Chapter VIII: Nuclear fission

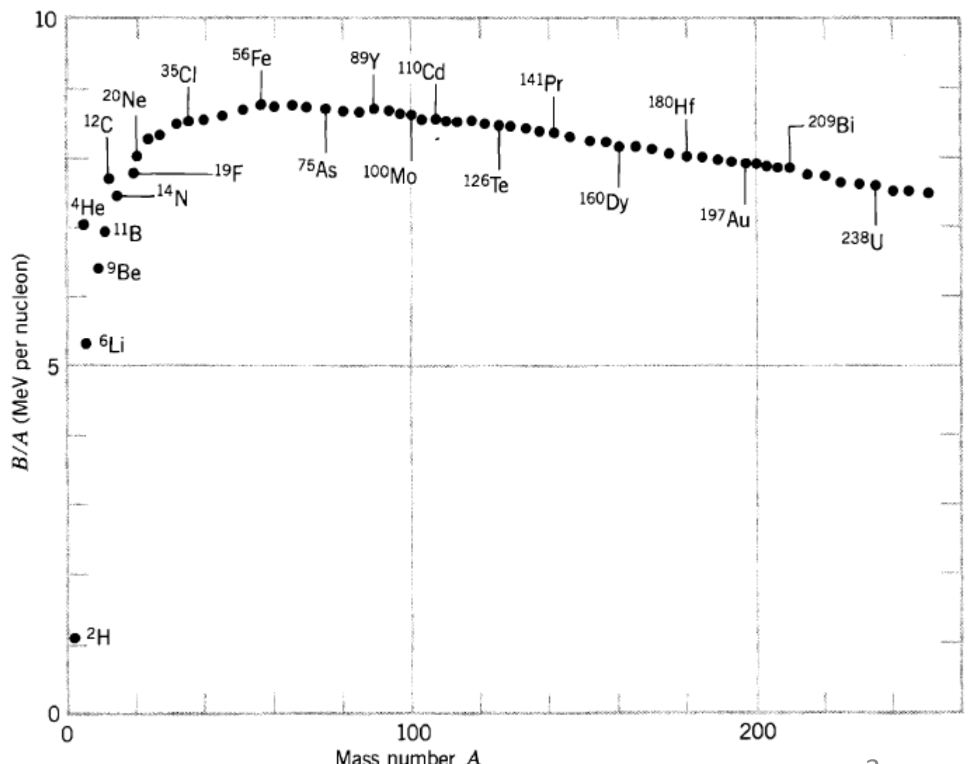
Summary

1. General remarks
2. Spontaneous and induced fissions
3. Nucleus deformation
4. Mass distribution of fragments
5. Number of emitted electrons
6. Radioactive decay processes
7. Fission cross section
8. Energy in fission
9. Nuclear structure
10. Applications

General remarks (1)

- Fission results from competition between nuclear and Coulomb forces in heavy nuclei → total nuclear binding energy increases roughly like A ↔ Coulomb repulsion energy of protons increase like Z^2 → faster

- Example of ^{238}U → binding energy B ≈ 7.6 MeV/nucleon → if division into 2 equal Pd fragments with A $\simeq$ 119 → B by nucleon ≈ 8.5 MeV → more tightly bound system → energy is released → (-238 $\times$ 7.6) - (-2 $\times$ 119 $\times$ 8.5) = 214 MeV

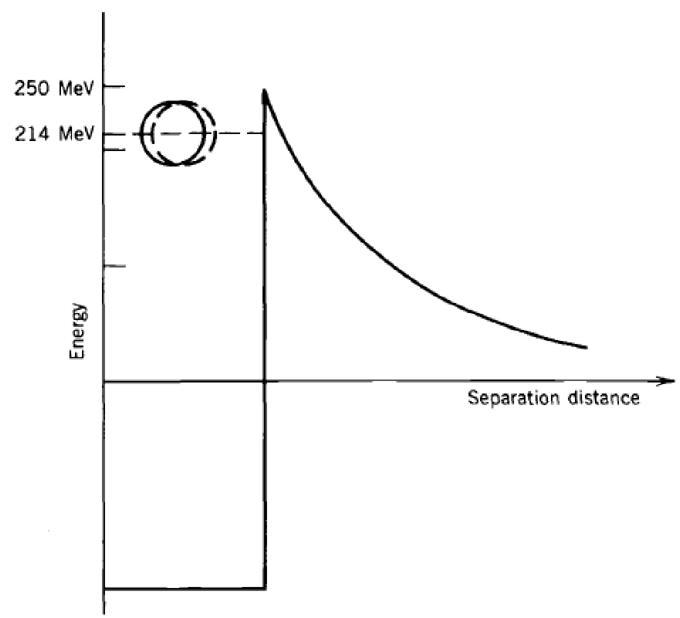


General remarks (2)

- To conserve energy → the final state must include an extra energy → variety of forms → neutrons, β and γ emissions from the fragments and primarily ($\sim$ 80%) as kinetic energy of the fragments as Coulomb repulsion drives them apart
- Generally fragments are not identical → binary fission if 2 fragments ↔ ternary fission if 3 fragments (rare and generally 1 of the 3 fragments is an α)
- Attention → not so obvious → for ^{238}U competition with spontaneous α decay ($T_{1/2}$ = 4.5 $\times$ 10^9 y) while $T_{1/2}$ for fission is ≈ 10^{16} y → not important decay mode for ^{238}U → become important for A ≥ 250

Spontaneous and induced fissions (1)

- Inhibition of the fission by the Coulomb barrier (analogous to Coulomb barrier of α decay) → improbable in general for a nucleus in its ground state
- In previous example of ^{238}U → ^{238}U may perhaps exist instantaneously as two fragments of ^{119}Pd but Coulomb barrier of about 250 MeV for ^{238}U prevents the fission

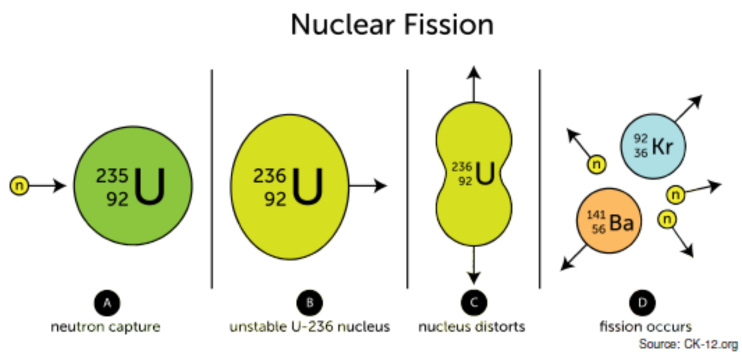


Spontaneous and induced fissions (2)

- If the height of the Coulomb barrier is roughly equal to the energy released in fission → reasonably good chance to penetrate the barrier
- This process is called spontaneous fission → in that case fission competes successfully with other decay processes
- Lightest nucleus for which spontaneous fission is the dominant decay mode → isotope ${}^{250}_{96}\mathrm{Cm}$ of the curium (80% of the disintegrations are fission and $T_{1/2}$ = 10^4 y)
- For ${}^{254}_{98}\mathrm{Cl}$ of californium ($T_{1/2}$ = 60 days) → lightest nucleus for which almost 100% of decay is spontaneous fission
- These 2 nuclei does not exist in natural state

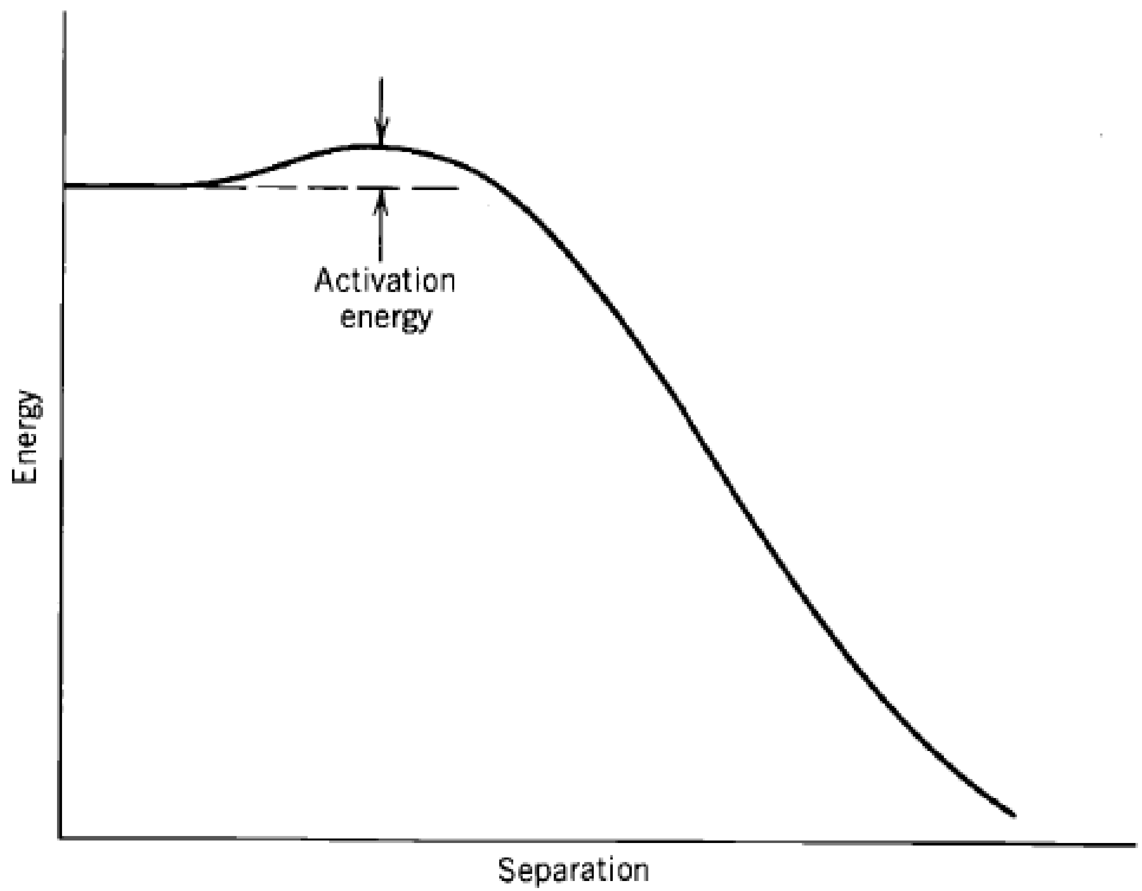
Spontaneous and induced fissions (3)

