Relevant High-energy Data for ADS

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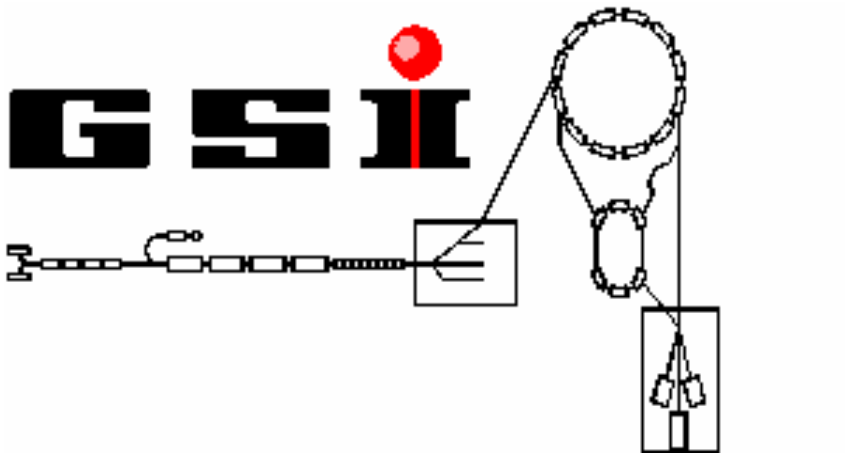


Second IP EUROTRANS Internal Training Course
Nuclear Data for Transmutation: status, needs and methods
Santiago de Compostela (Spain)
June 7 - 10, 2006

Lay out

- Introduction
- Physics
- Experimental methods
- Results
- Detailed considerations on the production of heavy residues (optional)

The ADS concept

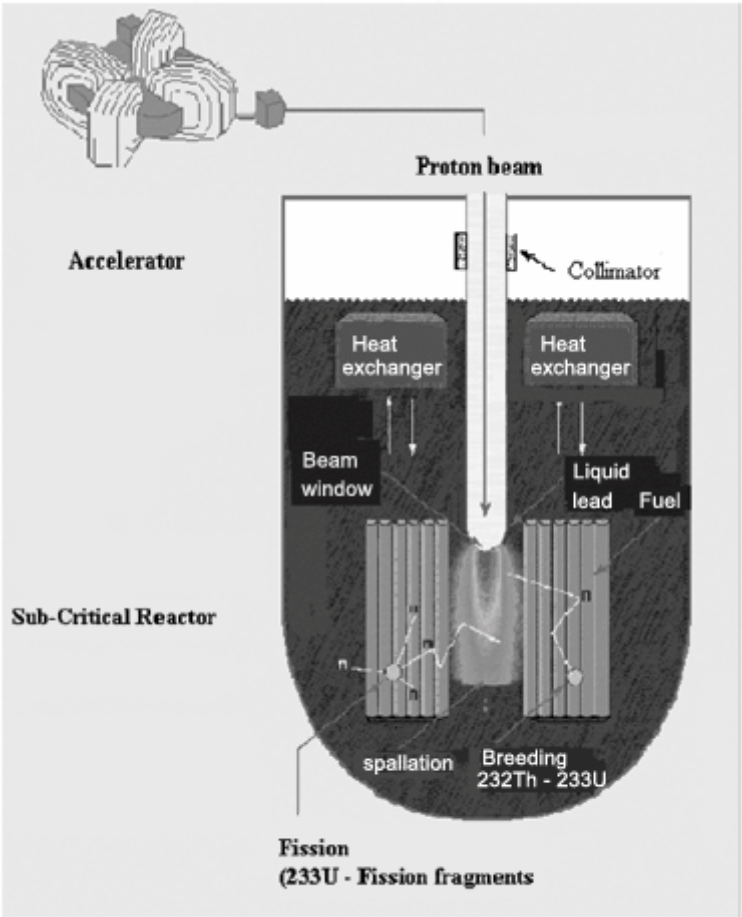


Figure 1. Schematic representation of Energy Amplifier proposed by Rubbia [4].

Proton accelerator (≈ 1 GeV)

Subcritical fission reactor

with

Spallation neutron source

Design of ADS requires knowledge on relevant nuclear data up to 1 GeV!

Concept for high-energy data

Variety of targets and energies cannot be fully covered by experiment

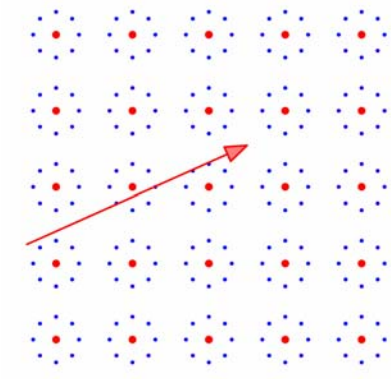
- **Selecting a few systems and energies for experimental studies.**
- **Development of realistic codes.**
- **Basic understanding needed, not always directly related to sensitivity.**

General interest in spallation reactions

- ADS
- Spallation neutron source
- Reactions of cosmic rays (high-energetic heavy nuclei) with 'interstellar matter (hydrogen)
- Behaviour of strongly heated nuclear matter (equation of state of nuclear matter)

Passage of protons through matter
1. Non-nuclear interactions → energy loss

Basic features of Coulomb interactions:



$$\Delta\vec{p} = \int \vec{F}\mathrm{d}t \propto \frac{Z_p \cdot e}{d_{eff}} \frac{1}{v}$$

$$\Delta E = \frac{\Delta p^2}{2m} \propto \frac{Z_p^{\ 2}}{v^2} \frac{1}{m}$$

- **Energy loss increases strongly with decreasing velocity (1/v^2).**
- **Energy loss is dominated by collisions with electrons (1/*m*).**
- **Source of heat load in the spallation target.**
- **Little angular scattering (like stopping a truck in gravel).**

More complete: Bethe-Bloch formula:

$$-\frac{\mathrm{d}E}{\mathrm{d}x} = \frac{4\pi}{m_c c^2} \cdot \frac{nZ_p^{\ 2}}{\beta} \cdot \left(\frac{e^2}{4\pi\varepsilon_0}\right)^2 \cdot \left[\ln\left(\frac{2m_e c^2 \beta^2}{I \cdot \left(1-\beta^2\right)}\right) - \beta^2\right] \; ; \; \beta = \frac{v}{c}$$

Relativistic increase of d*E*/d*x* above $\gamma = 3$ **;** γ **= Lorentz parameter.**

Passage of protons through matter
2. Nuclear interactions → attenuation

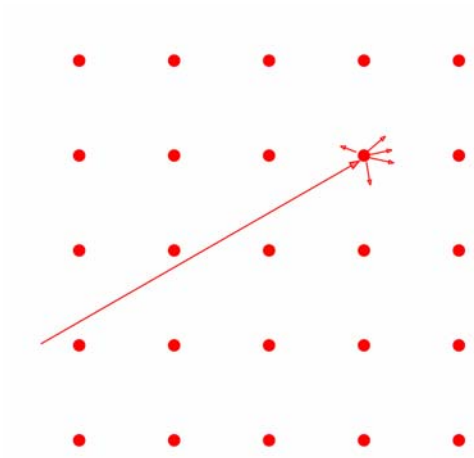
Basic features of nuclear interactions:
Glauber approach to the total reaction cross section (absorption on trajectory of the proton through the density profile of a target nucleus)

