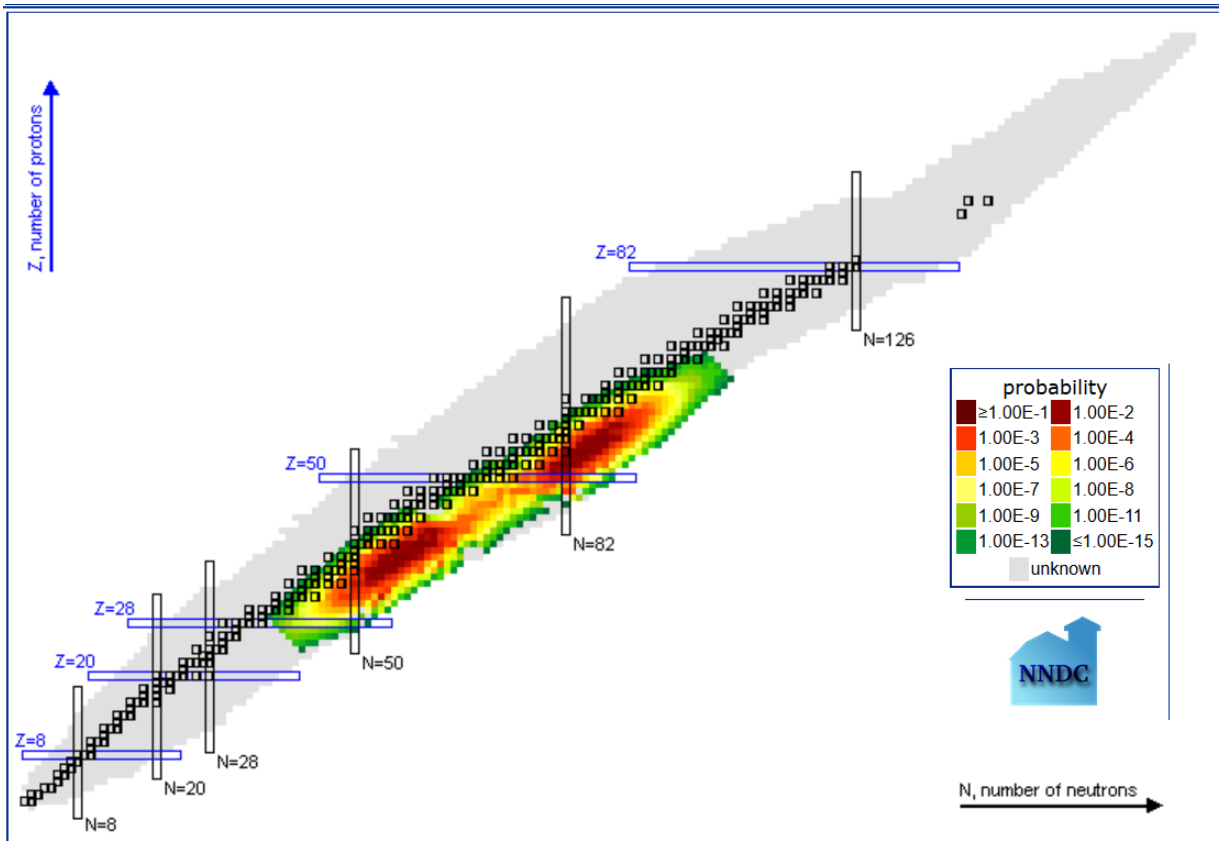- The individual fragments approximately maintain the original A/Z *(because neutrons and protons separate in the same way)*, but often high-A and low-A peaks exist

Seaborg & Loveland, The Elements Beyond Uranium (1990)
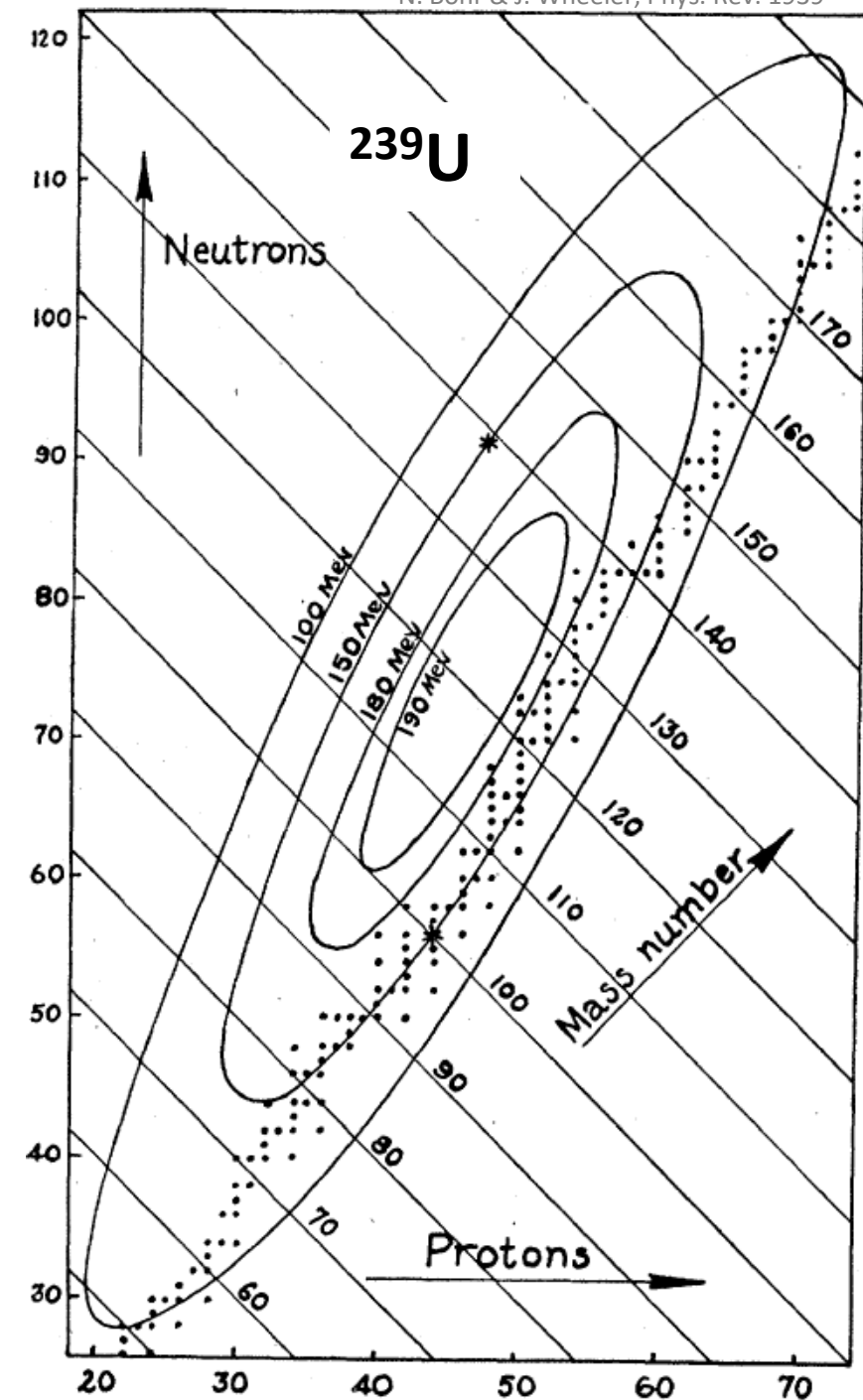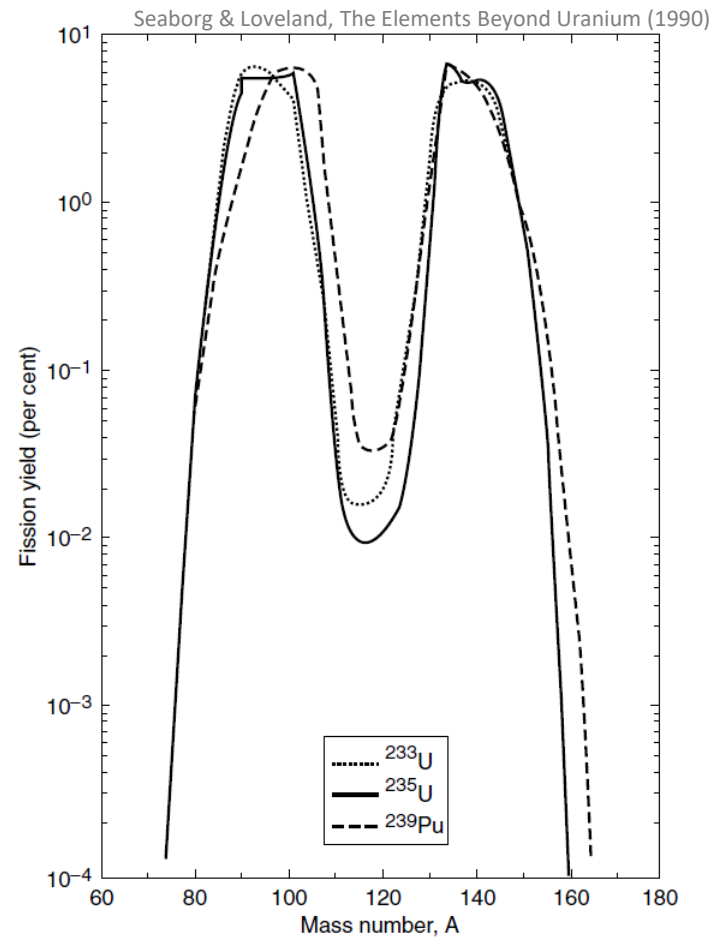
Fission fragment mass distribution

- The culprit for the asymmetric mass distribution are the $Z = 50$, $N = 82$ shell closures, which favors nuclei in this range for one of the fission fragments.