- Fission is much more frequent if the nucleus is in an excited state → occurs if a heavy nucleus absorb energy from a neutron or a photon → formation of an intermediate state in an excited state that is at or above the barrier → phenomenon called induced fission



- The ability of a nucleus to undergo induced fission depends critically on the energy of the intermediate system → for some absorption of thermal neutrons (≈ 0.025 eV) is sufficient ↔ for others fast (MeV) neutrons are required
- The height of the fission barrier above the ground state is called the activation energy E_{act}

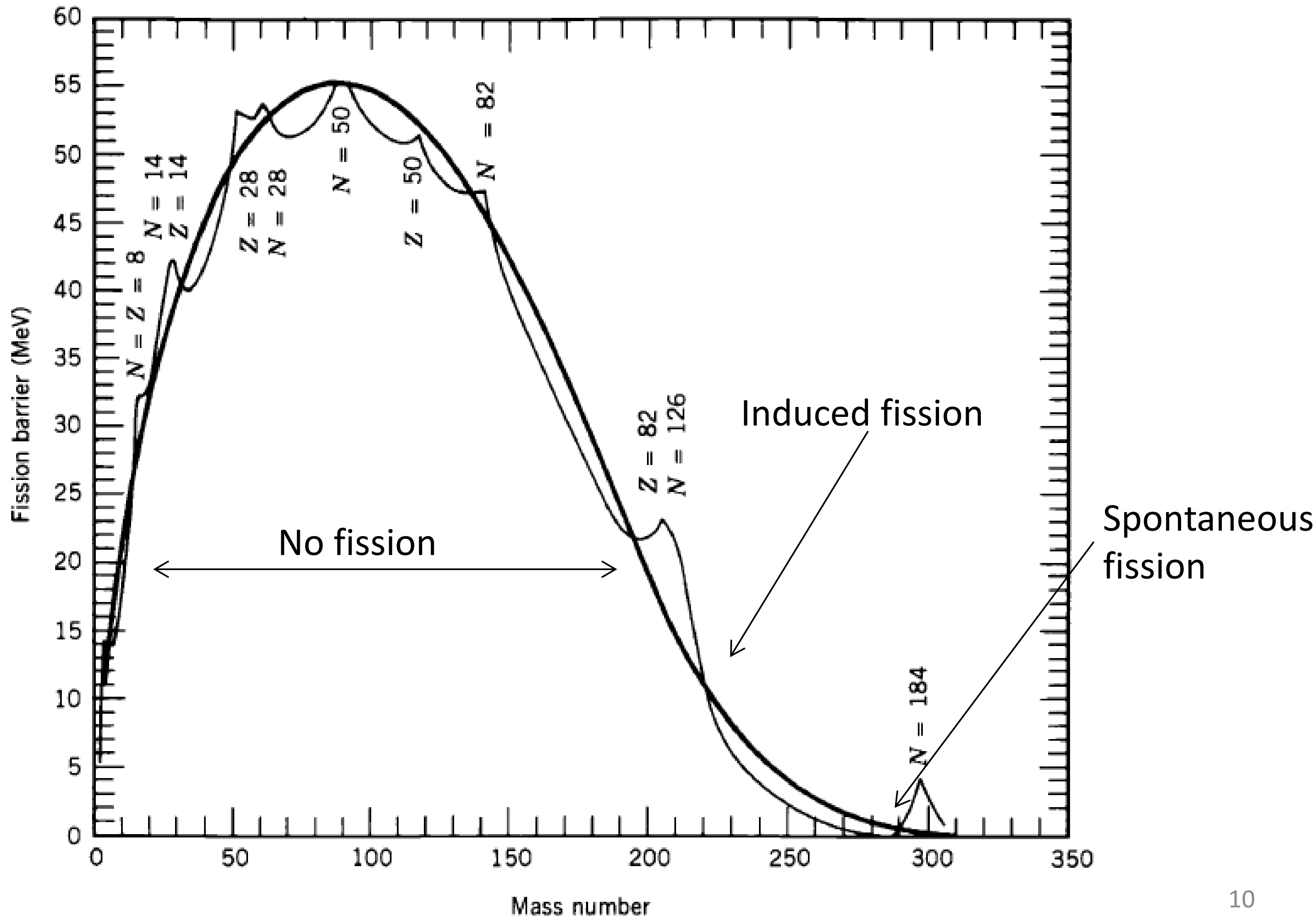
Spontaneous and induced fissions: activation energy (1)



Spontaneous and induced fissions: activation energy (2)

- Calculation of the barrier height is based on the liquid-drop model → the use of the shell model including more sophisticated effects modifies a bit the calculation (especially for magic numbers)
- Liquid-drop model implies the vanishing energy around mass 280 → these nuclei are thus extremely unstable to spontaneous fission
- Shell closure suggests that super-heavy nuclei around A = 300 are more stable against fission → research about super-heavy nucleons around the magic number N = 184 for neutrons
- Note the typical 5-MeV energies around uranium

Spontaneous and induced fissions: activation energy (3)



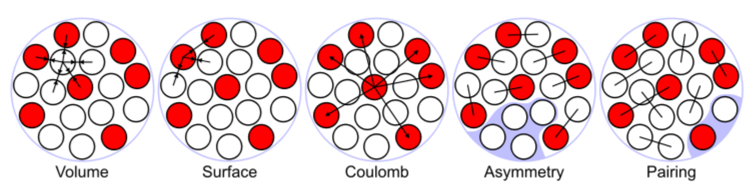
Nucleus deformation (1)

- To qualitatively understand fission → effect of the deformation on a heavy nucleus on semi-empirical Bethe-Weizsäcker equation →

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

- Effect on the binding energy of an initially spherical nucleus ($V = (4/3)\pi R^3$) that we gradually stretch → V is kept constant (cannot change because of the short-range of nuclear interaction) → stretched nucleus is an ellipsoid of revolution ($V = (4/3)\pi ab^2$) whit a the semimajor axis and b the semiminor axis → deviation of the ellipsoid from a sphere of is given in terms of the distortion parameter ϵ (eccentricity of the ellipse) →

$$\begin{aligned} a &= R(1+\epsilon) \\ b &= R(1+\epsilon)^{-1/2} \end{aligned}$$



Nucleus deformation (2)

- Distortion of a sphere to an ellipse → increase of area S →

$$S = 4\pi R^2(1 + \frac{2}{5}\epsilon^2 + \dots)$$

- Consequently → the absolute value of the surface energy term in the Bethe-Weizsäcker formula increases → decrease of the binding energy by ΔB_S →

$$\Delta B_S \simeq -a_S A^{2/3} \frac{2}{5}\epsilon^2$$

- Distortion of a sphere to an ellipse → decrease of the Coulomb term by a factor (1 - (1/5)ϵ^2 +…) → increase of the of the binding energy by ΔB_C →

$$\Delta B_C \simeq a_C \frac{Z^2}{A^{1/3}} \frac{1}{5}\epsilon^2$$

Nucleus deformation (3)

- The total variation of the binding energy is given by →

$$\Delta B \simeq A^{2/3}\frac{2}{5}(-a_s + \frac{a_C}{2}\frac{Z^2}{A})\epsilon^2$$

- If the second term is larger than the first → the energy difference ΔB is > 0 → gain energy due to the stretching → more the nucleus is stretched more energy is gained → amplification of the stretching → nucleus unstable → fission

- The condition for spontaneous fission is thus →

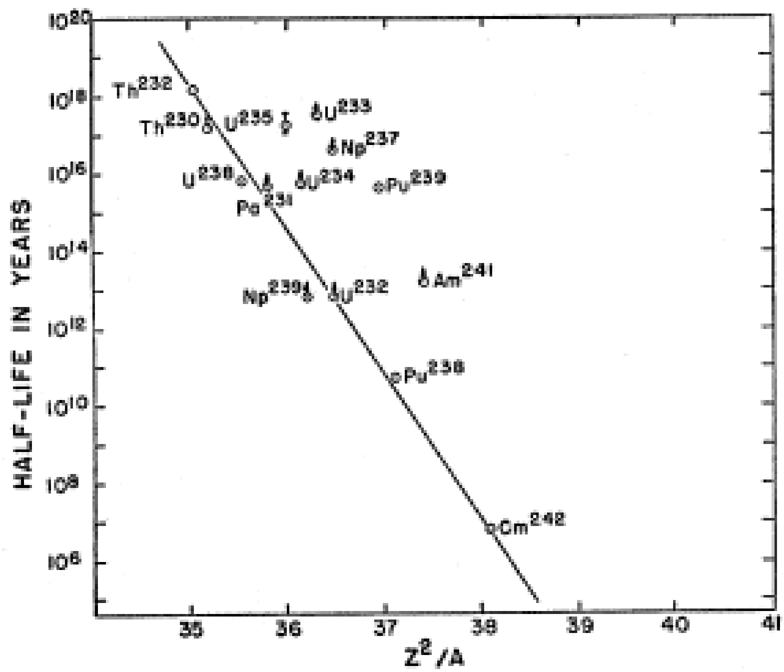
$$\Delta B > 0 \rightarrow \frac{a_C}{2}\frac{Z^2}{A} > a_S \rightarrow \frac{Z^2}{A} > 47$$

Nucleus deformation (4)