$$\sigma_R = 2\pi \int_0^\infty b[1 - T(b)]\mathrm{d}b$$

$T(b) =$ **Transparency function**

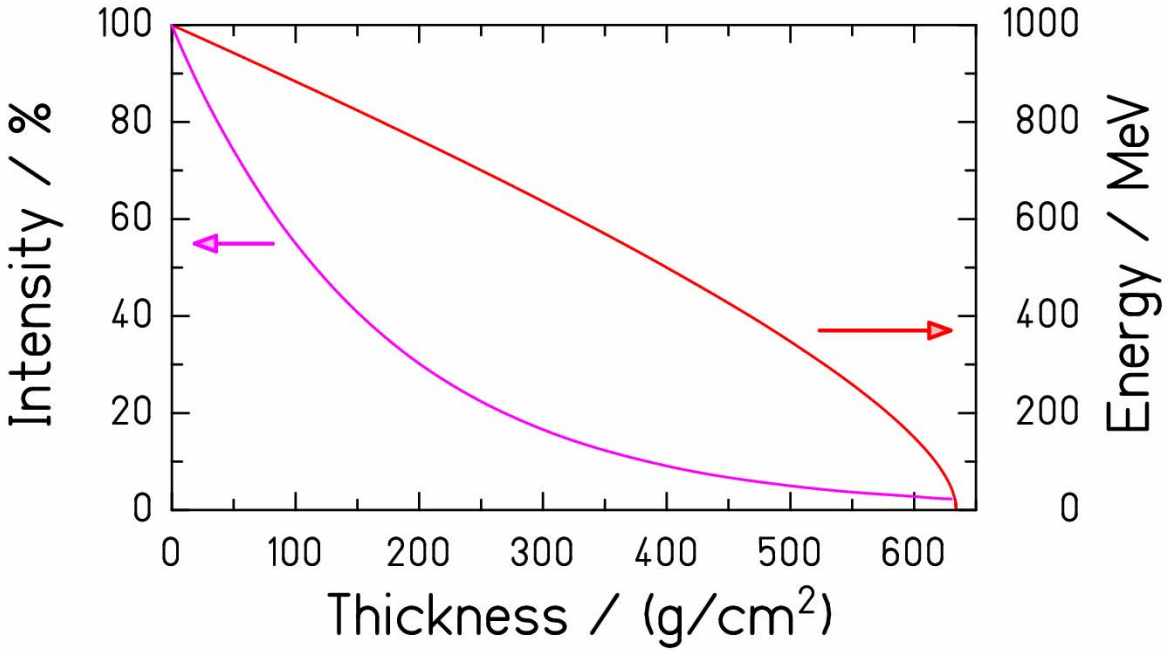
$$T(b) = \exp(-\sigma_{NN}\chi(b))$$

$\chi(b) =$ **Overlap integral of the nuclear densities along the trajectory**



- **Total cross section roughly equal to geometrical cross section**

Stopping 1 GeV protons in lead
Quantitative results

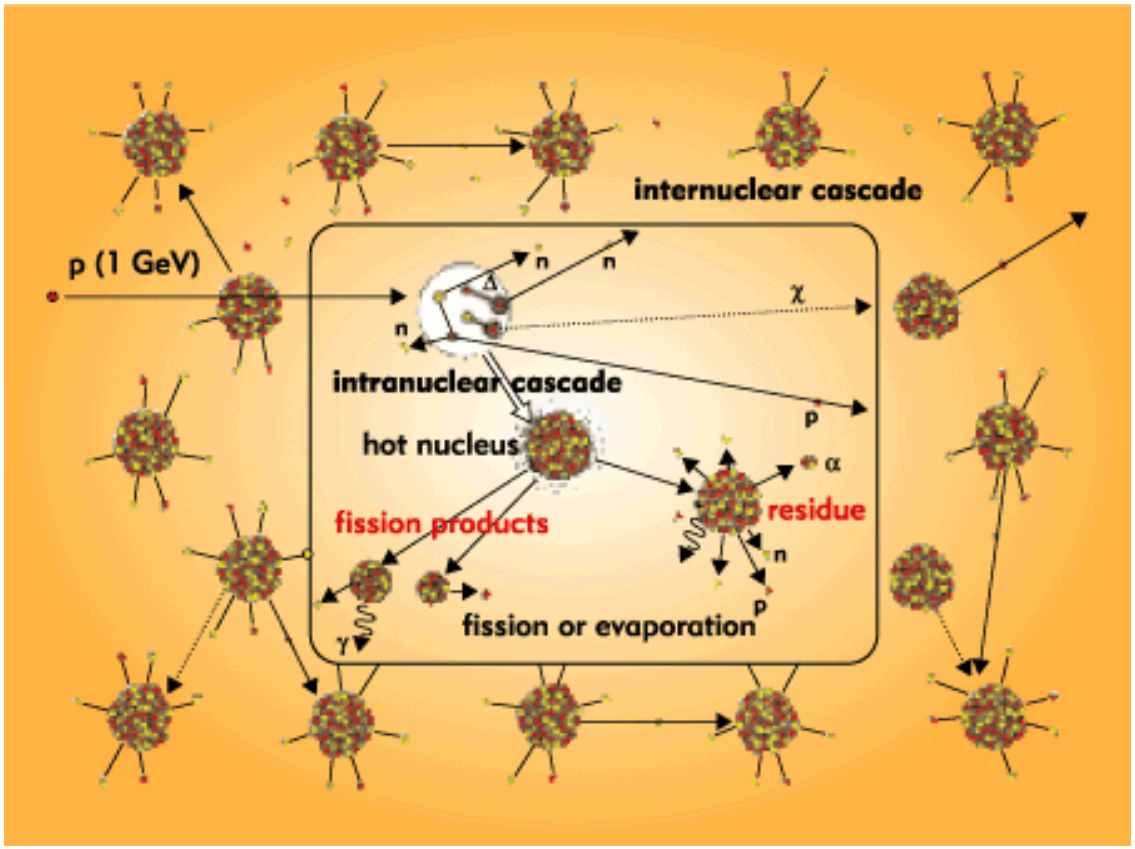


- **Total range of 1 GeV protons in lead is 634g/cm^2 = 56 cm.**
- **Proton beam is strongly absorbed along the target.**
- **Mean energy above 800 MeV.**
- **Reactions of primary protons mostly above 500 MeV.**

Passage of 1 GeV protons through lead
Important facts

- Slowing down by Coulomb interactions with electrons.
- Range is 56 cm in lead.
- dE/dx proportional to $1/v^2$ (for $\gamma < 3$).
- Straight-line trajectories.
- Attenuation length by nuclear reactions is 20 cm (≈ 1/3 of range).

How to model the spallation process?



Full quantum-mechanical treatment of the nucleon-nucleus collision process not available.

Nuclear-reaction mechanisms at ≈ 1 GeV

Decisive parameter:

de Broglie wavelength of a nucleon: $\lambda = \frac{h}{p}$.

E	*p*	λ
10 MeV	137 MeV/c	9.03 fm
100 MeV	443 MeV/c	2.79 fm
1 GeV	1692 MeV/c	0.73 fm

Compared to

- **nuclear radius (r = 1.16 fm × $A^{1/3}$)**
- **range of nuclear force (≈ 1 fm)**

Spallation reaction ≈ collisions of individual nucleons.

Intranuclear cascade codes treat the collision stage of spallation like billard

Scattering of nucleons calculated as classical quasi-free nucleon-nucleon collisions with nucleon-nucleon cross sections.

Problems:
Mean field (nuclear potential).
Pauli exclusion principle.
Fermi motion.
Excitation of the nucleon.

Quantum-mechanical features „screwed on“

Fermi gas

Volume in phase space $\Omega = \frac{4}{3}\pi r^3 \times \frac{4}{3}\pi p^3$

Number of states

$$N = \frac{\Omega}{\mathrm{h}^3} = \frac{\frac{4}{3}\pi r^3 \times \frac{4}{3}\pi p^3}{\mathrm{h}^3}$$

Fermi sphere in momentum space

Number of states per momentum interval

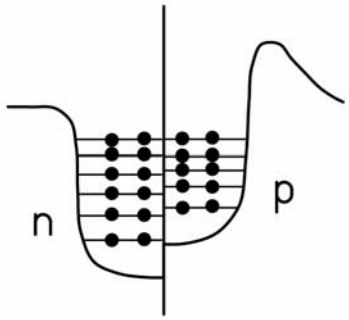
$$\mathrm{d}N = \frac{\frac{4}{3}\pi r^3 \times 4\pi p^2}{\mathrm{h}^3}\,\mathrm{d}p$$

Volume in space $\frac{4}{3}\pi r^3 = V$

sphere in local space

Momentum - energy $p^2 = 2mE$

$$p^2 dp = \sqrt{2m^3 E}\,\mathrm{d}E$$



levels in energy

Number of states per energy interval

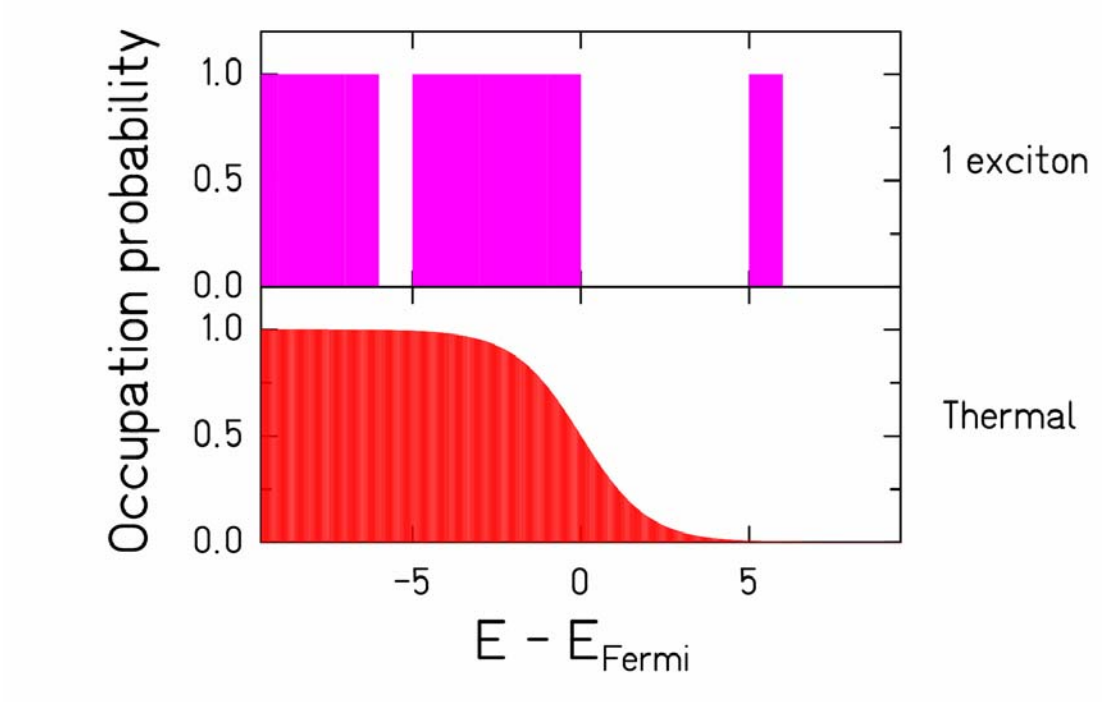
$$\mathrm{d}N = V \times \frac{4\pi\sqrt{2m^3}}{\mathrm{h}^3}\sqrt{E}\,\mathrm{d}E$$

(additional factor of 2 for spin degeneracy)

Pre-equilibrium emission (exciton model)

Simple initial single-particle excitations decay into more complex particle-hole excitations. Unbound particles may escape.

Evolution vs. thermal equilibrium.