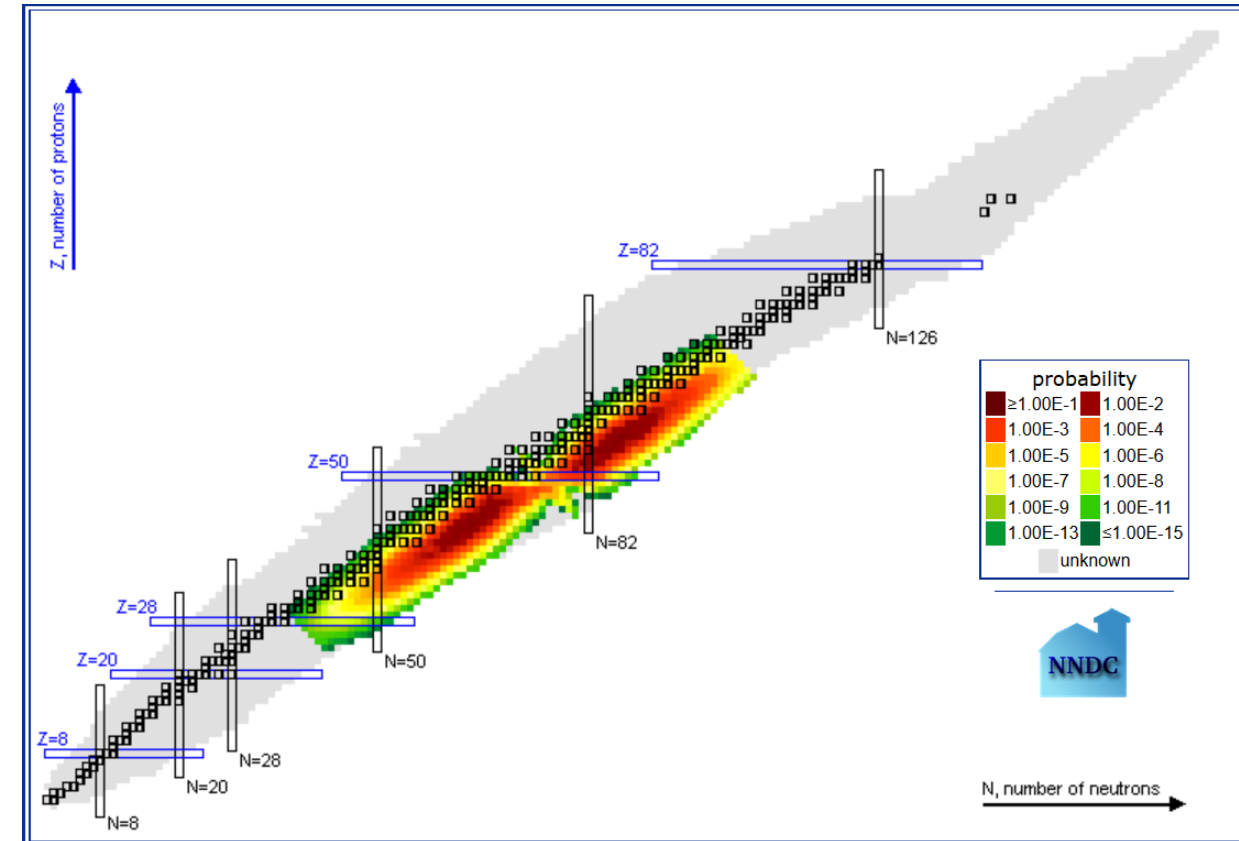- The other fragment has the remainder of most of the rest of the nucleons

Fission fragment mass distribution, *Examples*

²³⁵U induced fission (thermal neutrons)

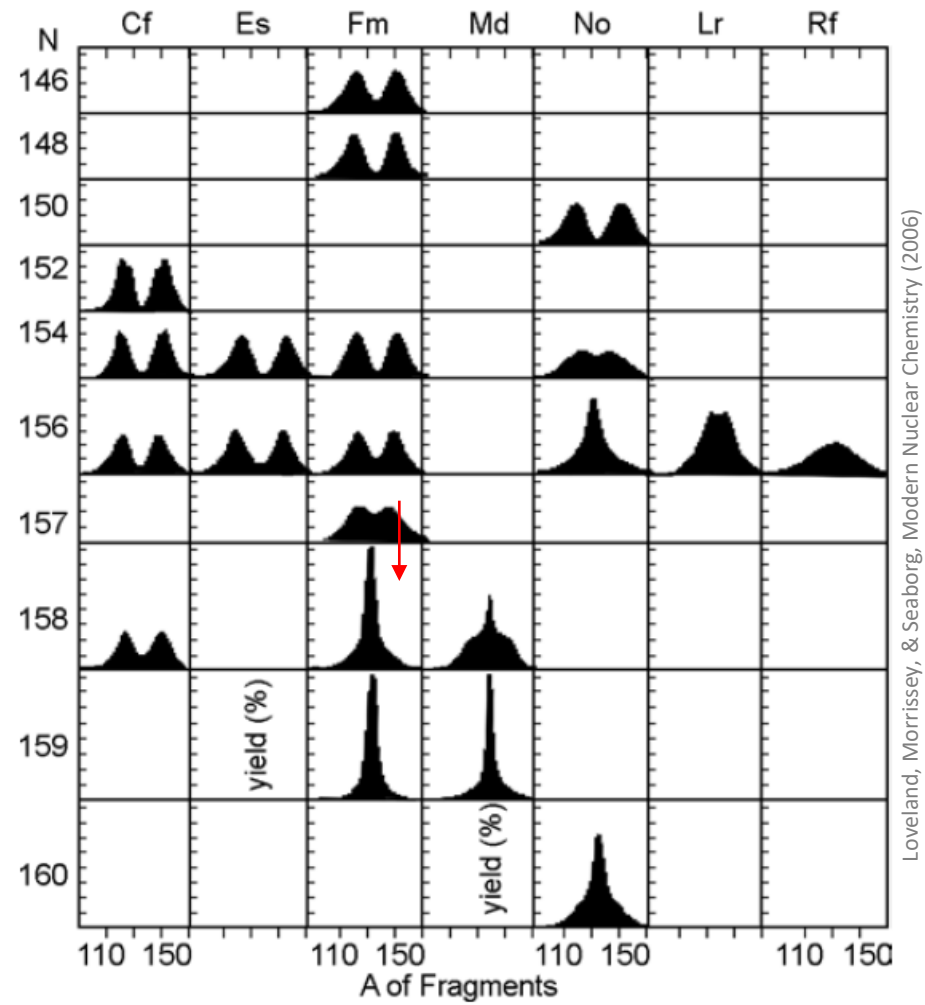## ²⁵²Cf spontaneous fission

Fission fragment mass distribution

- However, the signature of the closed shells is erased if the symmetric fragment distribution is too far from the shell closure...