- For heavy nuclei → $Z/A \approx 0.4$ → nuclei become instable for $Z > 117$
- In practice → modifications of this expression
  - Quantum mechanical barrier penetration could be possible even for negative energy deformation
  - Heavy nuclei have permanent deformation → the equilibrium shape is ellipsoidal
- However Z^2/A gives an indicator of the ability of a nucleus to fission spontaneously → the larger the value o Z^2/A the shorter is the half-life for spontaneous fission
- An approached expression for the half-life for spontaneous fission is:

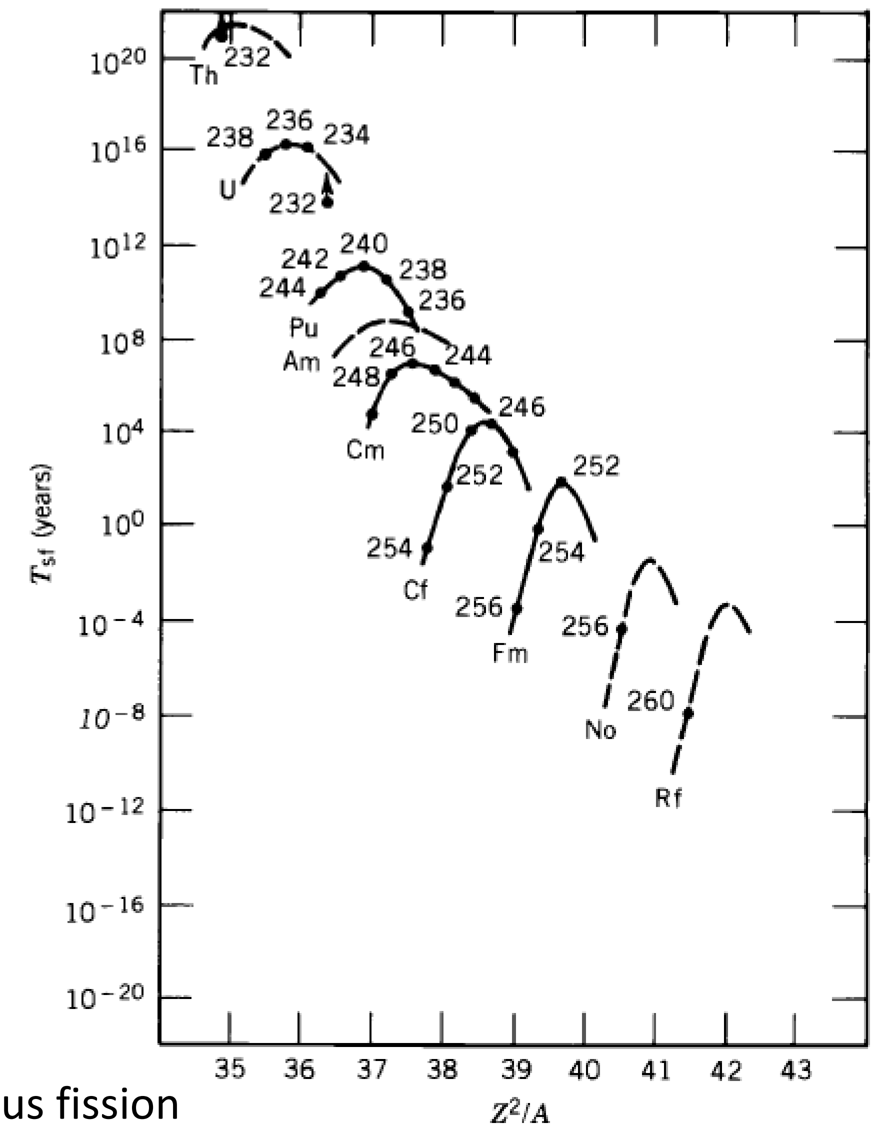
$$T_{1/2}^{SF} = 10^{-21} \times 10^{178-3.75Z^2/A}\ \mathrm{s}$$

Nucleus deformation (5)



- Attention → the real $T_{1/2}$ can be very different of the $T_{1/2}^{SF}$ due to other decay possibilities
- Extrapolation for 45 < Z^2/A < 50 → $T_{1/2}^{SF} = 10^{-20}$ s i.e. an instantaneous fission → it corresponds to A ≈ 280 as obtained previously
- For Z = constant → $T_{1/2}^{SF}$ are not constant → parabolic shape → more elaborated models are needed

Nucleus deformation (6)



Lifetimes for spontaneous fission

Mass distribution of fragments (1)

- Typical neutron-induced fission reaction is

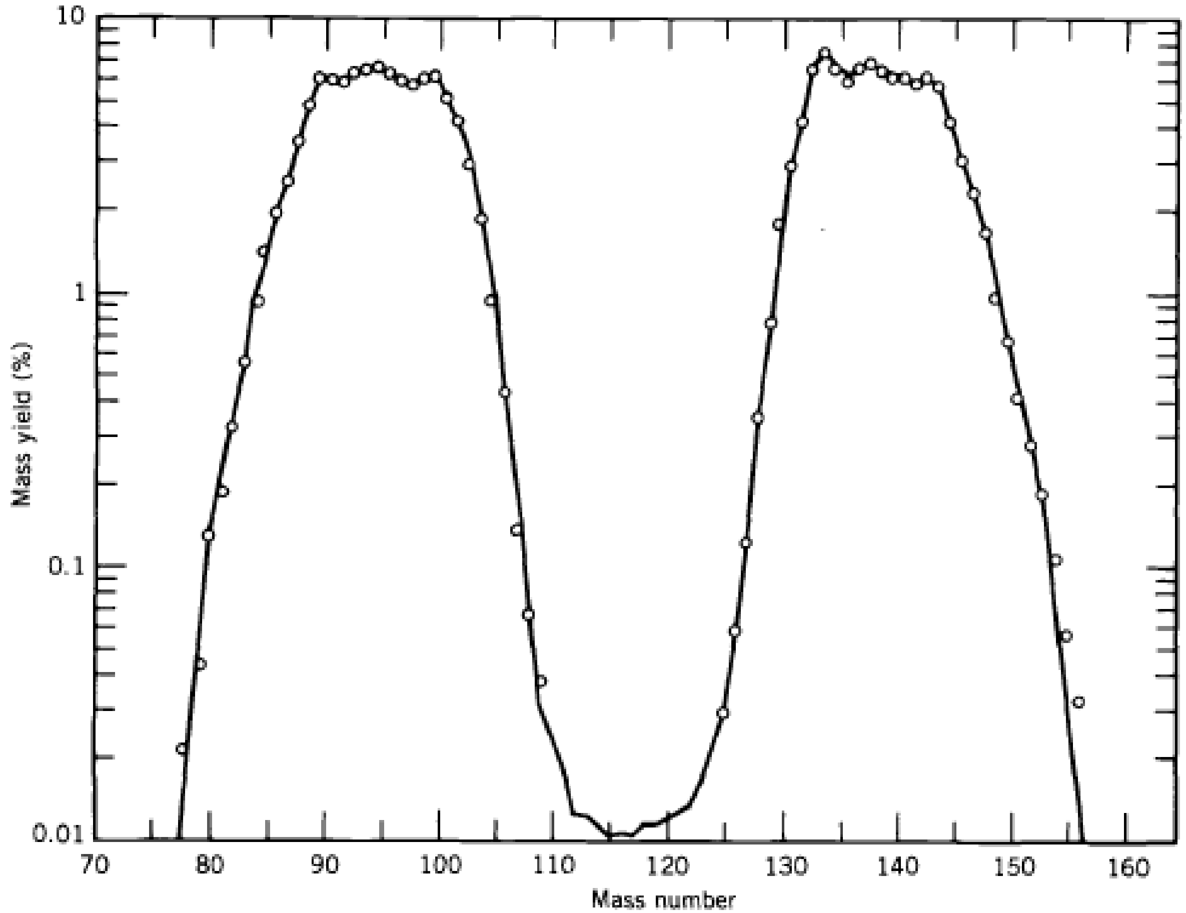
$$^{A}_{Z}X_{N} + \mathrm{n} \rightarrow {}^{A+1}_{Z}X^{*}_{N+1} \rightarrow {}^{A_1}_{Z_1}X_{N_1} + {}^{A_2}_{Z_2}X_{N_2} + \nu\mathrm{n}$$

- As for instance →

$$^{235}_{92}\mathrm{U}_{143} + \mathrm{n} \rightarrow {}^{93}_{37}\mathrm{Rb}_{56} + {}^{141}_{55}\mathrm{Cs}_{86} + 2\mathrm{n}$$

- This last reaction is particularly probable for low energy neutrons (thermal energies) but other reactions are only possible for large neutron energies (i.e. ^{238}U)
- Fission products are not determined uniquely → distribution of masses of the 2 fission products (ternary decay is rare) with condition $Z_1 + Z_2 = Z$ and $N_1 + N_2 + \nu = N + 1$
- For ^{235}U → distribution is symmetric about the center ($A \approx 116$) → an heavy fragment ($A_1 \approx 140$ → I, Xe, Ba) and a light fragment ($A_2 \approx 95$ → Br, Kr, Sr, Zr) → fission with $A_1 \approx A_2$ is less probable by a factor 600

Mass distribution of fragments (2)