Thermal expansion

$$S = 2\sqrt{aE}$$

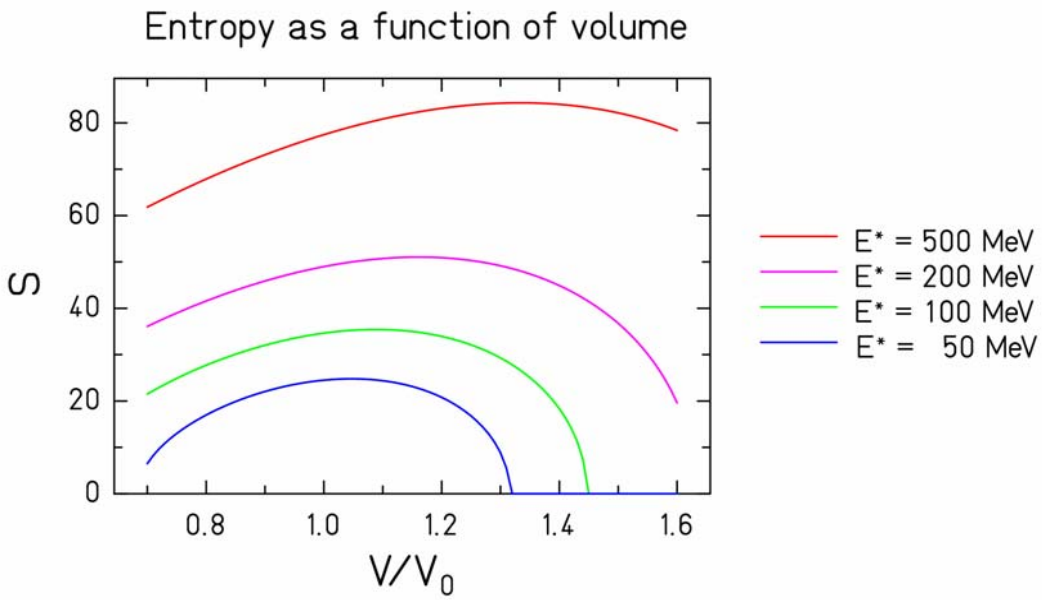
Fermi-gas level density

$$a \propto V$$

Level density parameter grows with volume

$$E = E_0 - c(V - V)^2.$$

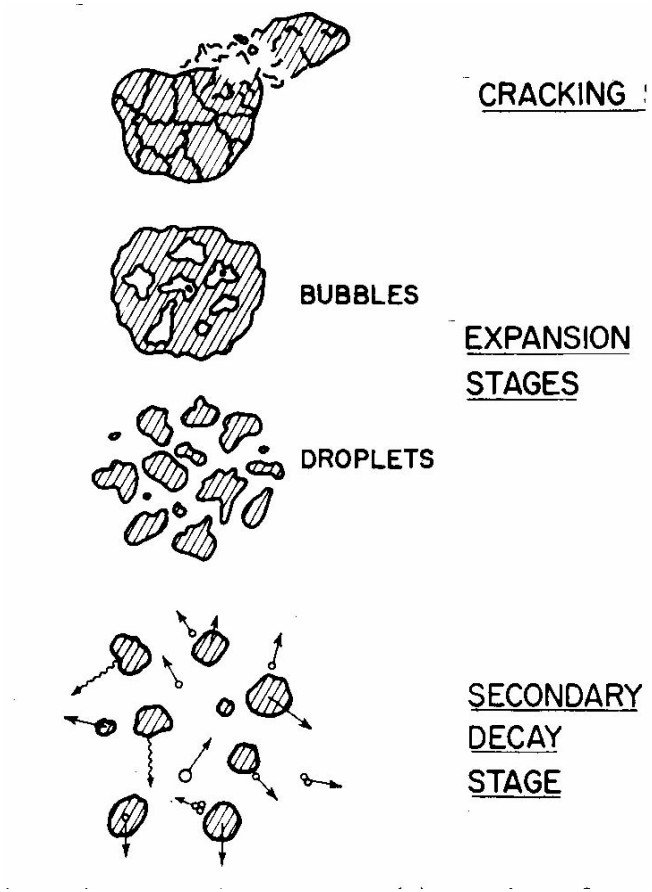
Compression energy relative to normal density



Highly excited nuclei expand – configuration of maximum entropy

Multifragmentation

Expansion may lead system into spinodal (liquid-gas) instabilities.
This may induce multifragmentation.



Multifragmentation occurs when E^*/A > 3 MeV.

- **High multiplicities of IMFs.**
- **Expanding kinematics.**

Particle evaporation (Weisskopf)

Basic principle: Thermo-dynamical equilibrium

Evaporation from a boiling liquid - Condensation from the surrounding saturated gas

$$W_n(E_\nu)\cdot\rho_A(E_A^*)=W_c(E_\nu)\cdot\frac{\mathrm{d}N}{\mathrm{d}E_\nu}\cdot\rho_B(E_B^*)$$

W_n: evaporation probability
ρ_A : state density of mother nucleus
W_c : capture probability
ρ_B : state density of daughter nucleus

E_ν : kinetic energy of evaporated particle
E^*_A : excitation energy of mother nucleus
$\mathrm{d}N/\mathrm{d}E_\nu$: kinematical phase-space density
E^*_B : excitation energy of daughter nucleus

$W_c(E_\nu)=\sigma_c(E_\nu)\cdot\Phi_\nu$ **$\sigma_\nu(E_\nu)$: capture cross section, $\Phi_\nu=\frac{v}{V}$: particle flux.**

Finally:

$$\Gamma_\nu=\frac{gm_\nu}{\pi^2\hbar^3\rho_A(E_A^*)}\int_0^{E_\nu^{\max}}\sigma_c(E_\nu)\rho_B(E_A^*-S_\nu-E_\nu)E_\nu dE_\nu$$

Γ_ν: particle decay width
m_ν: mass of evaporated particle

g: spin degeneracy
S_ν: particle separation energy

Partial widths Γ_n and Γ_p for emission of neutrons and protons.

$$\Gamma_n = \frac{2mR^2 g}{\hbar^2 2\pi\rho(E^*)} \int_0^{E^*-S_n} \varepsilon\rho(E^* - S_n - \varepsilon)\mathrm{d}\varepsilon$$

$$\Gamma_p = \frac{2mR^2 g}{\hbar^2 2\pi\rho(E^*)} \int_{\varepsilon_c}^{E^*-S_p} \varepsilon\left(1 - \frac{\varepsilon_c}{\varepsilon}\right)\rho(E^* - S_p - \varepsilon)d\varepsilon$$

(Approximation without considering tunneling, geometrical cross sections inserted)

S_n, S_p are the separation energies.
Γ_p is reduced by the Coulomb barrier ε_c with respect to Γ_n.

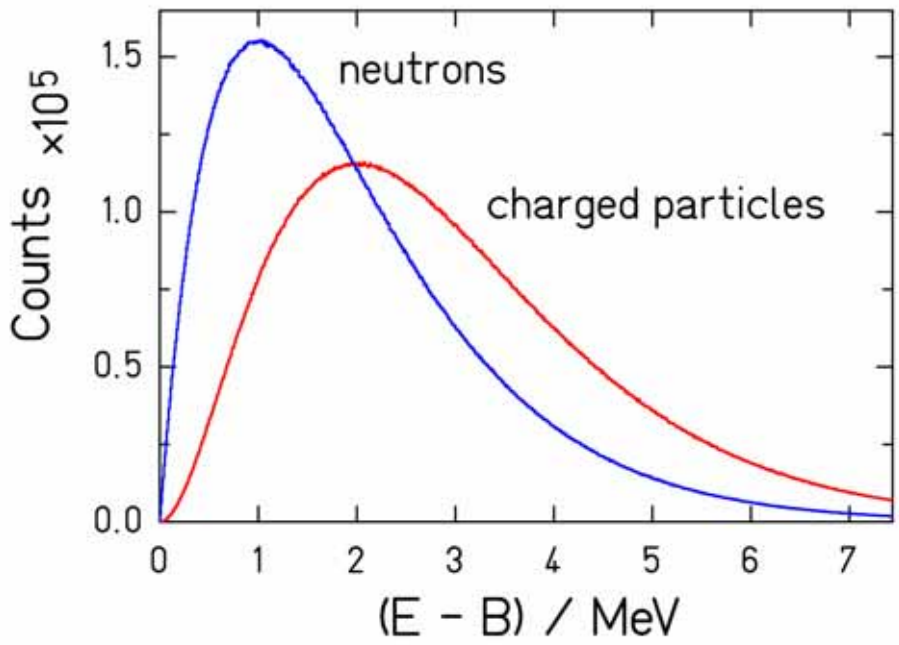
Similar expressions for evaporation of other charged fragments.

Energy spectra in evaporation

Neutrons:

Maxwellian distribution

$$\mathrm{d}N/\mathrm{d}E \propto E \cdot \exp(-E/T)$$



Charged particles:

Maxwellian modified by Coulomb factor

$$\mathrm{d}N/\mathrm{d}E \propto E \cdot \frac{E}{E+B} \cdot \exp(-E/T)$$

(tunnelling neglected)

Competition in the evaporation process

$$P_\nu = \frac{\Gamma_\nu}{\sum_i \Gamma_i}$$

Nuclear level density

Nuclear level density:

$$\rho(E^*) = \frac{\mathrm{d}N}{\mathrm{d}E^*}$$

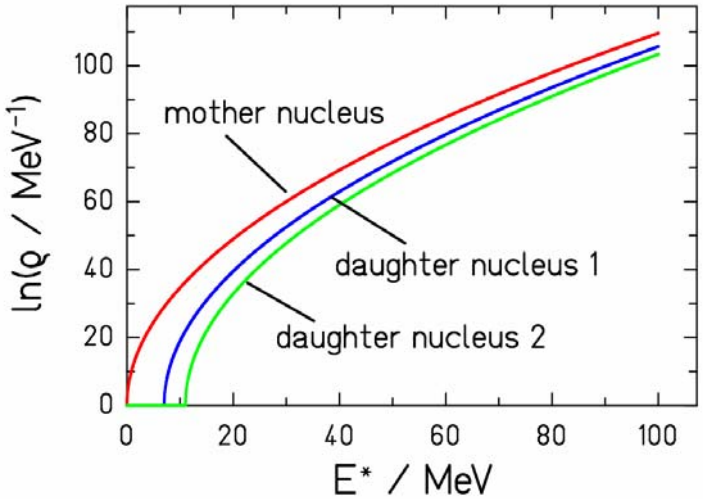
Number of combinations of all single-particle excitations, leading to a certain energy interval d*E.**

Approximate result:

$$\rho(E^*) \propto e^{2\sqrt{aE^*}}$$

a: level-density parameter

Approach starting from a constant single-particle level density.
(Also known as
"Fermi-gas level-density formula".)



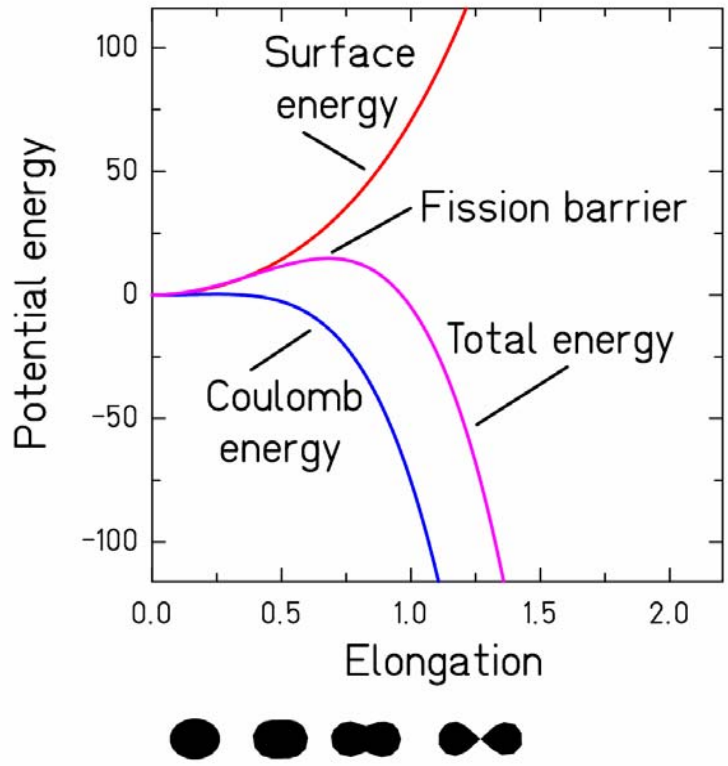
Competition between different decay channels is governed by the level densities above the thresholds.