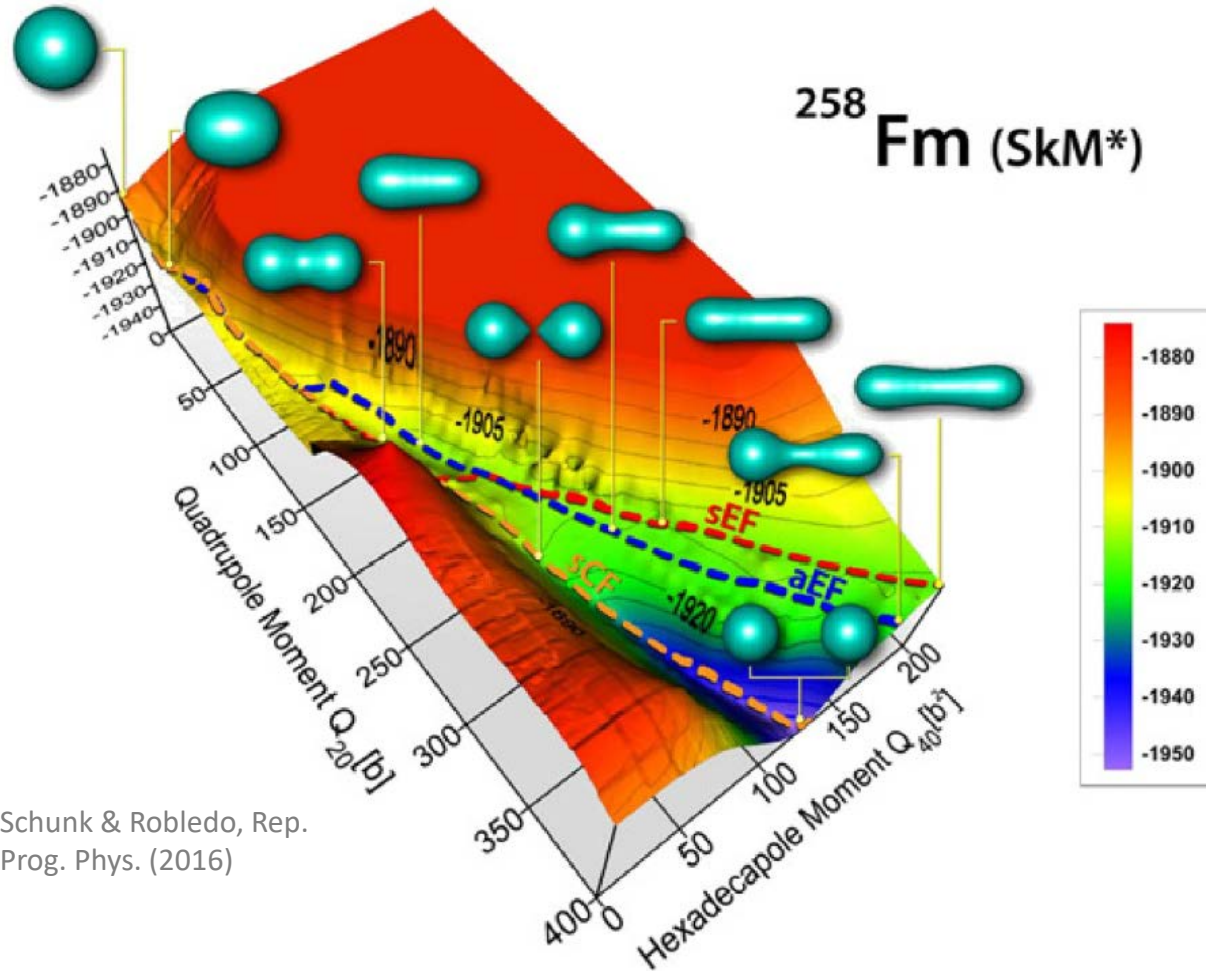- ...or if the excitation energy of the fissile nucleus is high *(e.g. from a high-E incoming neutron)*

Loveland, Morrissey, & Seaborg, Modern Nuclear Chemistry (2006)

Loveland, Morrissey, & Seaborg, Modern Nuclear Chemistry (2006)
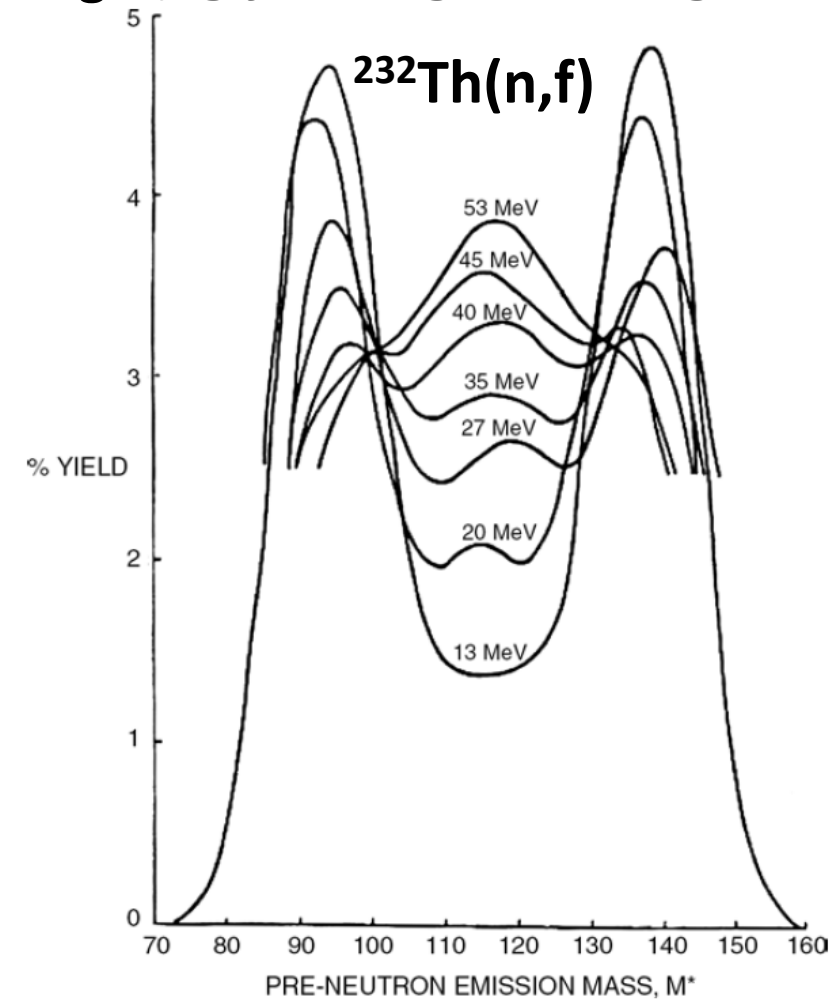
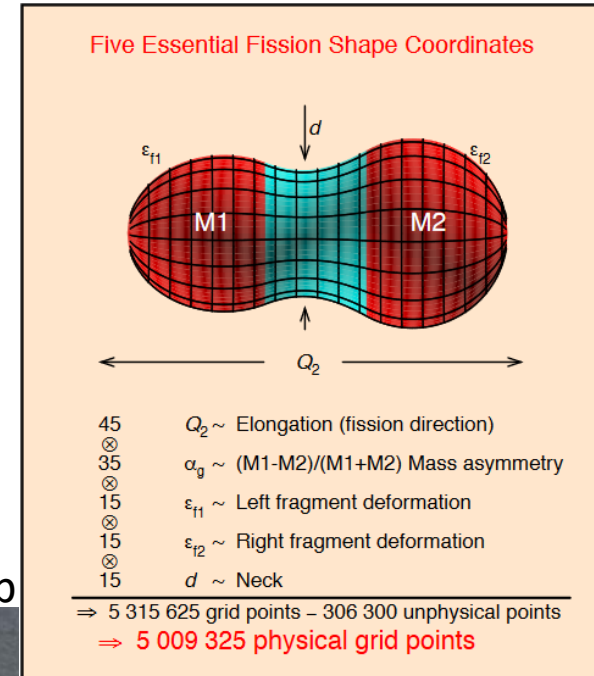# Induced-fission fragment mass distribution is independent of what did the inducing ("the Bohr Hypothesis")