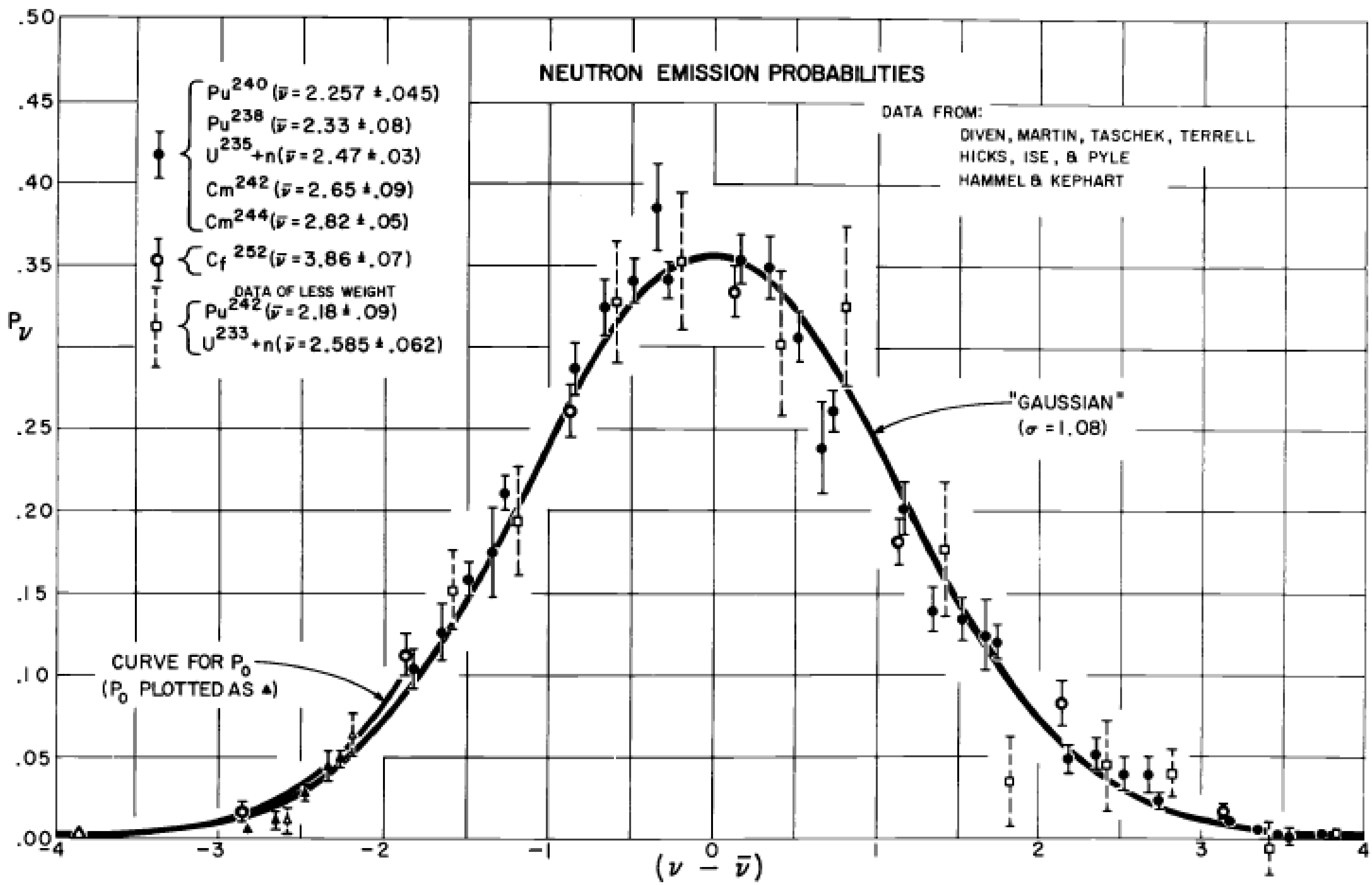


Mass distribution of fission fragments for $^{235}U + n$

Number of emitted neutrons: prompt neutrons (1)

- The ν neutrons of previous equation are emitted in a time < 10^{-16} s (time analog to the fragmentation duration) → they are called prompt neutrons
- To understand their origin → we consider again the case of ^{235}U → the fragments in the vicinity of *A* = 95 and *A* = 140 must share 92 protons → if it happens in rough proportion to their masses the nuclei formed are $^{95}_{37}\mathrm{Rb}_{58}$ and $^{140}_{55}\mathrm{Cs}_{85}$ → nuclei rich in neutrons
- These fission products have *Z/A* = 0.39 (i.e. same Z/A ratio as the initial nucleus ^{235}U)
- The stable *A* = 95 isobar has *Z* = 42 and the stable *A* = 140 isobar has *Z* = 58 → neutron excess emits at the instant of fission
- The average number of prompt neutrons depends of the nature of the 2 fragments and of the energy of incident particle for induced fission

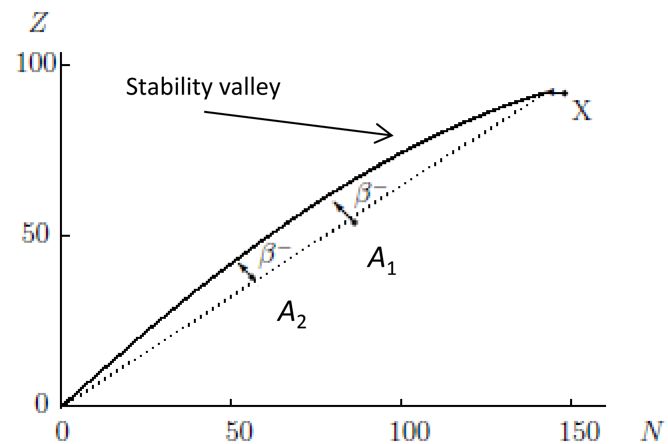
Number of emitted neutrons: prompt neutrons (2)



- Distribution of fission neutrons → the average number of neutrons changes with the fissioning nucleus but the distribution about the average is independent of the original nucleus

Number of emitted neutrons: delayed neutrons (1)

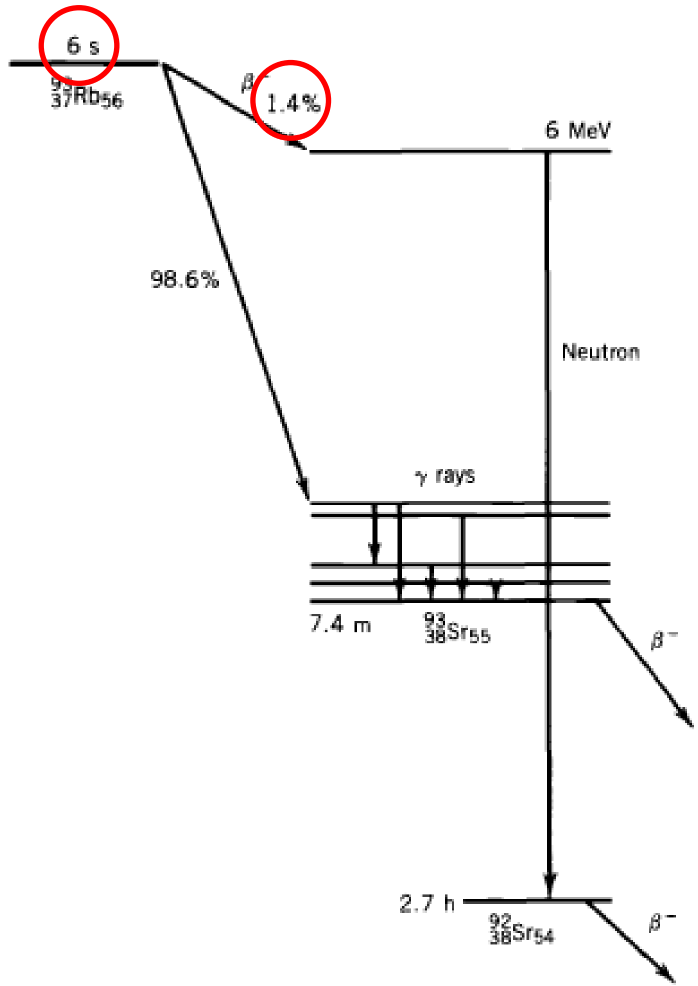
- Nuclei A_1 and A_2 are generally far from the stability valley → β^- decay



- Following this β^- decay → β-delayed nucleon emission (as explained in chapter V) → these neutrons are called delayed neutrons
- Nucleon emission occurs rapidly → nucleon emission occurs with a half-life characteristic of β^- decay → usually of the order of seconds

Number of emitted neutrons: delayed neutrons (2)

- Practical example: $^{93}Rb \rightarrow$



Number of emitted neutrons: delayed neutrons (3)

- The total intensity of delayed neutrons is ≈ 1 per 100 fissions
- Delayed neutrons are essential for the control of nuclear reactors
- No mechanical system could respond rapidly enough to prevent important variations in the prompt neutrons
- On the contrary → possible to achieve control using the delayed neutrons