Fission as an additional decay channel
Large quadrupole oscillations are unstable

Different shape dependences of surface and Coulomb energy create the fission barrier.

Schematic presentation →

Fission is an important additional decay channel for heavy nuclei.

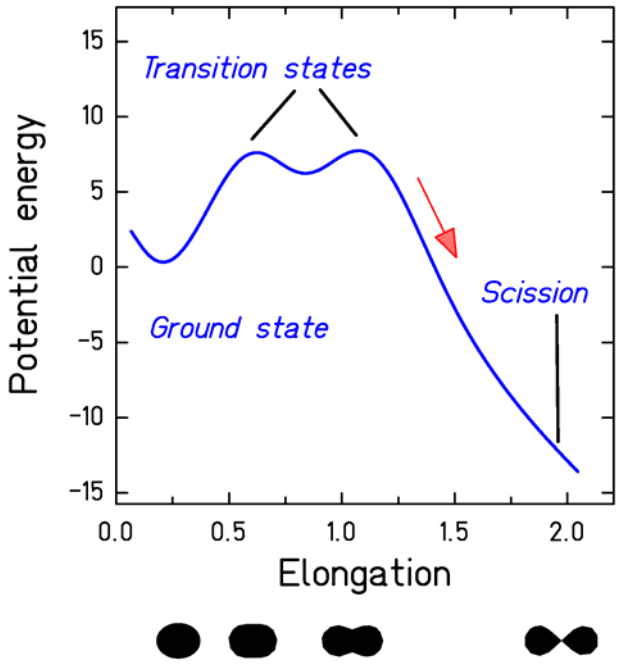


Transition-state model of Bohr and Wheeler

Bohr and Wheeler recognised that the fission probability is governed by the number of states above the fission barrier ("transition states") (not by the number of states of the final fission fragments).

The Bohr-Wheeler description of the fission decay width is very similar to the formula for particle decay:

$$\Gamma_f = \frac{1}{2\pi\rho(E^*)} \int_0^{E^*-B_f} \rho_{saddle}(E)\,\mathrm{d}E$$



The picture is complicated by shell effects: four configurations have to be considered: ground state, inner saddle, second minimum, outer saddle.

The decision whether fission occurs is made at the outer saddle, there is no return possible beyond this point.

Langevin equations

$$\frac{dq}{dt} = \frac{p}{\mu(q)}$$

Derivative of position (deformation) *q* is determined by the momentum *p*.

$$\frac{dp}{dt} = \frac{1}{2}\left(\frac{p}{\mu(q)}\right)^2 \frac{d\mu(q)}{dq} - \frac{dV(q)}{dq} - \frac{\gamma(q)}{\mu(q)} p + \sqrt{D(q)}\, f_L(t)$$

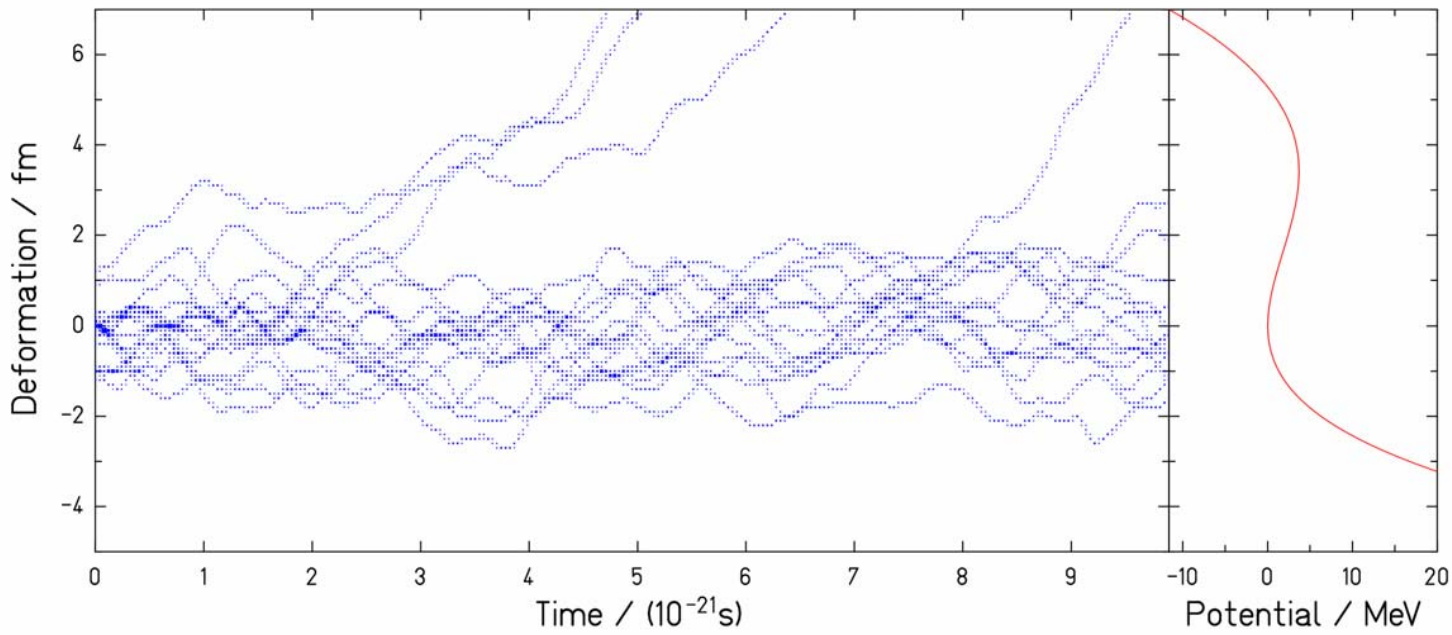
Derivative of momentum is determined by

inertia term **potential-energy term** **friction term** **fluctuating term**

$\mu(q)$ **and** $\gamma(q)$ **are the nuclear inertia and friction coefficient, respectively.** $V(q)$ **is the nuclear potential. The Langevin random force describes the fluctuating, or *Brownian*, part of the surrounding medium on the motion of the particle. In the framework of the fluctuation-dissipation theorem, the diffusion coefficient** $D(q)$ **is related to friction via the Einstein relation:** $D(q) = \gamma(q)T$ **.**

Dynamical and dissipative effects in fission

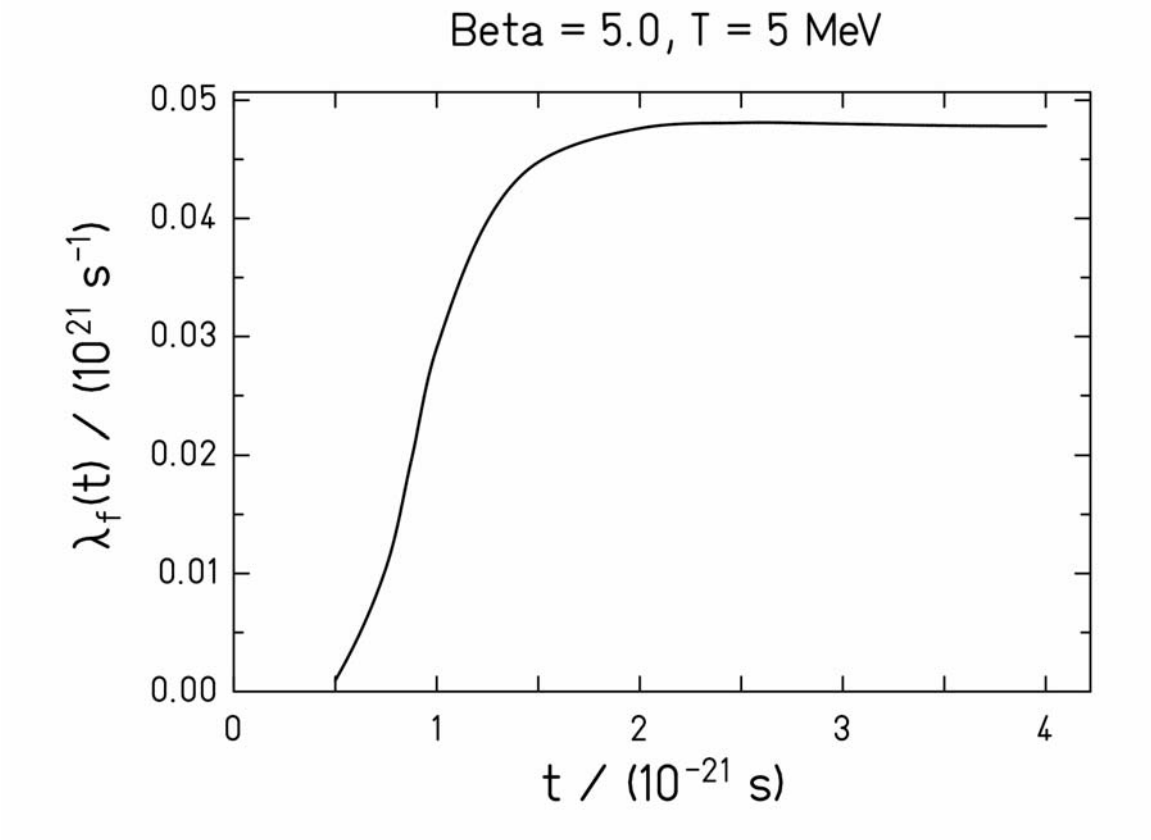
- **Fission is slow compared to the intrinsic excitations.**
- **Bohr-Wheeler model misses these dynamical aspects.**
- **Fission dynamics is treated by Fokker-Planck or Langevin equation.**



- **Highly excited nuclei may cool down by evaporation before being able to fission.**

Transient effects

Solution of the Fokker-Planck equation (integral form of the Langevin equations):



Flux over fission barrier starts with a time delay.