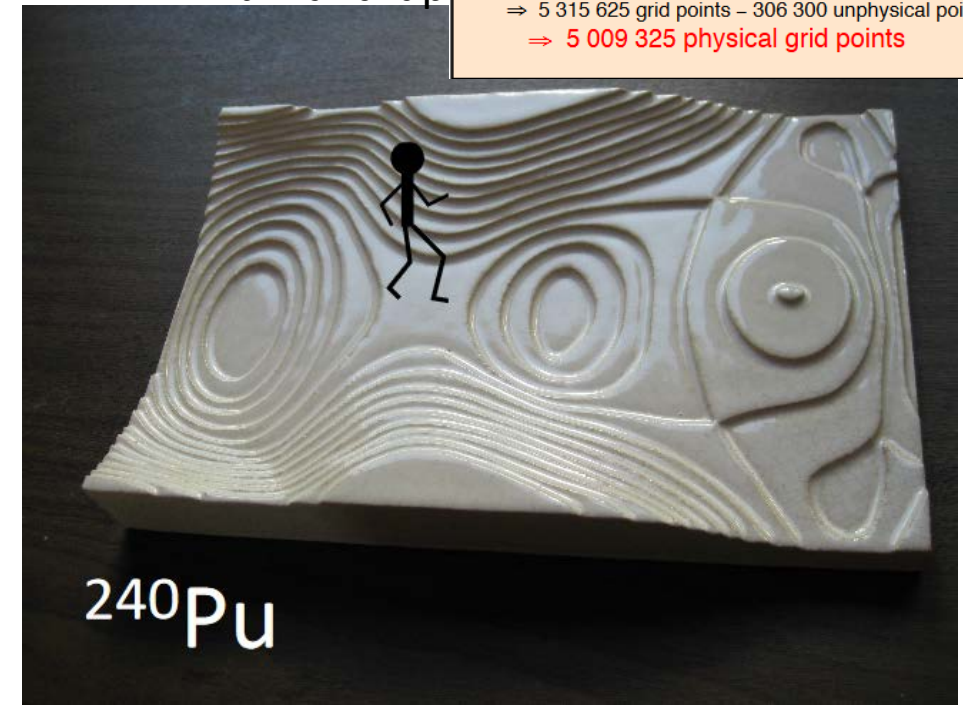*we'll revisit this in the Statistical Reactions lecture*
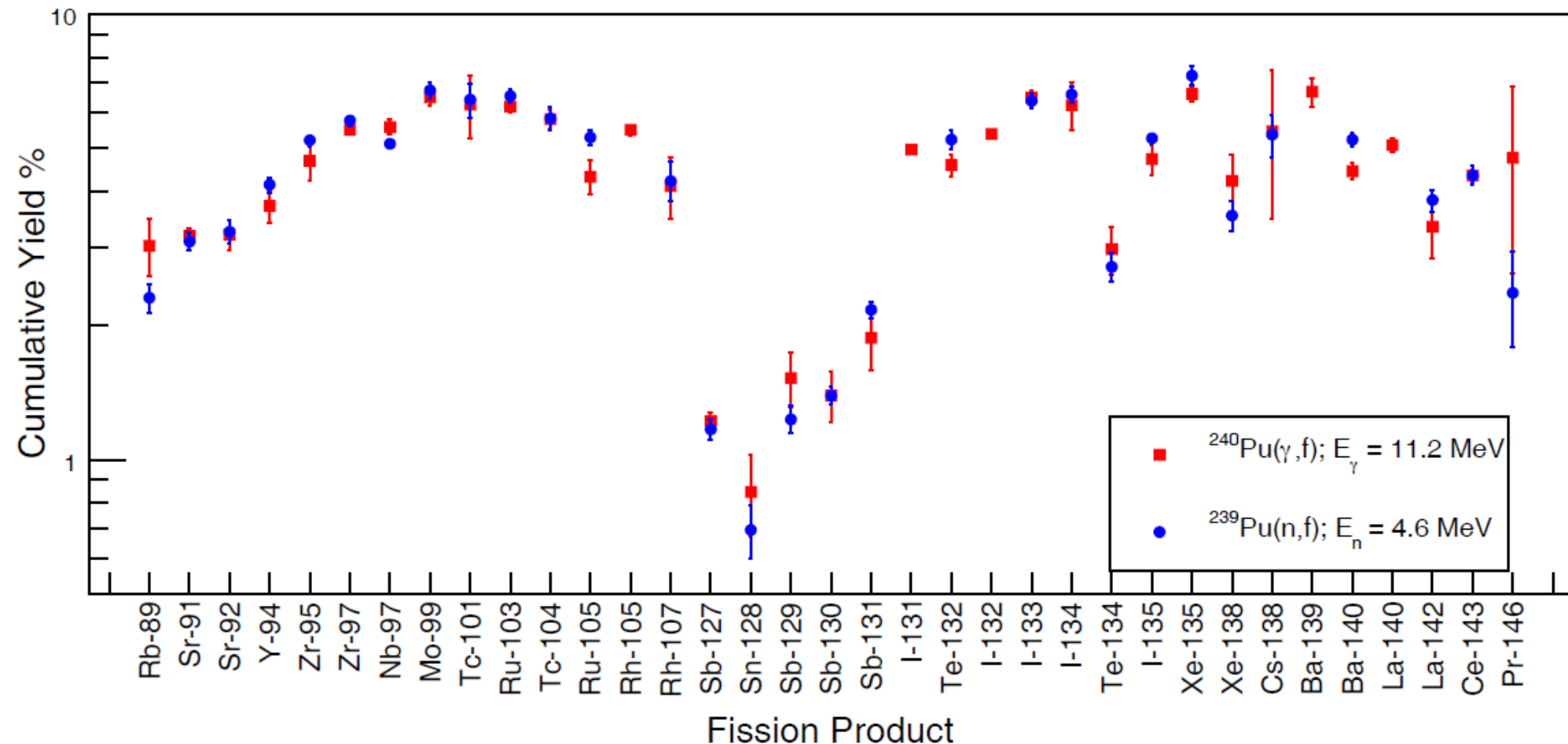
Modern fission fragment calculations:
Random walk in a shape-based energy space

^{258}Fm (SkM*)

Schunk & Robledo, Rep. Prog. Phys. (2016)

Five Essential Fission Shape Coordinates

45 ⊗	$Q_2 \sim$ Elongation (fission direction)
35 ⊗	$\alpha_g \sim$ (M1-M2)/(M1+M2) Mass asymmetry
15 ⊗	$\varepsilon_{f1} \sim$ Left fragment deformation
15 ⊗	$\varepsilon_{f2} \sim$ Right fragment deformation
15	$d \sim$ Neck

⇒ 5 315 625 grid points – 306 300 unphysical points
⇒ 5 009 325 physical grid points

J. Randrup

^{240}Pu

This walk is extremely sensitive to the nuclear temperature, which factors into the probability of taking a given step: $P \sim exp(-\Delta U/T)$
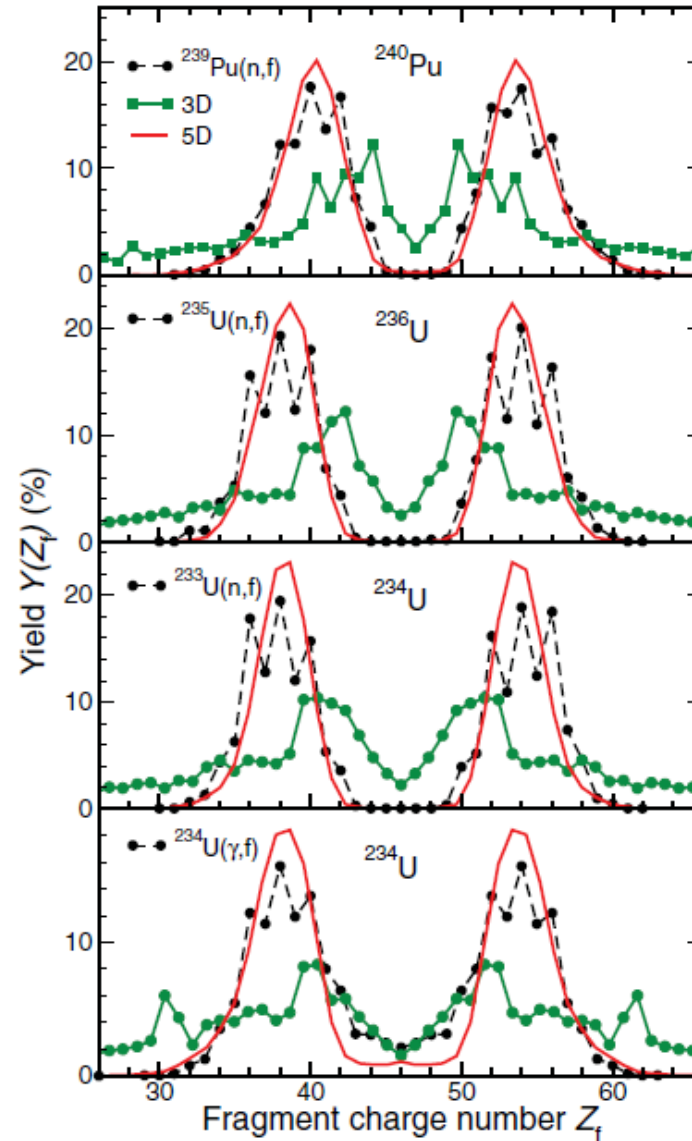
Modern fission fragment calculations:
Random walk in a shape-based energy space