Radioactive decay processes

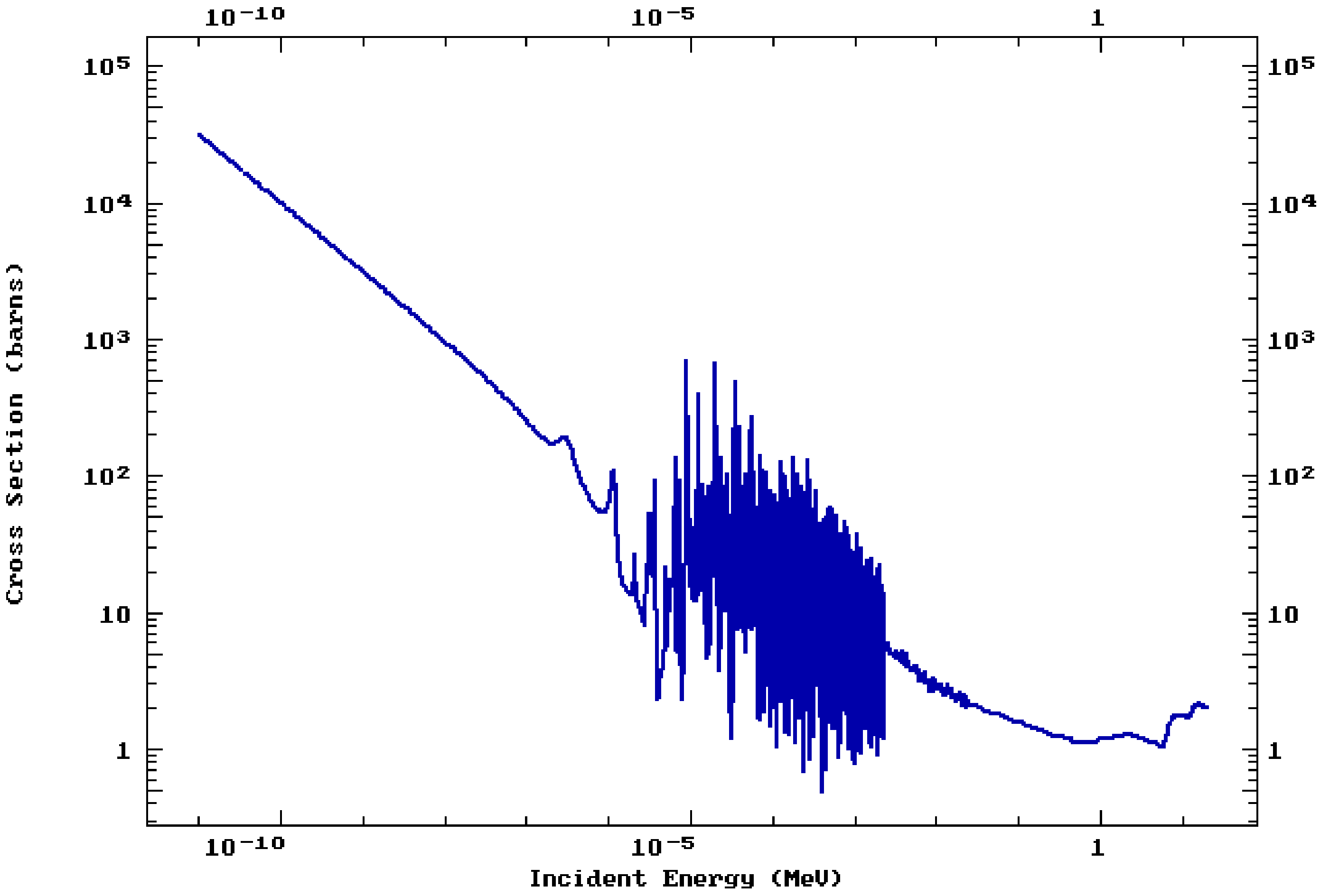
- Initial fission products are highly radioactive → they decay toward stable isobars by emitting many β and γ → these radiations contribute to the total energy release during fission
- Examples of decay chains →

$$^{93}\mathrm{Rb} \xrightarrow{6\ \mathrm{s}} {}^{93}\mathrm{Sr} \xrightarrow{7\ \mathrm{m}} {}^{93}\mathrm{Y} \xrightarrow{10\ \mathrm{h}} {}^{93}\mathrm{Zr} \xrightarrow{10^{6}\ \mathrm{y}} {}^{93}\mathrm{Nb}$$

$$^{141}\mathrm{Cs} \xrightarrow{25\ \mathrm{s}} {}^{141}\mathrm{Ba} \xrightarrow{18\ \mathrm{m}} {}^{141}\mathrm{La} \xrightarrow{4\ \mathrm{h}} {}^{141}\mathrm{Ce} \xrightarrow{33\ \mathrm{d}} {}^{141}\mathrm{Pr}$$

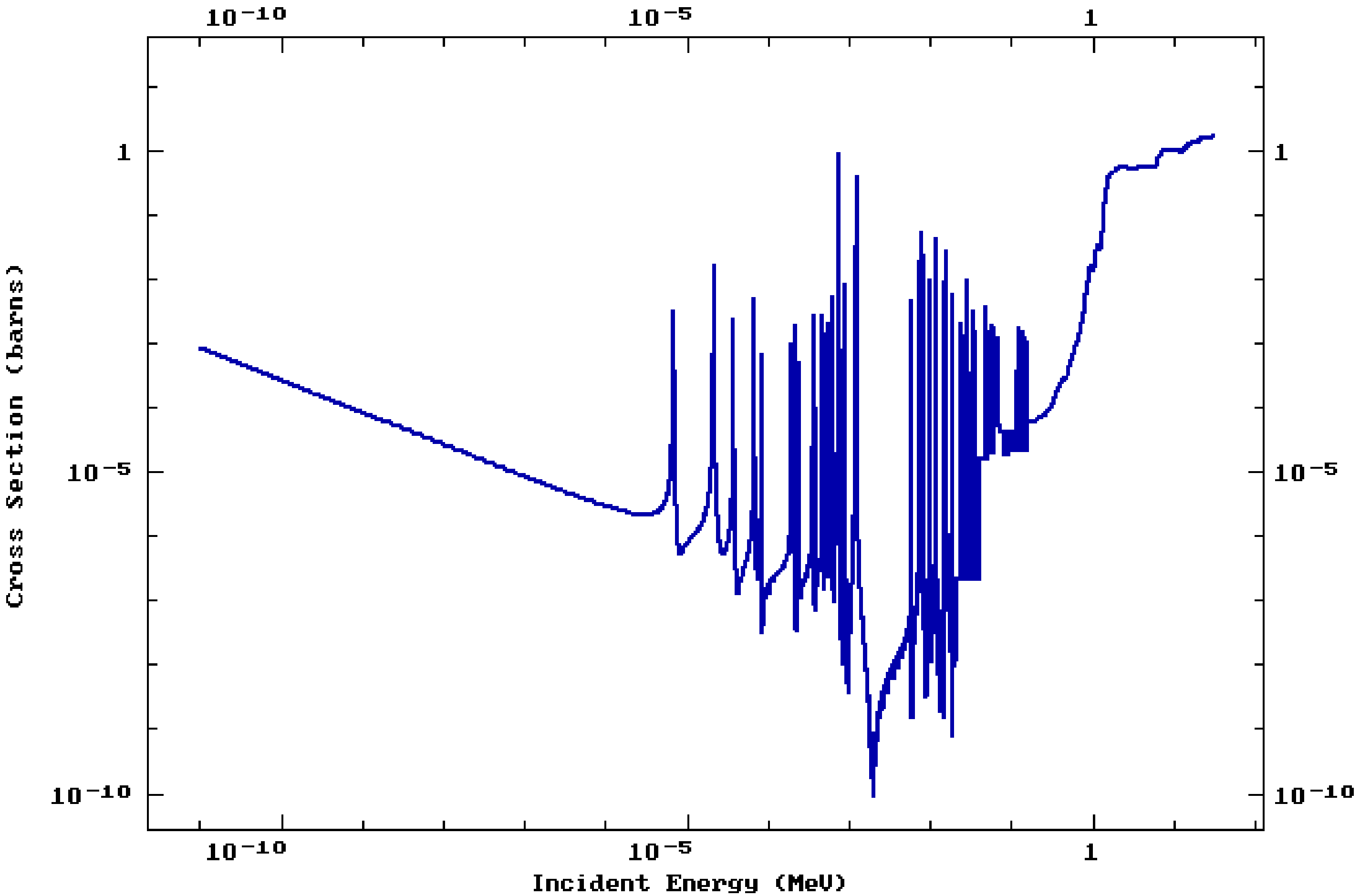
- These radioactive products are the waste products of nuclear reactors
- Many decay very quickly ↔ others have long half-lives (especially near the stable members of the series)

Fission cross section: ^{235}U



http://www.nndc.bnl.gov/exfor/endf00.jsp

Fission cross section: ^{238}U



Fission cross section: Simple model

- Simple estimation of energy dependence is provided by the Ramsauer model
- The effective size of a neutron is $\propto$ to its de Broglie wavelength →

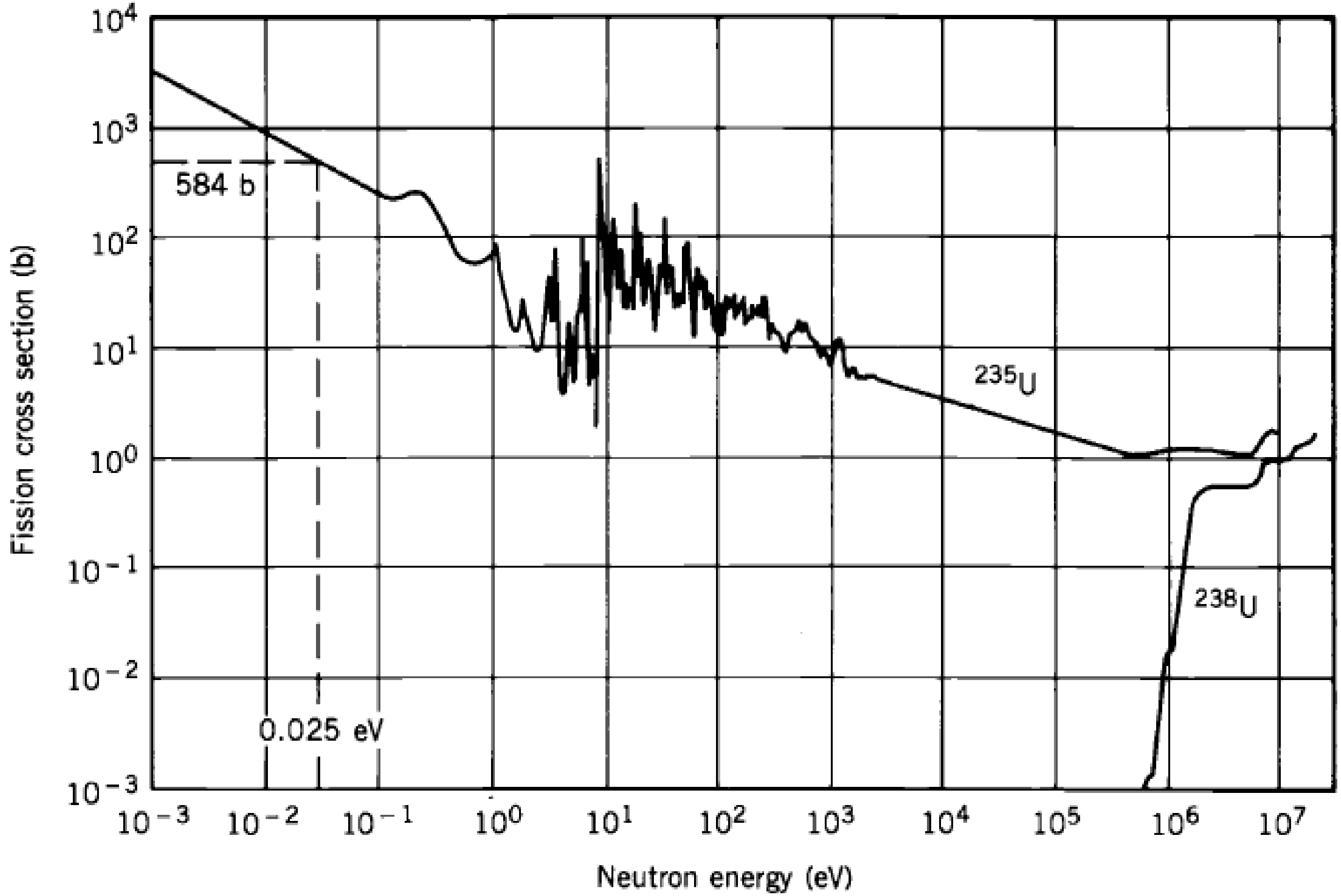
$$\lambda(E) = \frac{1}{k} = \frac{h}{p} = \frac{h}{\sqrt{2mE}}$$