Stages of the spallation reaction - Important facts

- **Nucleon-nucleus collisions (INC stage)**
  - **Quasi-free nucleon-nucleon collisions**
  - **Excitations of the nucleons (Δ resonance)**
  - **Deposition of excitation energy (typical ≈ 20 % of initial energy)**

- **Thermal expansion (multifragmentation stage)**
  - **If E/A > 3 MeV → multifragmentation**

- **Spreading of-particle-hole excitations (pre-equilibrium stage)**
  - **Pre-equilibrium emission (exciton model)**
  - **Thermalization of single-particle excitations**

- **Statistical decay from a compound nucleus (evaporation-fission stage)**
  - **Evaporation of nucleons and light fragments**
  - **Fission (dynamical treatment)**
  - **Gamma deexcitation (mostly below the particle threshold)**

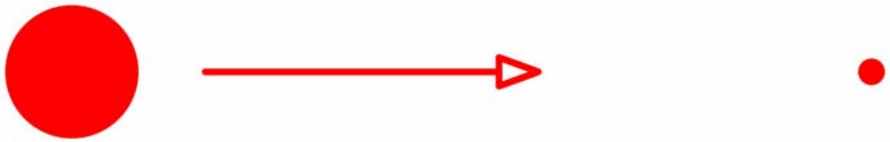
- **Radioactive decay**
  - **Mostly beta decay towards the beta-stability valley**

Detector systems

- **Normal kinematics (Proton projectiles on heavy target nuclei):**

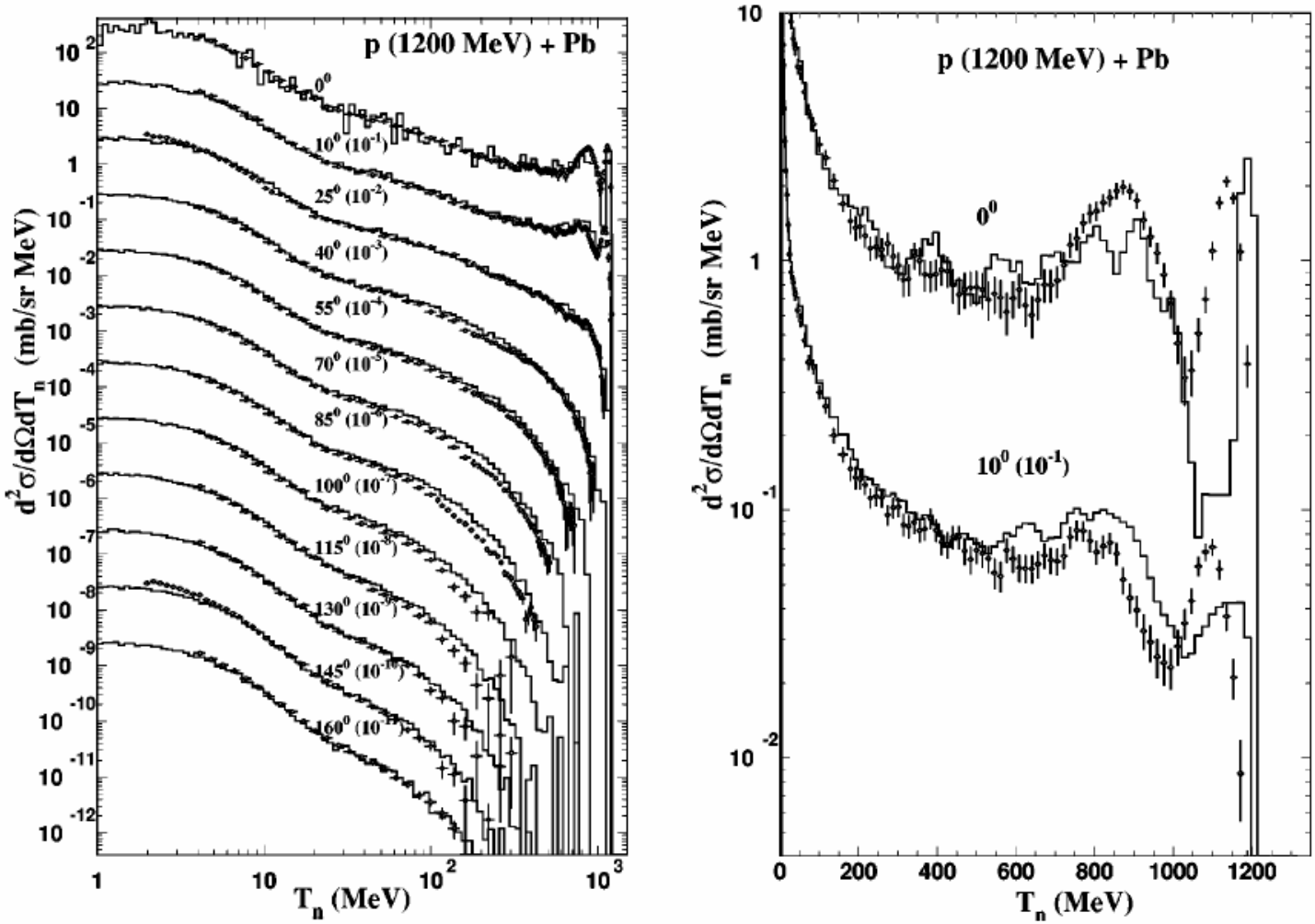
  - **Neutrons** ($d^2Y/(dE\, d\theta)$) **(kinematical detectors)**
  - **Neutrons** **(total yield)** **(moderation and capture of neutrons)**
  - **Light charged particles** ($d^2Y/(dE\, d\theta)$) **(ΔE -E, e.g. silicon)**
  - **Heavy residues** **(a few independent yields, cumulative yields)**
    (activation, gamma spectroscopy, accelerator mass spectrometry)

- **Inverse kinematics (Heavy projectiles on hydrogen target):**

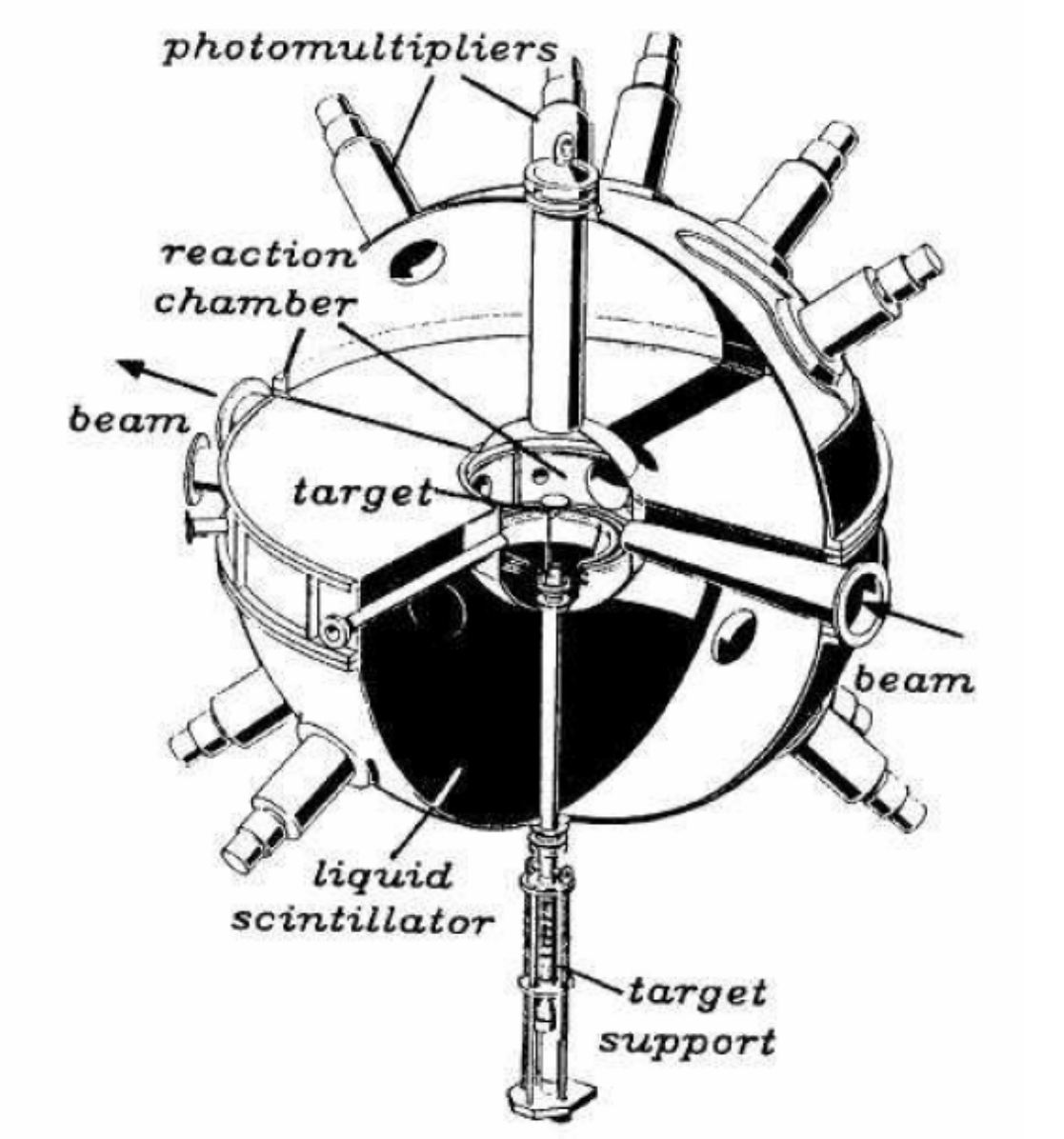


  - **Heavy residues** ($Y(Z,A)$, dY/dv)
    (in-flight identification $B\rho$ – ToF – ΔE)