Second minimum

^{222}Th

Outer saddle

^{222}Th

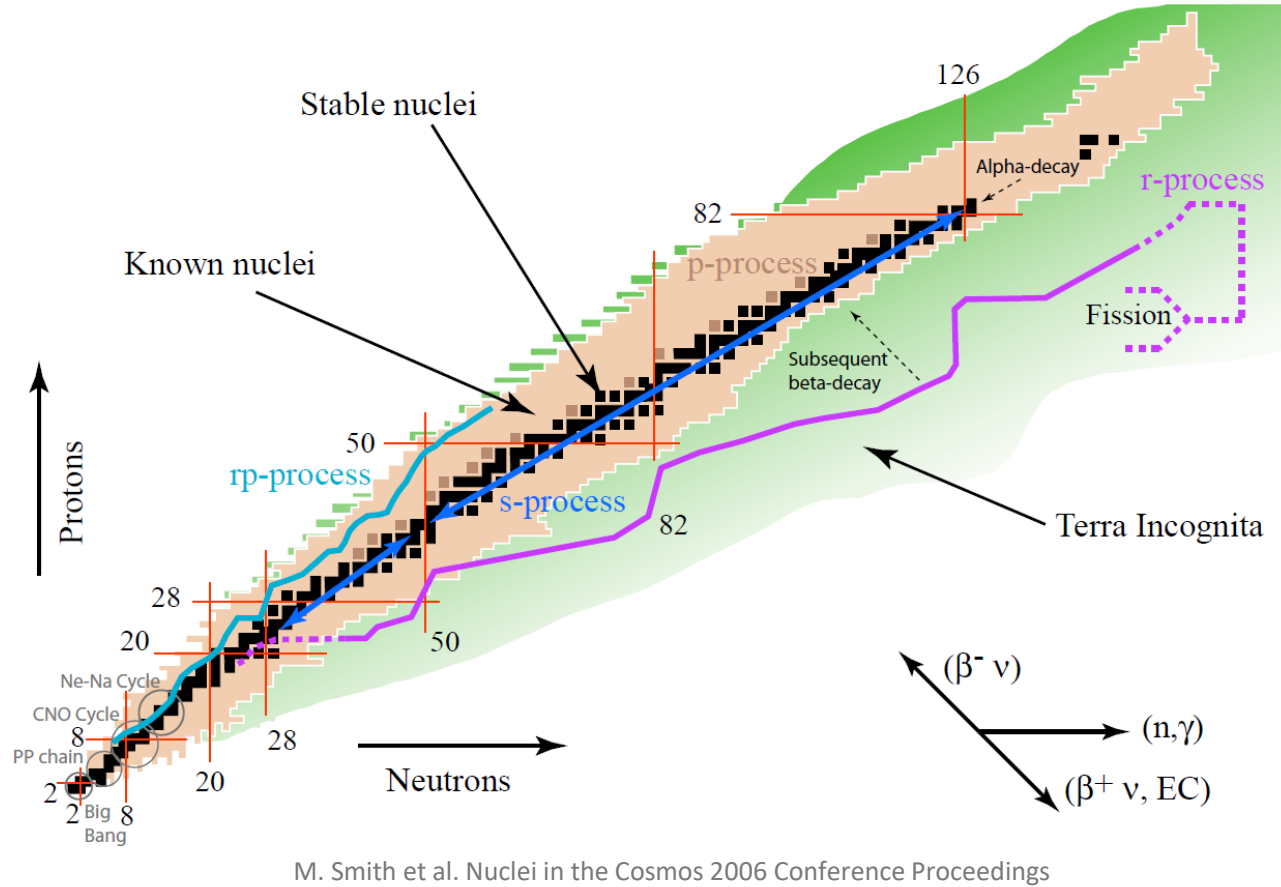Most probable fragment mass division

^{111}Re ^{111}Re
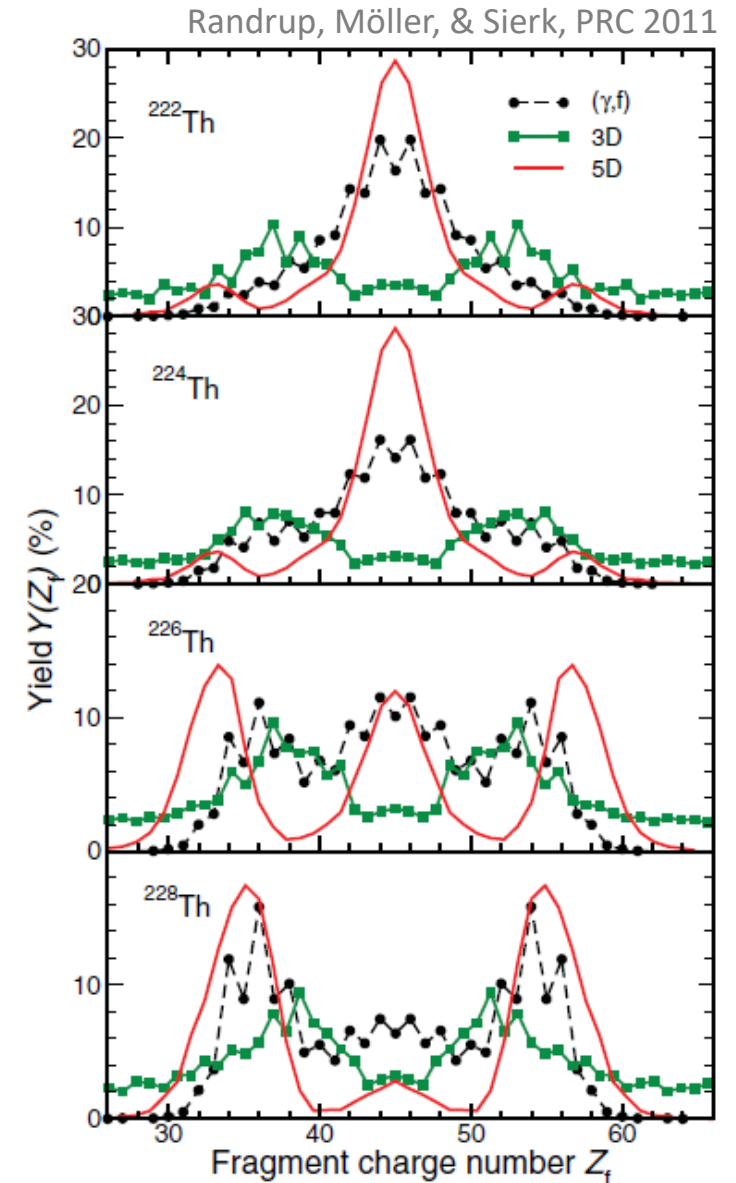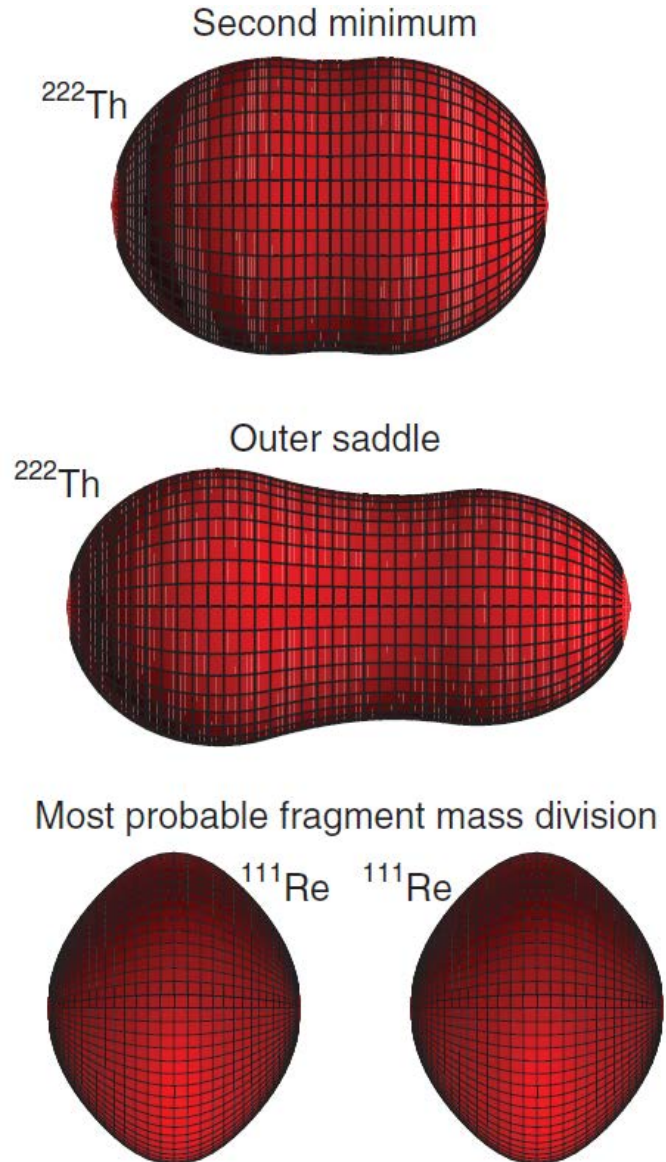
Fission fragment mass distribution, *systematics*