- R is the effective radius of the nucleus → the cross section of interaction $\sigma(E) \propto \pi[R + \lambda(E)]^2 \times T$ (T is the transmission probability of crossing the barrier potential, written $4kK/(k + K)^2$ with $k = (2mE/\hbar^2)^{1/2}$ and $K = (2m(E + V_0)/\hbar^2)^{1/2}$ for a barrier of depth $-V_0$)
- For low energy neutron = large wavelength → R can be neglected, $E \ll V_0$ and $k \ll K$ → $\sigma \propto \lambda$ → $\sigma(E)$ is inversely proportional to neutron velocity
- For high energy neutron = small wavelength → $\sigma(E) \propto R^2$ → constant
- Attention → presence of resonances → precisely defined states of the composed nucleus

Fission cross section: For thermal neutrons

Nuclide	Cross Section (b)	$A + 1$ Activation Energy (MeV)
$^{227}_{90}Th_{137}$	200 ± 20	
$^{228}_{90}Th_{138}$	< 0.3	
$^{229}_{90}Th_{139}$	30 ± 3	8.3
$^{230}_{90}Th_{140}$	< 0.001	8.3
$^{230}_{91}Pa_{139}$	1500 ± 300	7.6
$^{231}_{91}Pa_{140}$	0.019 ± 0.003	7.6
$^{232}_{91}Pa_{141}$	700 ± 100	7.2
$^{233}_{91}Pa_{142}$	< 0.1	7.1
$^{231}_{92}U_{139}$	300 ± 300	6.8
$^{232}_{92}U_{140}$	76 ± 4	6.9
$^{233}_{92}U_{141}$	530 ± 5	6.5
$^{234}_{92}U_{142}$	< 0.005	6.5
$^{235}_{92}U_{143}$	584 ± 1	6.2
$^{238}_{92}U_{146}$	$(2.7 \pm 0.3) \times 10^{-6}$	6.6
$^{234}_{93}Np_{141}$	1000 ± 400	5.9
$^{236}_{93}Np_{143}$	3000 ± 600	5.9
$^{237}_{93}Np_{144}$	0.020 ± 0.005	6.2
$^{238}_{93}Np_{145}$	17 ± 1	6.0
$^{239}_{93}Np_{146}$	< 0.001	6.3
$^{238}_{94}Pu_{144}$	17 ± 1	6.2
$^{239}_{94}Pu_{145}$	742 ± 3	6.0
$^{240}_{94}Pu_{146}$	< 0.08	6.3
$^{241}_{94}Pu_{147}$	1010 ± 10	6.0
$^{242}_{94}Pu_{148}$	< 0.2	6.2
$^{241}_{95}Am_{146}$	3.24 ± 0.15	6.5
$^{242}_{95}Am_{147}$	2100 ± 200	6.2
$^{243}_{95}Am_{148}$	< 0.08	6.3
$^{244}_{95}Am_{149}$	2200 ± 300	6.0
$^{243}_{96}Cm_{147}$	$610 + 30$	6.1
$^{244}_{96}Cm_{148}$	1.0 ± 0.5	6.3
$^{245}_{96}Cm_{149}$	2000 ± 200	5.9
$^{246}_{96}Cm_{150}$	0.2 ± 0.1	6.0

Fission cross section: ^{235}U and ^{238}U



Energy in fission: Excitation energy (1)

- We consider ^{235}U capturing a neutron → compound state ^{236}U*
- The excitation energy E_{ex} is

$$E_{ex} = [m(^{236}\mathrm{U}^*) - m(^{236}\mathrm{U})]c^2$$

- Energy of ^{236}U* is given by (assuming a negligible kinetic energy for the incident neutron ↔ thermal neutron) →

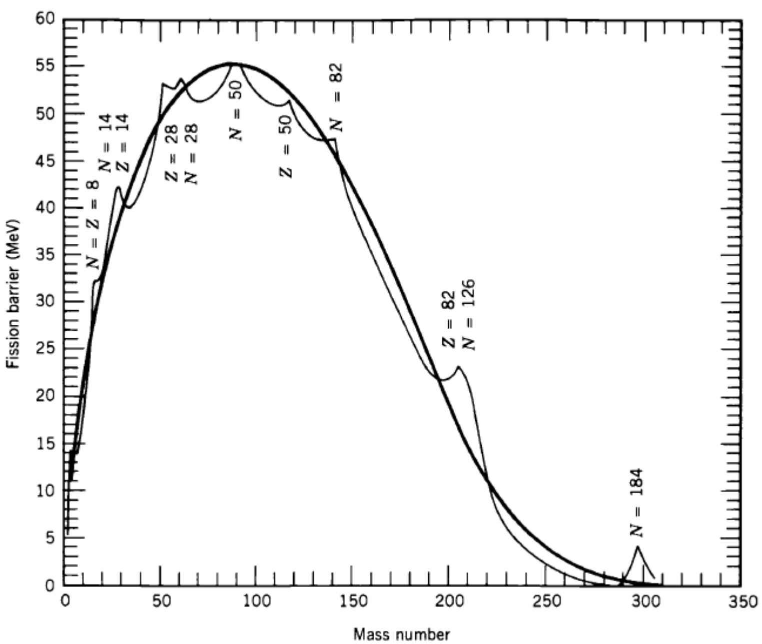
$$\begin{aligned} m(^{236}\mathrm{U}^*) &= m(^{235}\mathrm{U}) + m_n = 235.043924 \text{ u} + 1.008665 \text{ u} \\ &= 236.052589 \text{ u} \end{aligned}$$

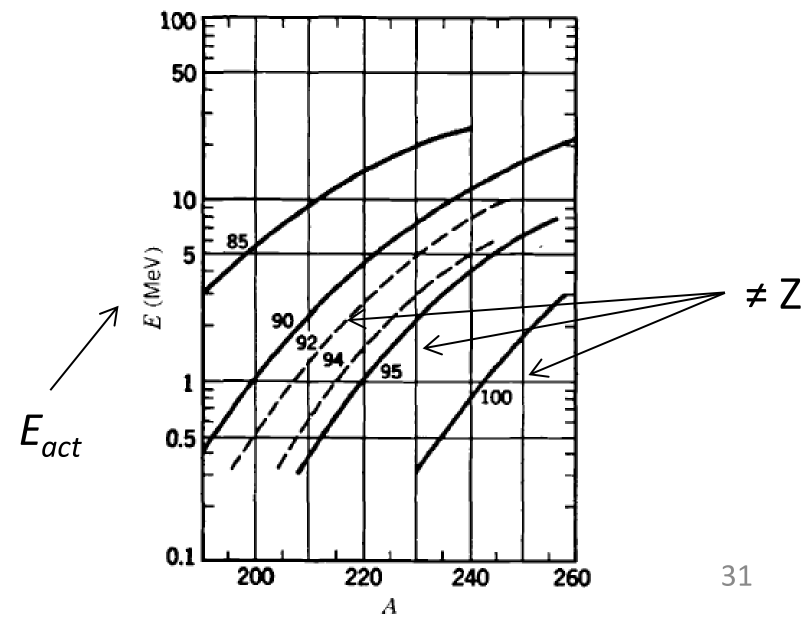
$$\Rightarrow$$

$$\begin{aligned} E_{ex} &= (236.052589 \text{ u} - 236.045563 \text{ u})931.502 \text{ MeV/u} \\ &= 6.5 \text{ MeV} \end{aligned}$$

Energy in fission: Excitation energy (2)

- The activation energy obtained for ^{236}U is 6.2 MeV
- We have $E_{ex} > E_{act}$
- ^{235}U can be fissioned with ≈ 0-energy neutrons
- For $^{238}U + n \rightarrow {}^{239}U^* \rightarrow E_{ex} = 4.8$ MeV and $E_{act} = 6.6$ MeV → neutrons of a few MeV are required for fission → threshold in energy





Energy in fission: Excitation energy (3)

- The extreme differences in the fissionability of ^{235}U and ^{238}U is due to the difference between their excitation energies: 6.5 and 4.8 MeV
- This ≠ in E_{ex} is explained by the pairing energy term $\delta = \pm 12 A^{-1/2}$ in the Bethe-Weizsäcker formula ↔ only significant ≠ between A and A+1

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

- The role of δ in the excitation energy →
  - ^{235}U (N-odd – Z-even → δ = 0) + n → ^{236}U (N-even – Z-even → $\delta \approx +12/(235)^{1/2}$ = 0.78 MeV) → gain of 1 $\delta \approx$ +0.78 MeV
  - ^{238}U (N-even – Z-even → δ = 0. 78 MeV) + n → ^{239}U (N-odd – Z-even → $\delta \approx$ 0 MeV) → decrease of 1 $\delta \approx$ -0. 78 MeV
  - The difference in excitation energy between ^{235}U + n and ^{238}U + n is therefore about $2\delta \approx$ +1.6 MeV → corresponds to observed difference