Double-differential neutron spectra



S. Leray et al. (2002)



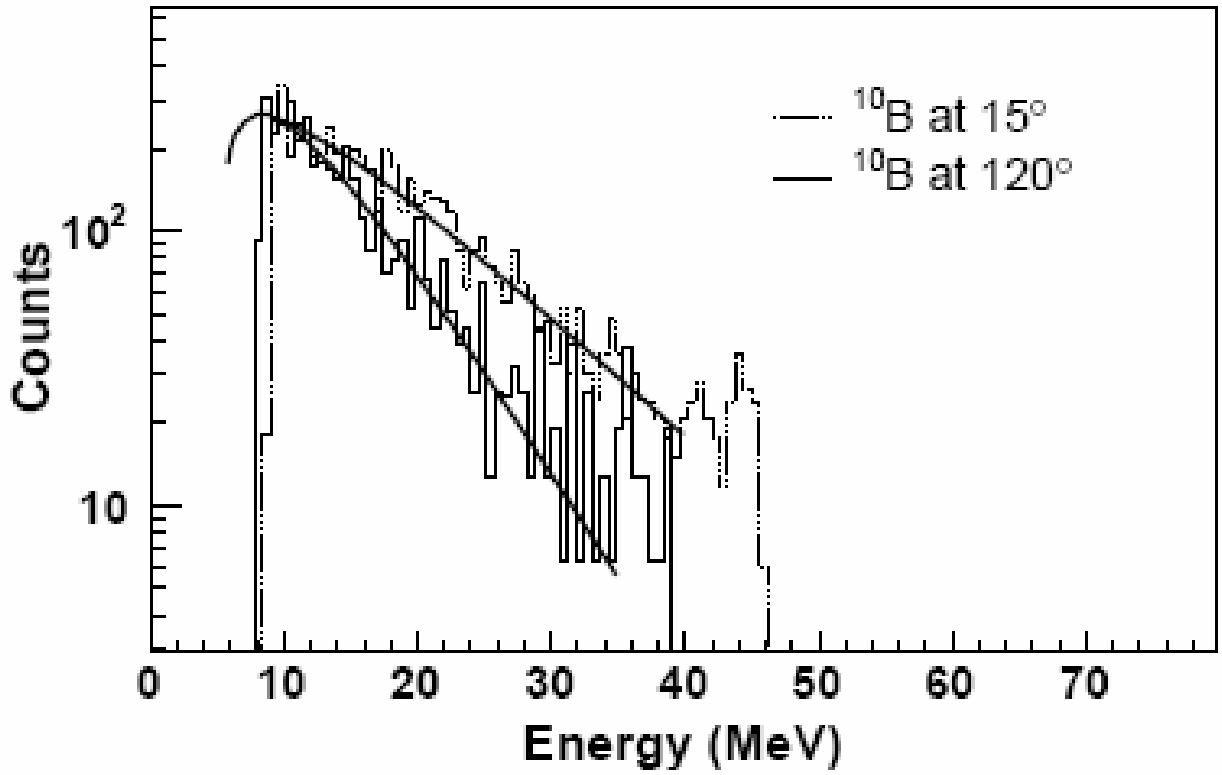
The Berlin neutron ball

1.5 m^3 0.4% Gd-loaded liquid scintillator

Neutrons are moderated and captured (in about 10 µs).

Measures total neutron yields in thick-target experiments.

Energy spectra of fragments from the PISA experiment



(a) ^{10}B from 1.9 GeV p+Ni at 15° and 120°

Goldenbaum et al. (2003)

Excitation functions of independent and cumulative yields

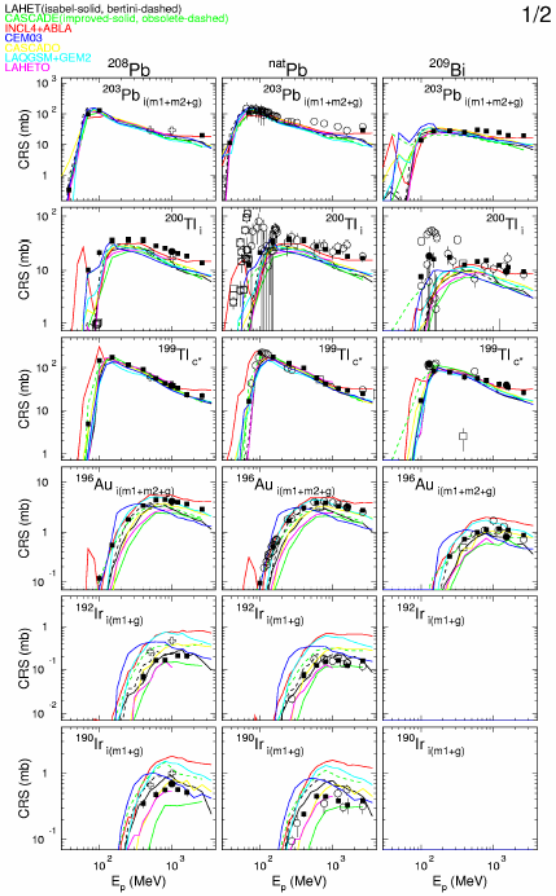


Fig. 2. Experimental and simulated excitation functions of ^{203}Pb, ^{200}Tl, ^{199}Tl, ^{196}Au , ^{192}Ir, and ^{190}Ir produced in ^{208}Pb (left), ^{nat}Pb (center), and ^{209}Bi(right). (■ – this work, ● – [2],

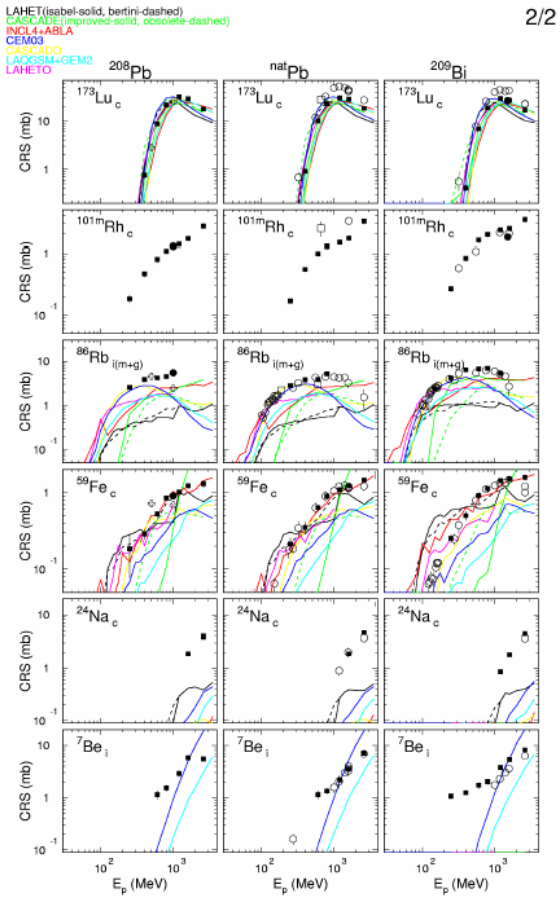
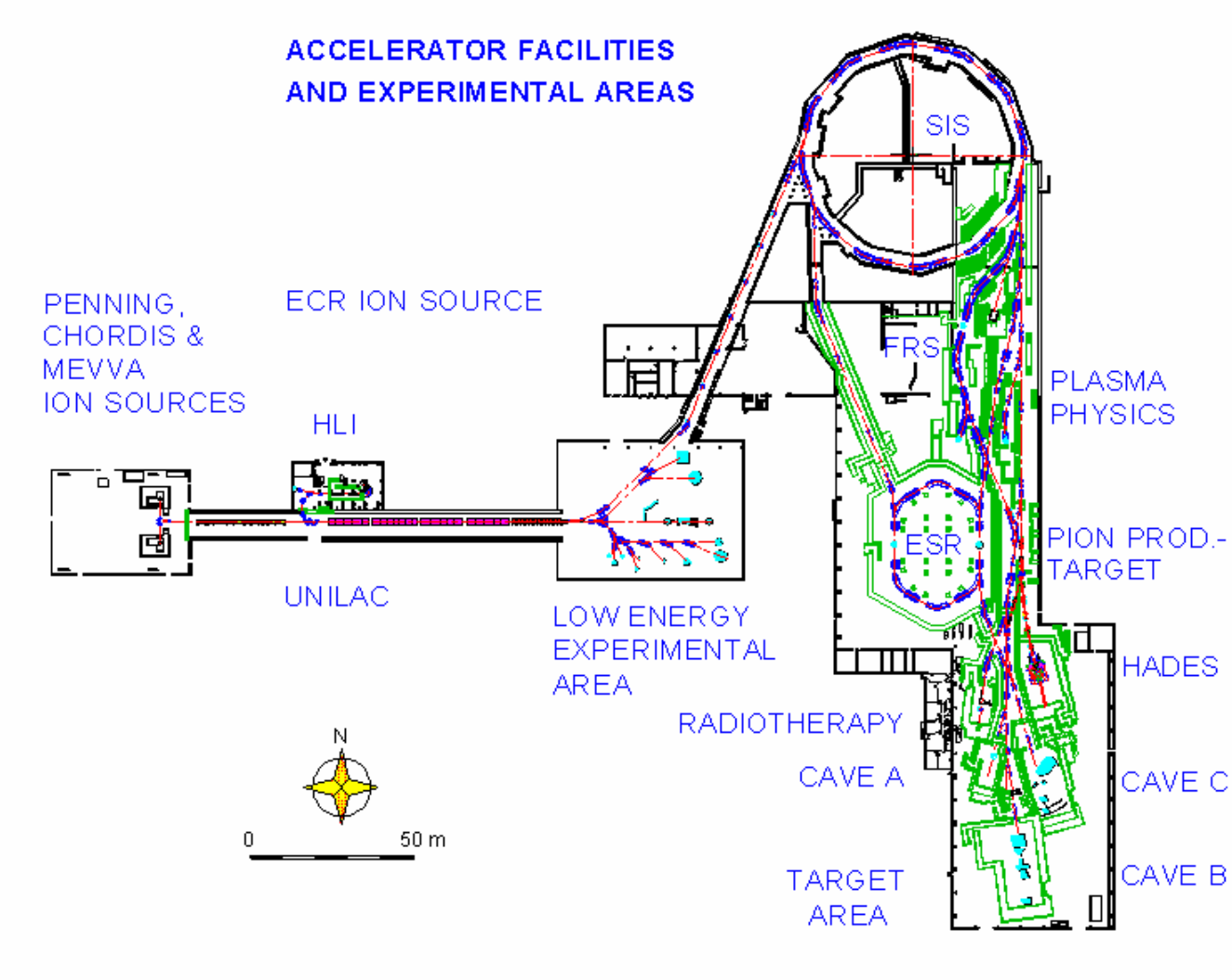


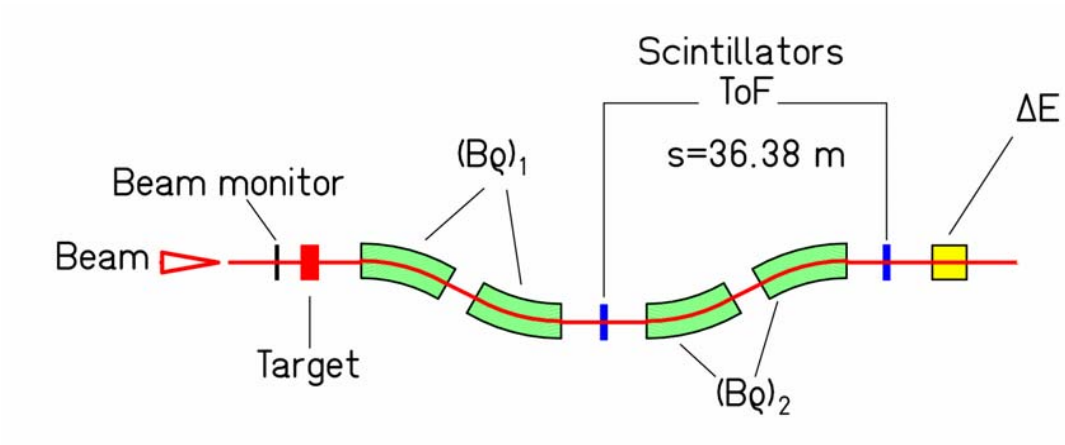
Fig. 3. The same as Fig. 2 but for ^{173}Lu, ^{101m}Rh, ^{86}Rb, ^{59}Fe, ^{24}Na, and ^{7}Be.