Calculated with FRLDM
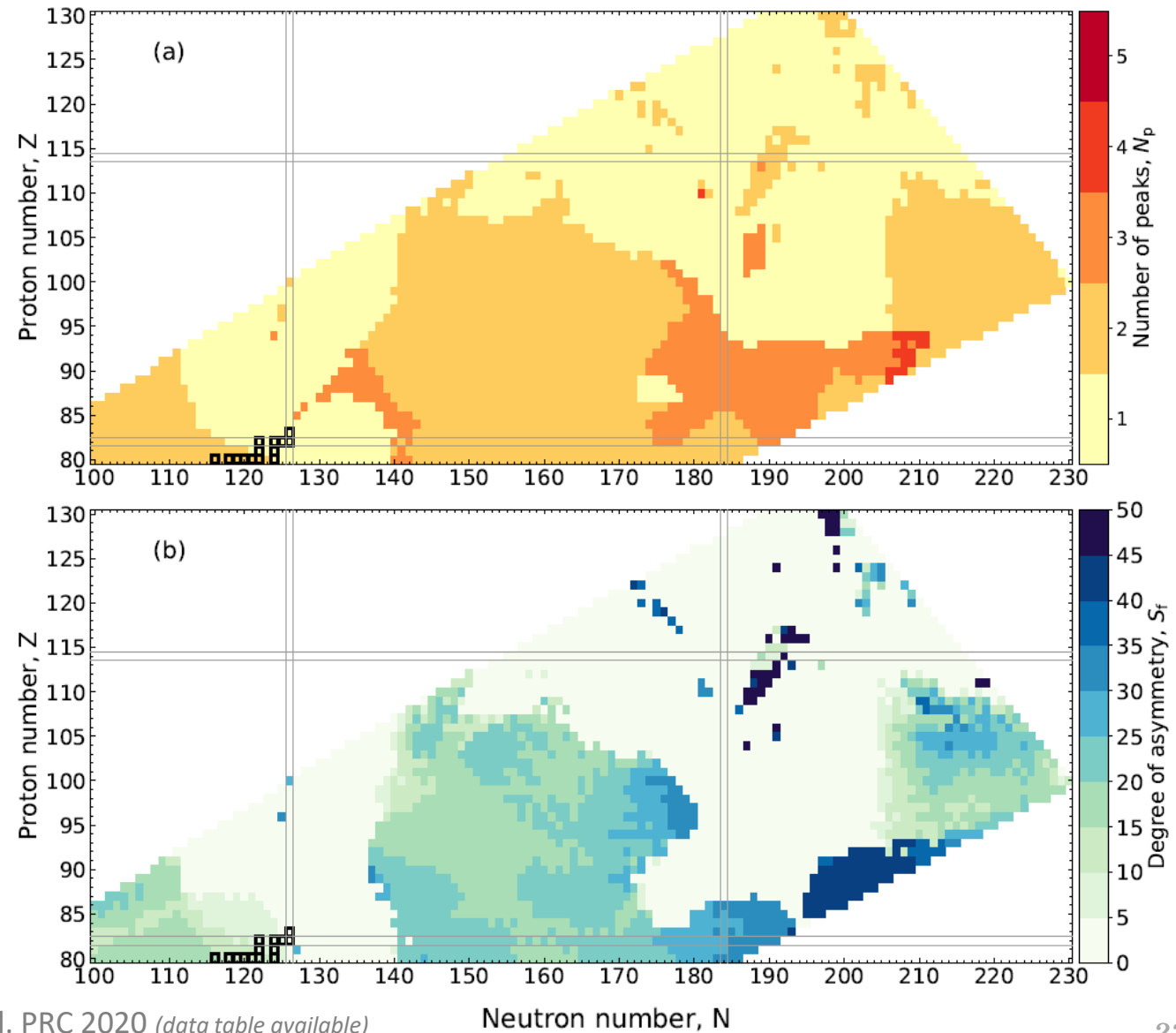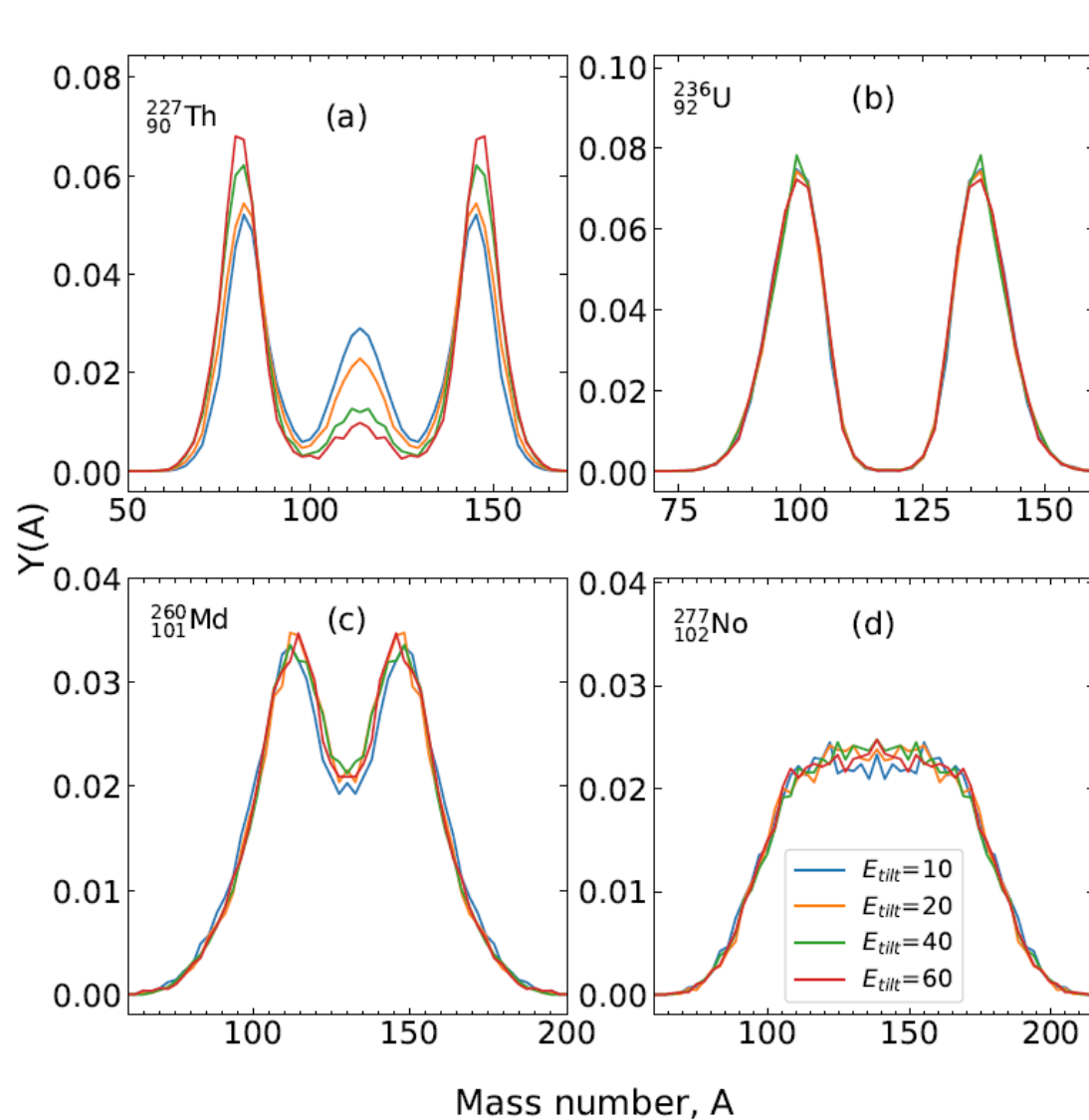
Why care about the fragment distribution?
r-process fission "recycling" & kilanova light curves