Energy in fission: Excitation energy (4)

^{A}X			^{A+1}X			ΔE_{X^*}
N	Z	$\delta(^{A}X)$	N	Z	$\delta(^{A+1}X)$	
even	even	$+\delta$	odd	even	0	$-\delta$
even	odd	0	odd	odd	$-\delta$	$-\delta$
odd	even	0	even	even	$+\delta$	$+\delta$
odd	odd	$-\delta$	even	odd	0	$+\delta$

- In a general way → if we call ΔE_{X*} the contribution to the excitation energy due to the pairing energy term δ → if we consider the 4 possible case types → we obtain (for initial *N*) →

$$\Delta E_{X^*} \approx (-1)^{N+1}\delta$$

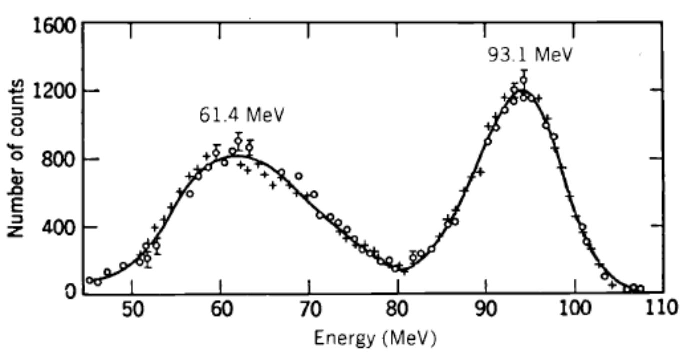
- The difference between nuclei with even neutrons and odd neutrons is 2δ → ≈ 1.6 MeV for A ≈ 240

Energy in fission: Released energy (1)

- We consider again the reaction →

$$^{235}_{92}\mathrm{U}_{143} + \mathrm{n} \rightarrow {}^{93}_{37}\mathrm{Rb}_{56} + {}^{141}_{55}\mathrm{Cs}_{86} + 2\mathrm{n}$$

- Using the binding energy/nucleon (see for instance http://amdc.in2p3.fr/masstables/Ame2012/Ame2012b-v2.pdf) → Q ≈ 180 MeV → other final products gives energy releases of roughly the same magnitude → quite reasonable to take 200 MeV as an average value for the energy released for ^{235}U fission
- Experiments allows obtaining the energy distribution of the two fission fragments → the 2 higher energies are at 66 MeV for heavy fragment and 98 MeV for light fragment



Add 5 MeV due to miscalibration

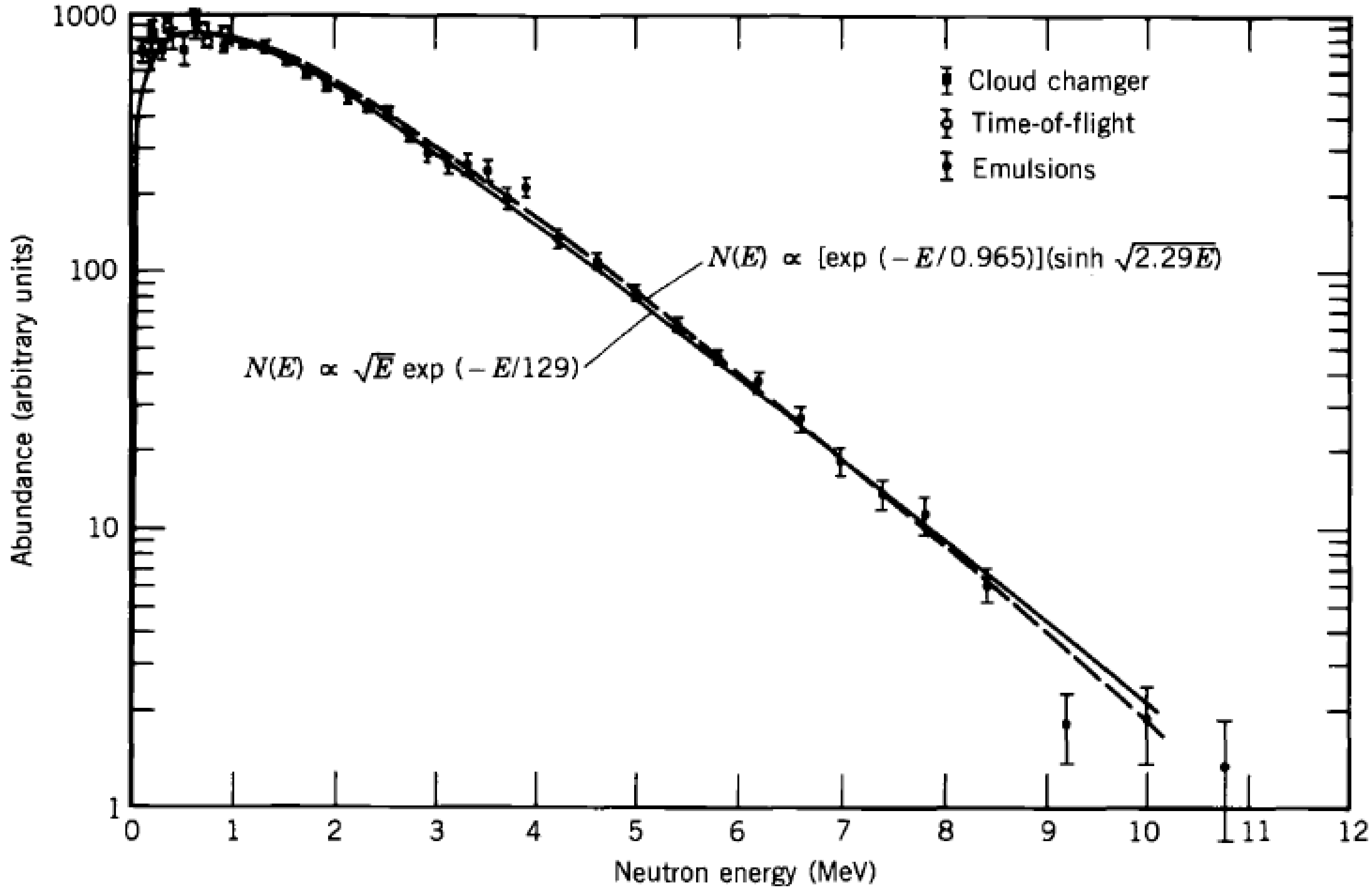
Energy in fission: Released energy (2)

- Conservation of momenta gives (neutrons carry very little momentum) → $m_1 v_1 = m_2 v_2$ → ratio between kinetic energies is the inverse of the ratio of the masses →

$$\frac{\frac{1}{2} m_1 v_1^2}{\frac{1}{2} m_2 v_2^2} = \frac{m_2}{m_1}$$

- The ratio of the energies 66 MeV / 98 MeV = 0.67 is consistent with the ratio of the masses 95 / 140 = 0.68
- The total energy carried by the 2 fragments = 164 MeV ≈ 80% of the total fission energy
- The average energy carried by 1 prompt neutron is about 2 MeV → with 2.5 neutrons per fission → the total average energy carried by the neutrons in fission is ≈ 5 MeV (3% of the fragments energy → can be neglected in the equation of momentum conservation)

Energy in fission: Released energy (3)



Energy spectrum of prompt neutrons emitted during fission of ^{235}U → mean value ≈ 2 MeV

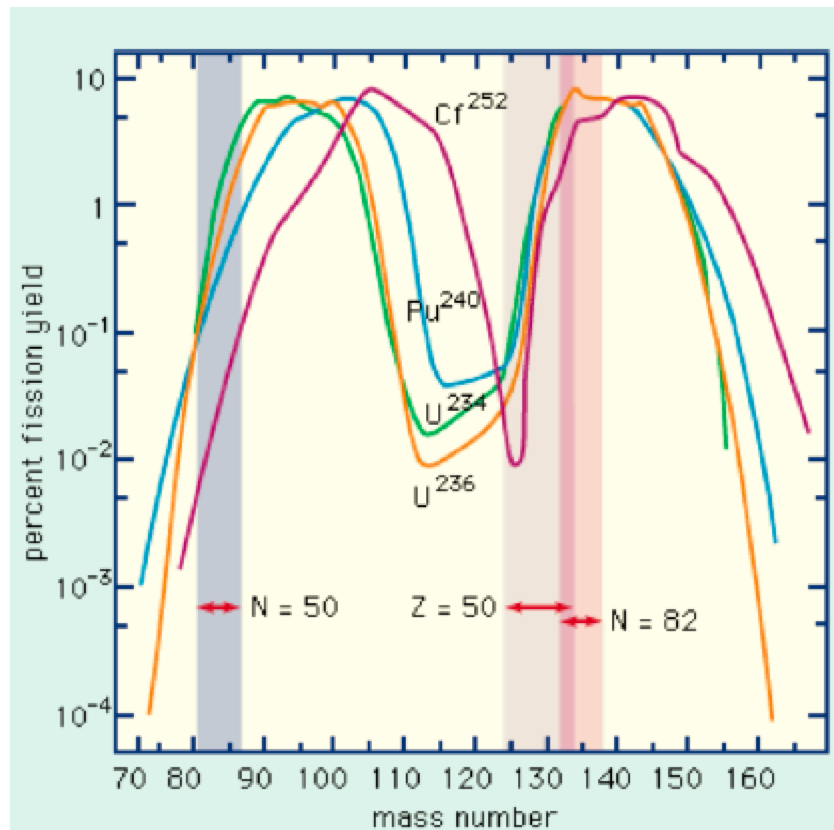
Energy in fission: Released energy (4)