Titarenko et al., 2005

The GSI facility (Darmstadt, Germany)

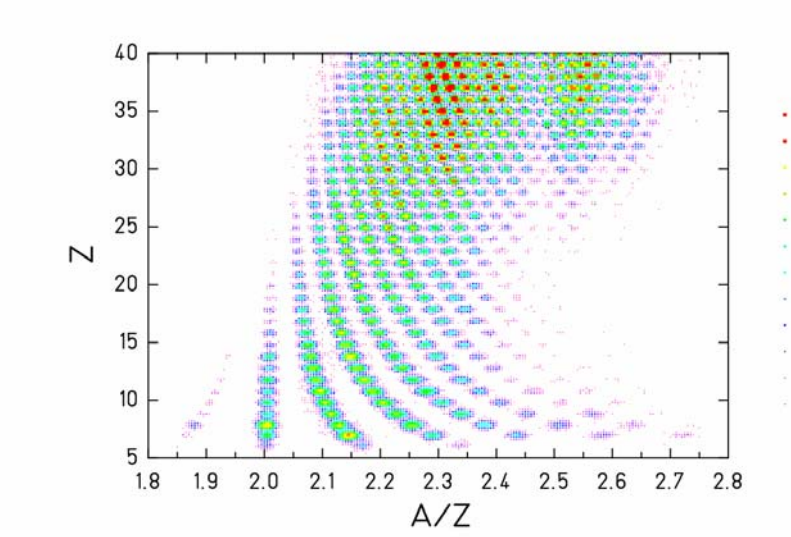


The GSI fragment separator as a magnetic spectrometer



Spectrometer

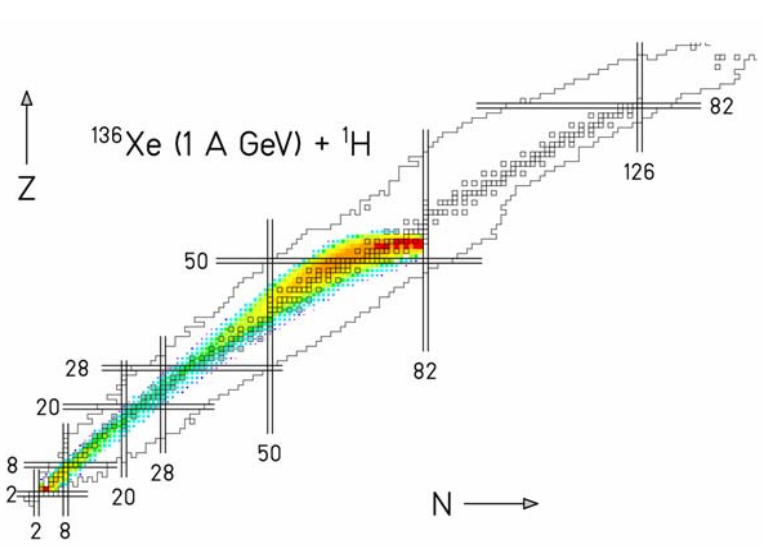
^{238}U (1A GeV) + ^{1}H

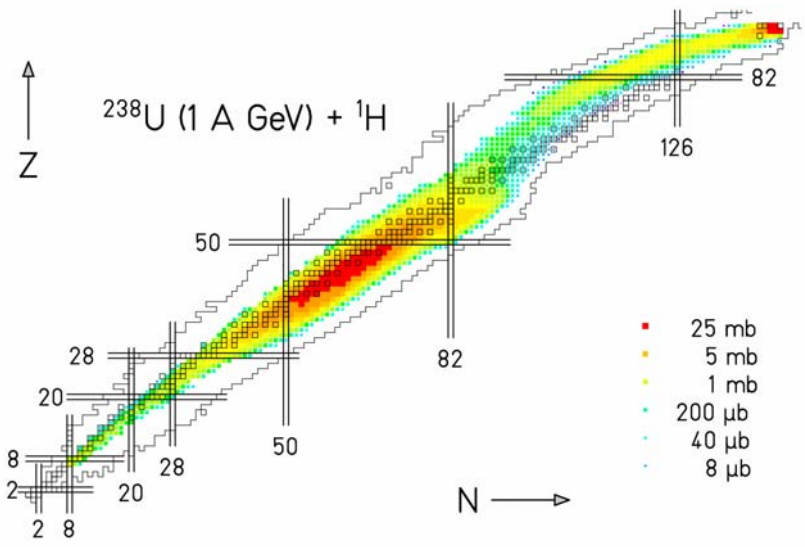


Nuclide identification
(M. V. Ricciardi)

Resolution $p/\Delta p \approx 2000$.
Acceptance $\Delta B\rho / B\rho = 3\%$ and Θ_{max}=15 mr.
Identification in *Z* and *A* by magnetic deflection, tracking, *ToF* et ΔE.
Basic equations: $B\rho = m_0 A c \beta \gamma / (e Z)$ et $\Delta E \propto Z^2 / v^2$

General features of spallation reactions

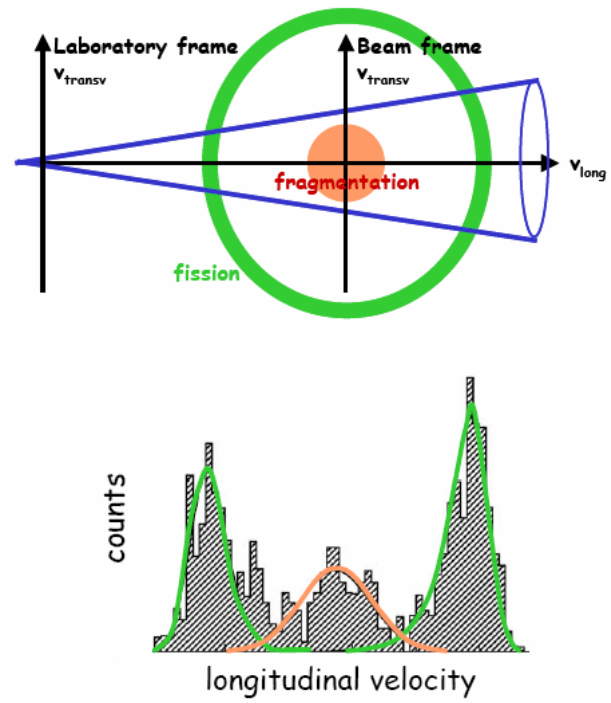


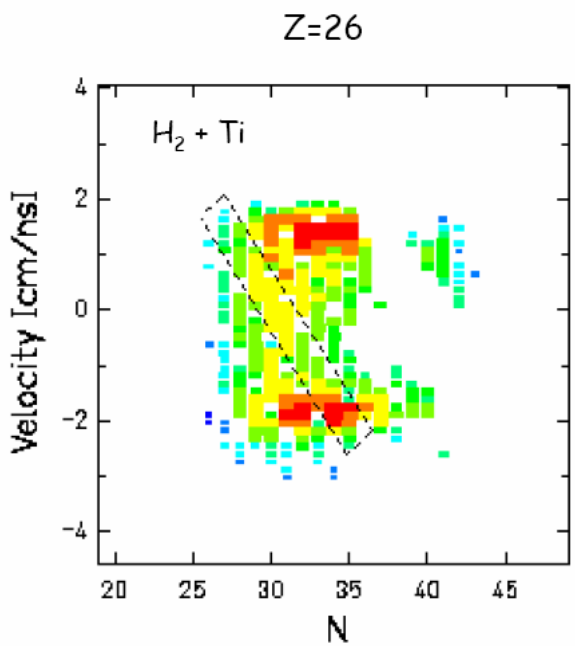


- Spallation-evaporation produces nuclides reaching from the projectile to about 10 to 15 elements below. (A few neutron-rich, most neutron-deficient)
- Spallation-fission (from Th, U) produces neutron-rich nuclides up to Z = 65.