The kilanova light curve is powered by radioactive heating. Amount of heating and heat deposition efficiency varies by type of heating:

M. Smith et al. Nuclei in the Cosmos 2006 Conference Proceedings

Y.L. Zhu et al. ApJ2021

(b) $Y_e = 0.16$
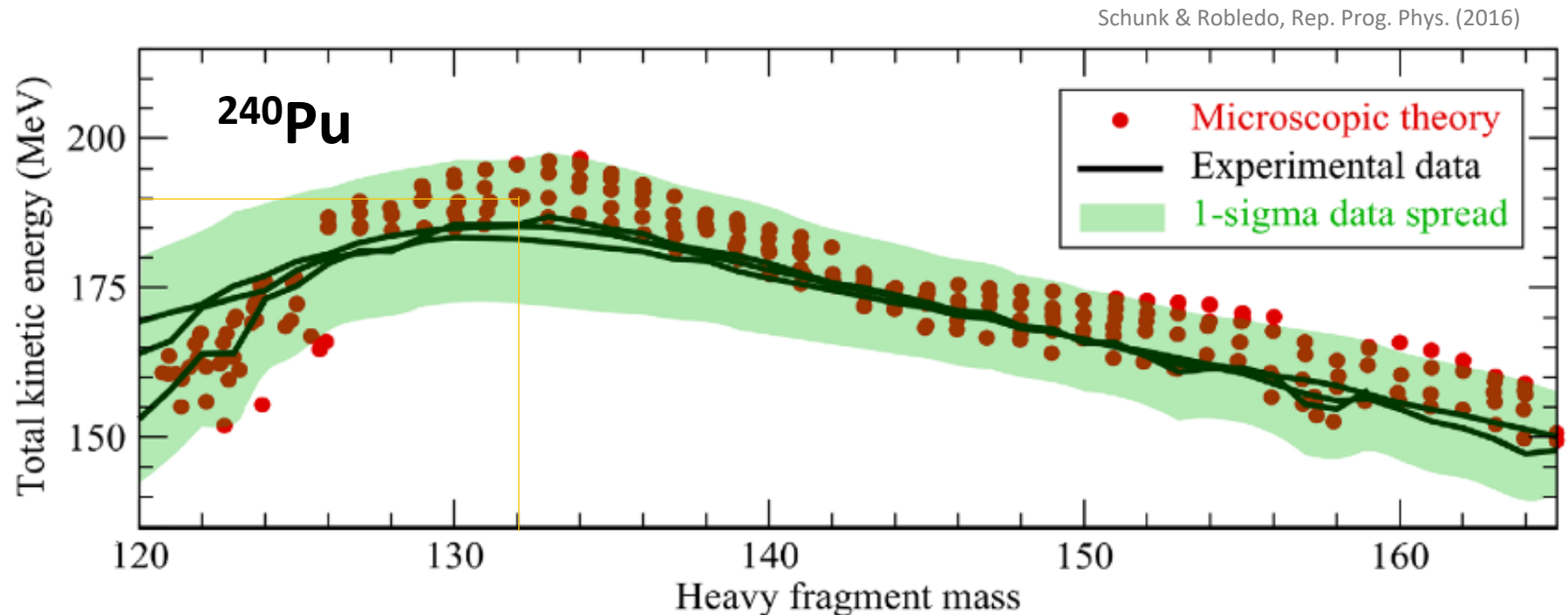
Fission fragments: *kinetic energy*

- The total kinetic energy of fission products will roughly be the coulomb repulsion energy of the two main fission fragments,

  - $TKE = \dfrac{Z_1 Z_2 \alpha \hbar c}{1.8\left(A_1^{1/3} + A_2^{1/3}\right)}$ …*where r$_0$=1.8fm is used instead of 1.2 because of the strong deformation at scission*

- E.g. ^{240}Pu

  - $A/Z \approx 2.55$. If $Z_{high} = 50, N_{high} = 82$, then $Z_{low} = 94 - 50 = 44, N_{low} = 240 - 132 - 44 = 64$

  - $TKE = \dfrac{(50)(44)\alpha \hbar c}{1.8\left(132^{1/3} + 108^{1/3}\right)} \approx 178 MeV$

Schunk & Robledo, Rep. Prog. Phys. (2016)

Fragment de-excitation & neutron emission

- The total excitation energy is obtained from energy conservation, $TXE = \left[E_{cm} + B_n + Q_f\right] - TKE$, where the Q-value plus any reaction energy can pay for the fragment kinetic energy and to unbind a neutron

- Typically TXE is several, up to tens of, MeV

- A rough estimate is that $\sim 10 MeV$ of excitation energy is removed from γ emission and β-decay and the associated ν emission, and the rest is for neutrons

- It takes $\sim 5 MeV$ to create 1 neutron, so a few neutrons are emitted per fission

- *Deviations from the linear trend with mass (b/c TXE is higher with A) are mostly due to shell corrections*

P. Jaffke et al. PRC 2018

Loveland, Morrissey, & Seaborg, Modern Nuclear Chemistry (2006)
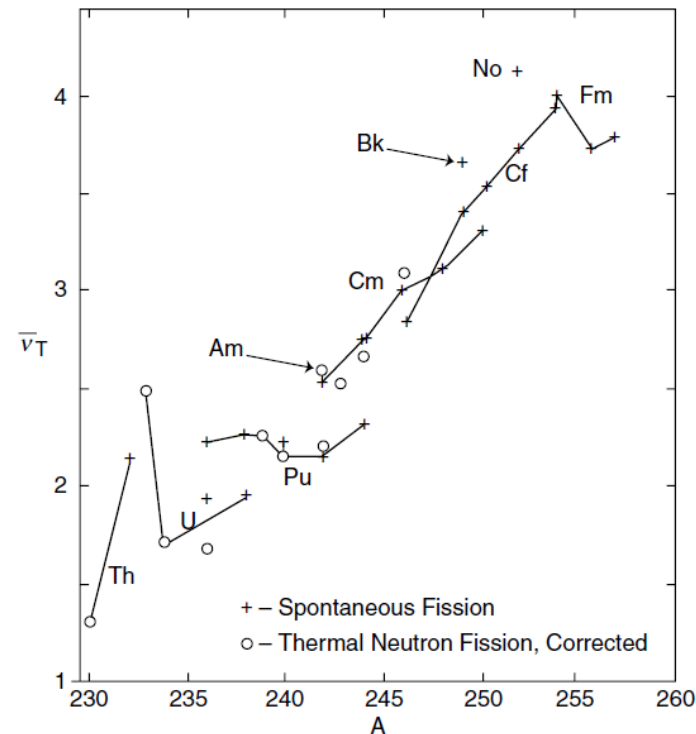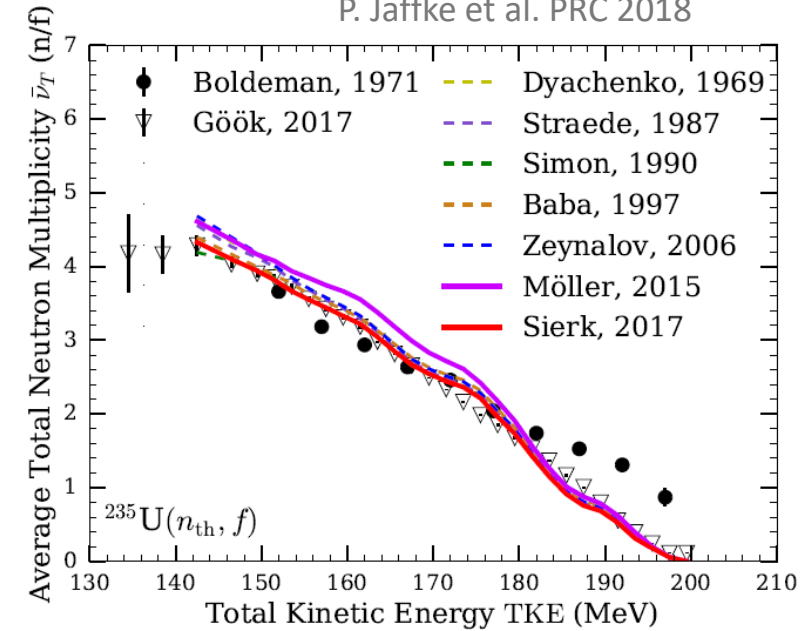
Fission neutrons kinetic energy

- Neutrons are emitted from the nearly fully accelerated fission fragments and so are emitted along the direction of the fragments