- Measurements allows identification of other released energy →
  - prompt γ rays (at the instant of fission → within 10^{-14} s) → 8 MeV
  - β decays from radioactive fragments → 19 MeV
  - γ decays from radioactive fragments → 7 MeV
- Remark → the energy released during the β decay is shared between β particle and neutrino → about 30-40% is given to β particles → the remainder (≈ 12 MeV) goes to neutrinos → the neutrino energy is lost and have no contribution in practice

Nuclear structure (1)

- Previous results obtained from the liquid-drop model
- However shell effect (↔ shell model) also plays an important role
- Effect 1 → mass distribution of fragments →

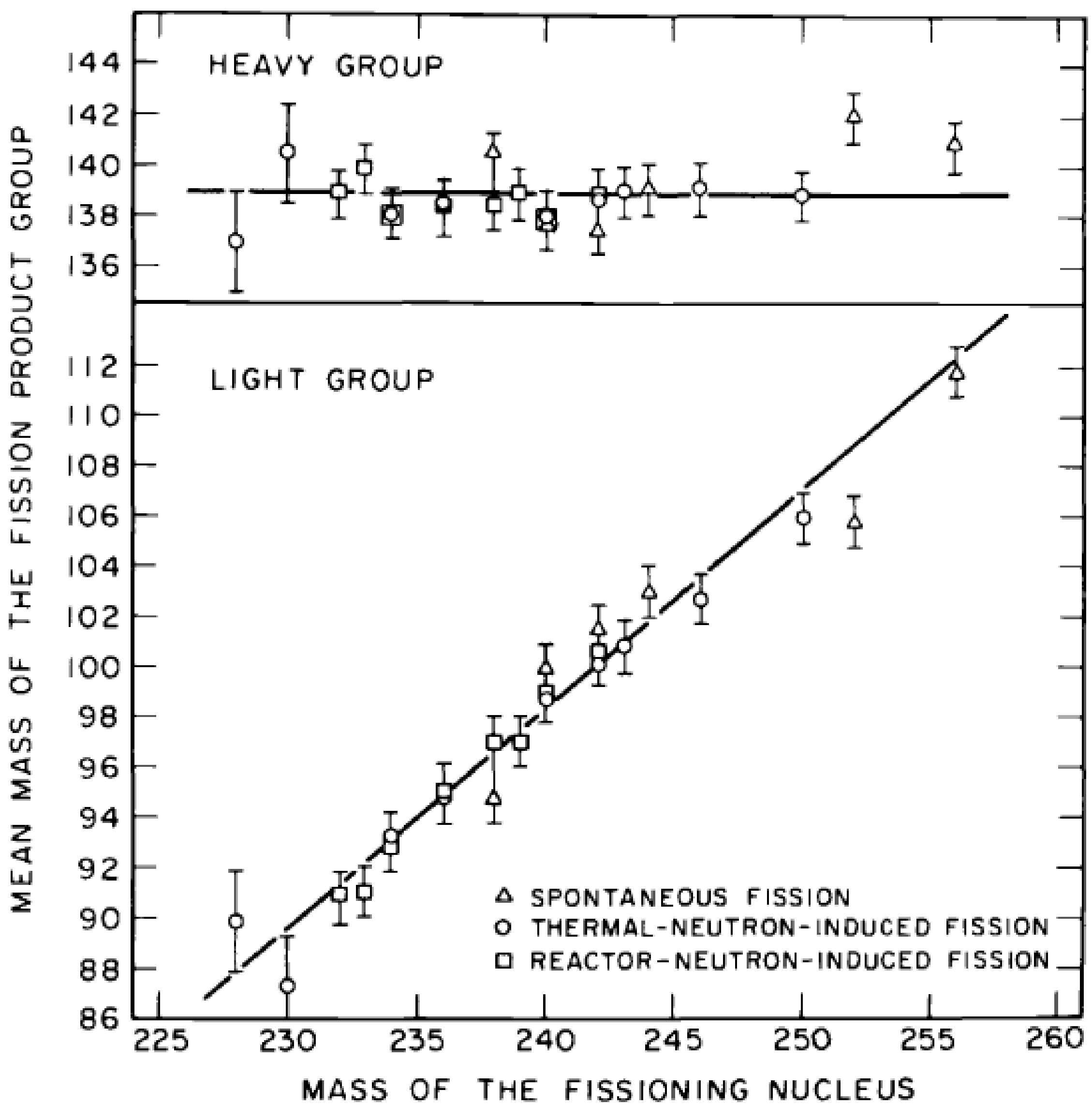
- For heavy fragments → the mass distributions overlap quite well
- For lighter fragment → large variation



Nuclear structure (2)

- Comparing ^{236}U and ^{256}Fm → *Z, N* and *A* ↗ by ≈ 8.5%
- If the liquid-drop model of fission is completely correct → shift of both the heavy and light fragment distributions by ≈ 8.5% between ^{236}U and ^{256}Fm → the average masses should go from ≈ 95 and 140 in ^{236}U to about 103 and 152 in ^{256}Fm
- Practically → the observed average masses in ^{256}Fm are ≈ 114 and 141 → the 20 additional mass goes to the lighter fragment
- More generally → looking for the average masses of the light and heavy fragments over a mass range from 228 to 256 → for heavy fragment it stays constant at ≈ 140 ↔ for light fragment it ↗ linearly with *A* → the added nucleons all go to the lighter fragment
- This is in contradiction with the liquid-drop model for which the masses would be uniformly shared

Nuclear structure (3)

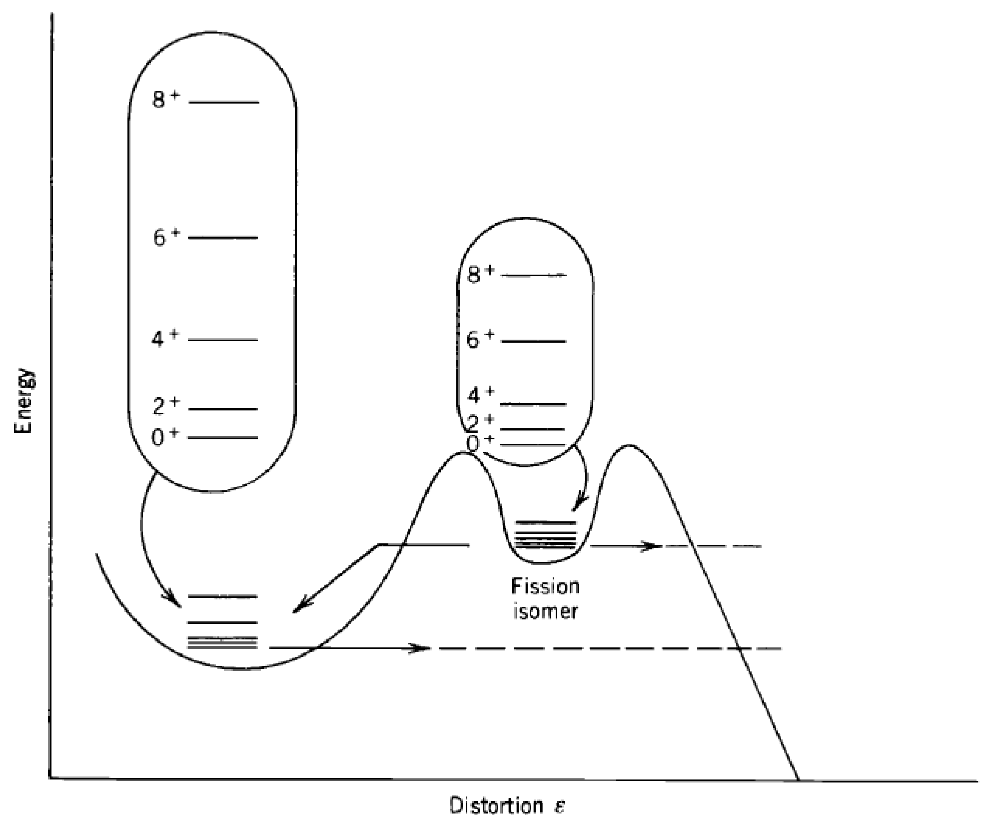


Nuclear structure (4)

- Difference can be explained with the shell model
- In previous figure are shown regions with fission fragments with shell-model magic numbers of protons or neutrons
- For heavy fragment → presence of one of this regions → especially presence of a double magic nucleus (*Z* = 50 and *N* = 82): ${}^{132}_{50}\mathrm{Sn}_{82}$
- This exceptionally stable configuration determines the low edge of the mass distribution of the heavier fragment
- No such effect occurs for the lighter fragment → unaffected by shell closures

Nuclear structure (5)

- Effect 2 → modification of the fission barrier
- Very often → deformed nuclei are stable due to the presence of shells → introduction of a double-humped barrier



Nuclear structure (6)

- For these nuclei → $E_{ex} \approx$ 2-3 MeV (far below the barrier height of 6-7 MeV) → but their half-lives for spontaneous fission are in the range of 10^{-6}-10^{-9} s
- These isotopes have states in the intermediate potential well → they could decay either by fission (through a relatively thin barrier) or by γ emission back to the ground state
- They are called fission isomers or shape isomers ↔ the word « isomer » is used because they have a long-life for γ decay
- Properties of the fission isomers controlled by the relative height of the 2 barriers →
  - For U and Pu → they are close
  - For Z < 93 (neptunium) → the left barrier is the lowest → γ decay
  - For Z > 97 (berkelium) → the right barrier is the lowest → rapid fission
- Moreover when energy states are closed in the 2 wells → resonances

Applications

- Fission reactors → see « Introduction to reactor physics » →
  - Power reactors → extraction of the kinetic energy of the fission fragments as heat → conversion of that heat energy to electrical energy
  - Research reactors → production of neutrons for research (nuclear physics, solid-state physics,…) → particular case: MYRRHA (Multi-purpose hYbrid Research Reactor for High-tech Applications) → nuclear reactor coupled to a proton accelerator (Accelerator-driven system or ADS)
- Fission explosives (no comment…)
- Neutron detectors based on fission reactions → Ionization chamber with fissile coating → see « Nuclear Metrology Techniques »