- The neutron kinetic energy (typically $\sim 2 MeV$) is just described by a Maxwell-Boltzmann distribution, because the nucleus is "evaporating" neutrons
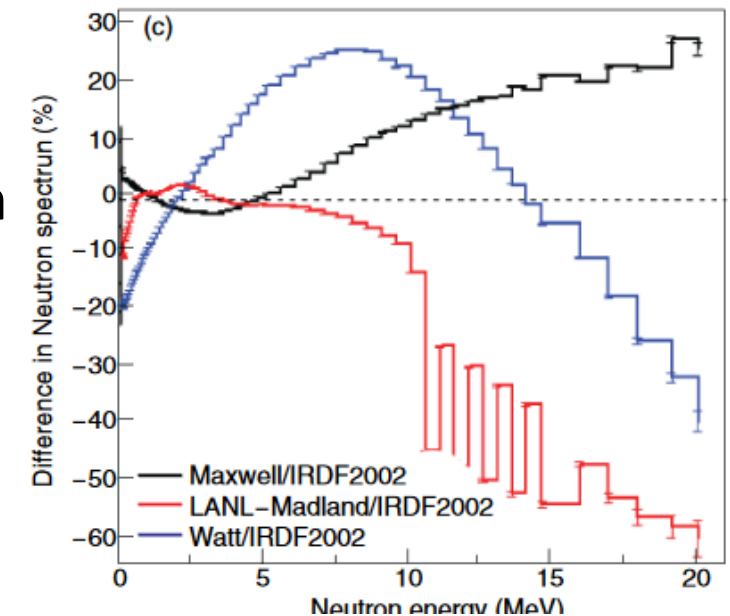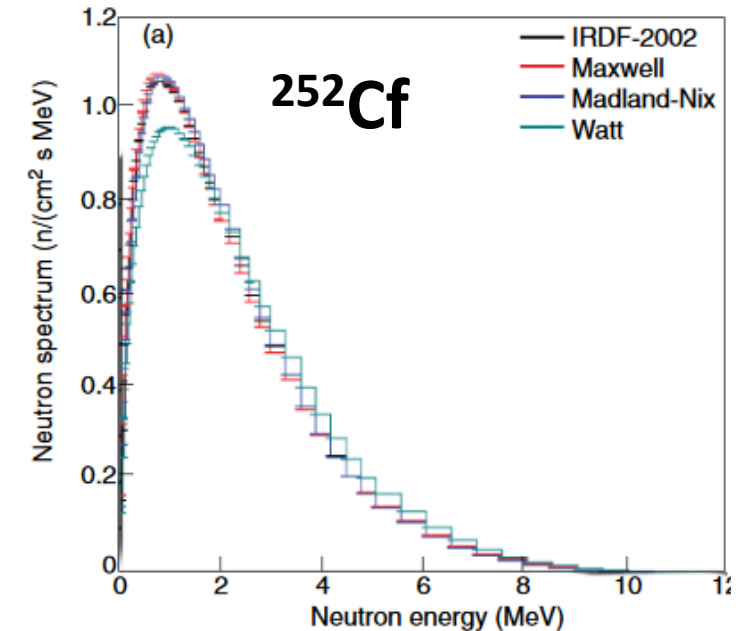$$P(KE_n) = KE_n \exp(-E_n/k_B T)$$

- Transforming into the lab frame, the result is the Watt fission spectrum,

$$P(KE_n) = \exp(-KE_n/k_B T)\sinh\left(\sqrt{4E_n E_f/(k_B T)^2}\right), \text{where } E_n$$

and E_f are the laboratory neutron and fission fragment energies in

MeV/u and $T = \sqrt{\dfrac{8E^*}{A}}$ is the nuclear temperature

- Parameterized forms of the Watt fission spectrum, with the associated parameter values for a given fission fragment are available. E.g. in the appendix of the MCNP user manual

(a) ^{252}Cf
IRDF-2002 — Maxwell — Madland-Nix — Watt
Neutron spectrum (n/(cm² s MeV))
Neutron energy (MeV)

(c)
Difference in Neutron spectrum (%)
Maxwell/IRDF2002
LANL–Madland/IRDF2002
Watt/IRDF2002
Neutron energy (MeV)

Fission fragment angular distributions

- Induced fission is slow enough that the nucleus reaches statistical equilibrium

- This results in an angular distribution of fission fragments that is symmetric about 90° in the center of mass, where 90° marks the plane perpendicular to the direction of motion of the fissioning system.

- This is because forward and backward angles maximize the amount of angular momentum the fragments carry away. Prior to scission, the fissioning nucleus will be highly excited relative to the absolute ground state, and higher excitations correspond on average to larger angular momenta.

- The proper formula for the angular distribution is complicated and related to the deformation of the fissioning nucleus
  [See Loveland, Morrissey, & Seaborg]

- From momentum conservation, we know that the fragments themselves are emitted 180° apart (in the CM), which is handy for gating on fission events in reaction-induced fission studies

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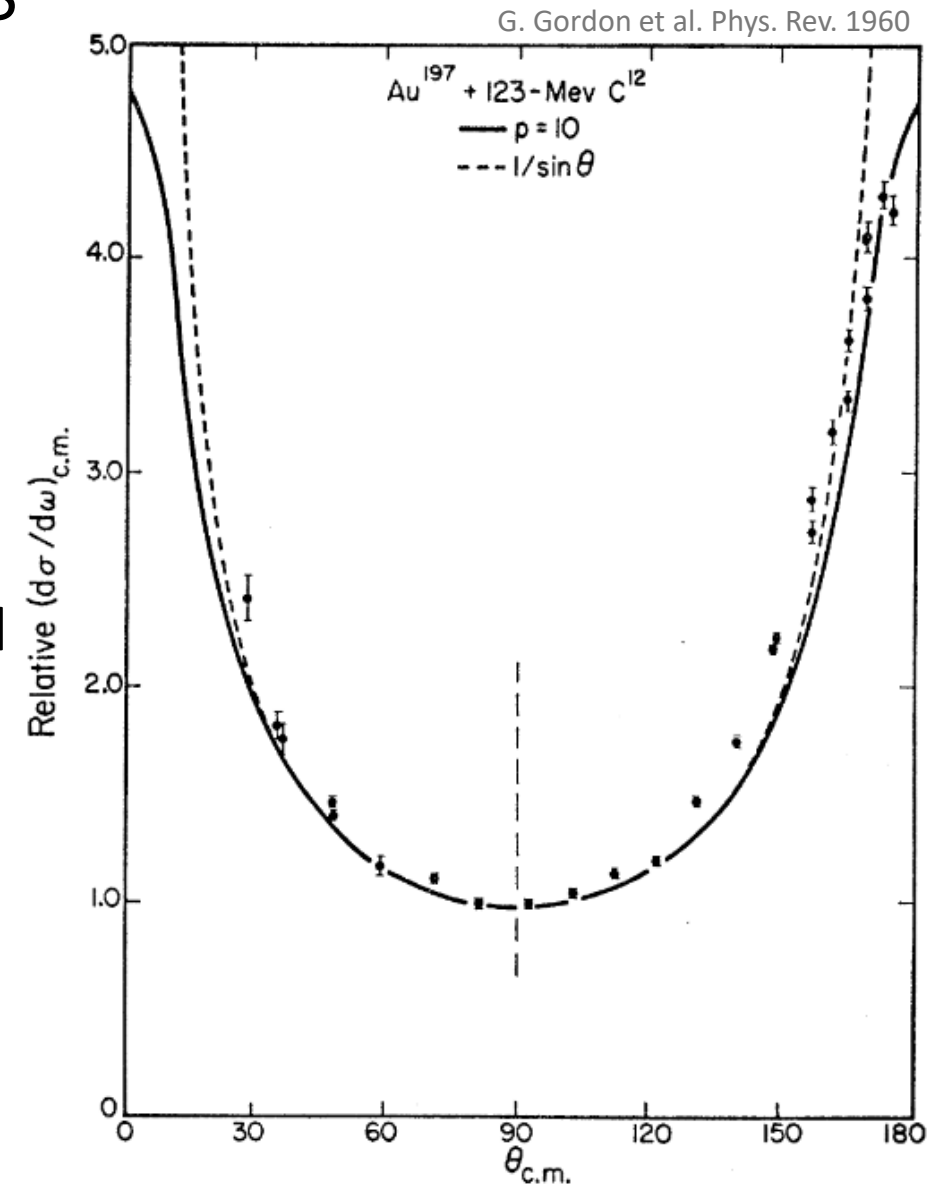
Further Reading

- Chapter 11: Modern Nuclear Chemistry (Loveland, Morrissey, Seaborg)
- Chapters 2 & 9 : Nuclear & Particle Physics (B.R. Martin)