Experiments performed at GSI

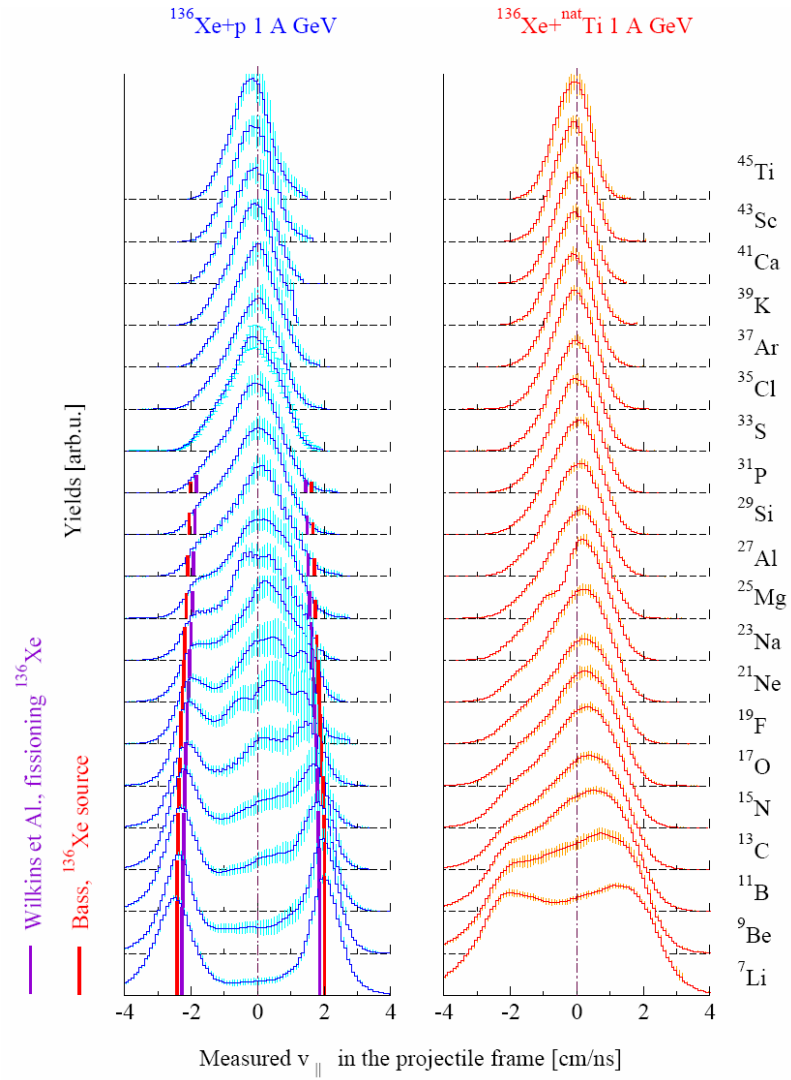
Kinematical signature of fission



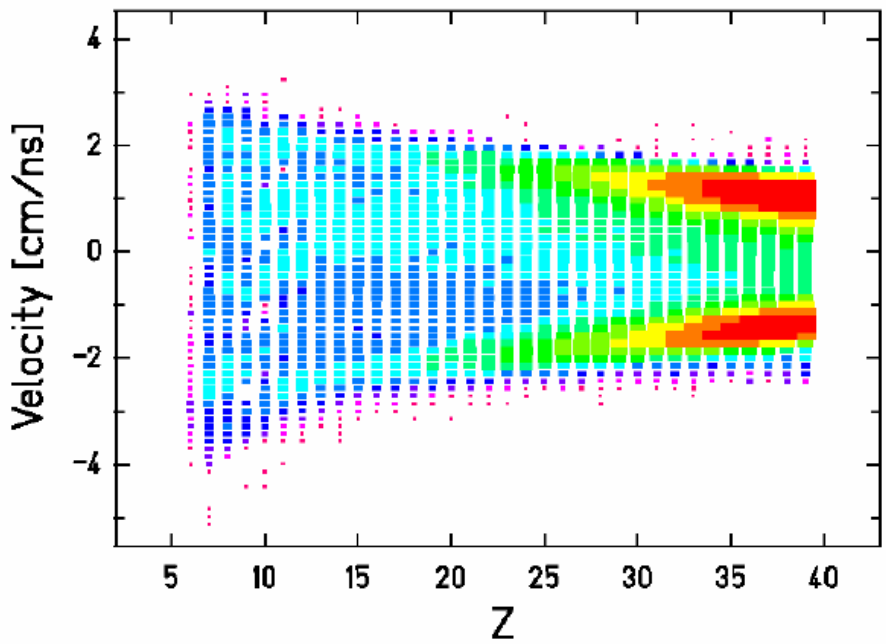


The Coulomb repulsion of the fission fragments causes a double-humped profile in the measured velocity distribution.

Velocity spectra ($v_{||}$ inside angular acceptance of FRS)

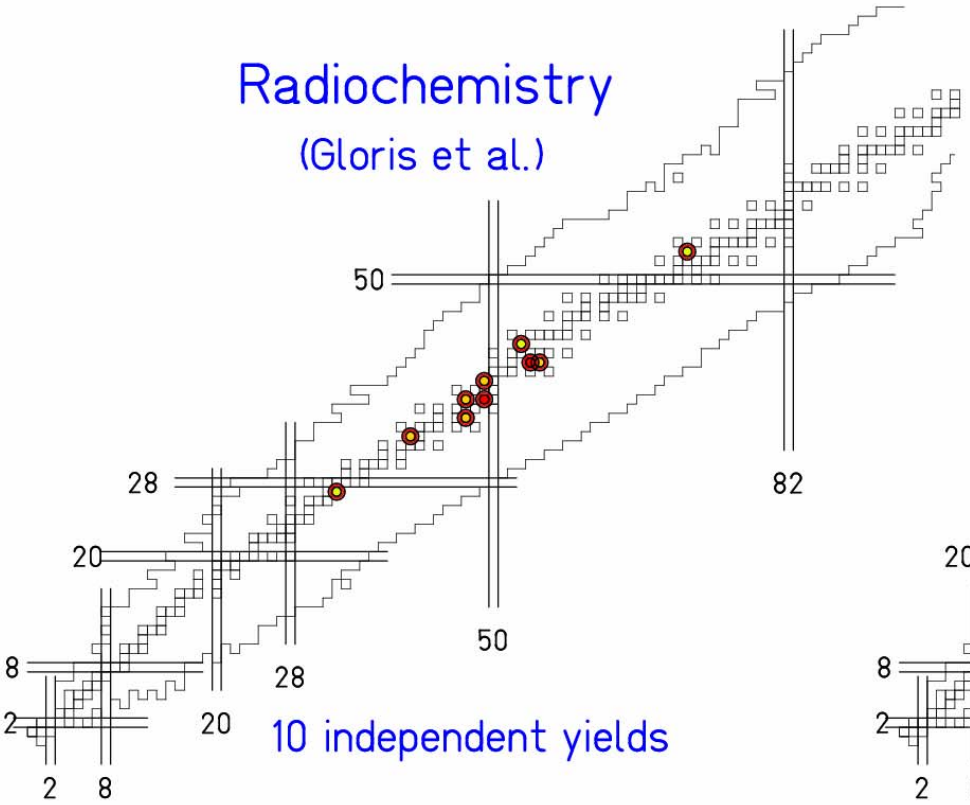


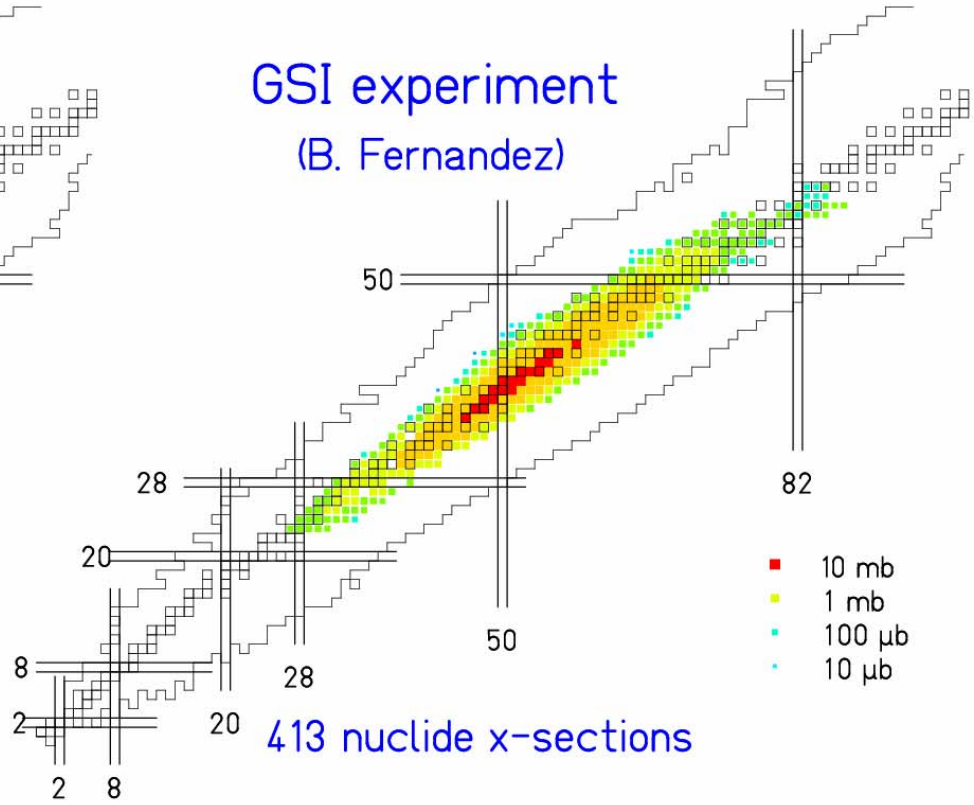
^{238}U + p, 1 A GeV



Experimental progress by inverse-kinematics method

Example: <u>Fission</u> of lead induced by ≈ 500 MeV protons

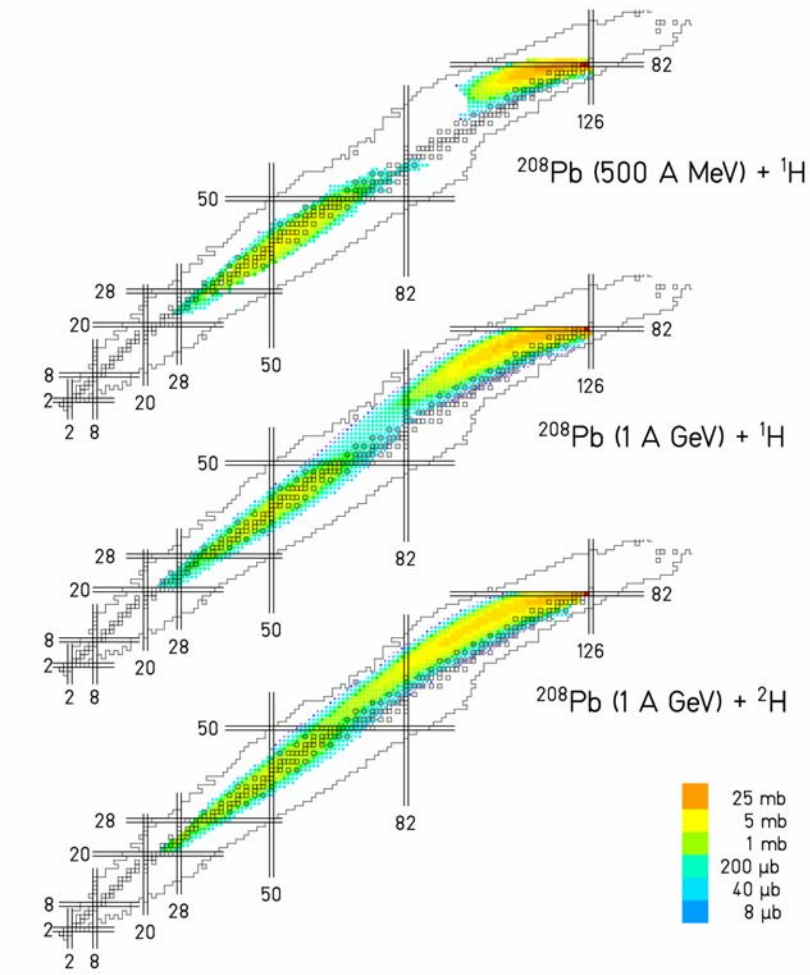




protons (553 MeV) on lead

^{208}Pb (500 A MeV) on hydrogen

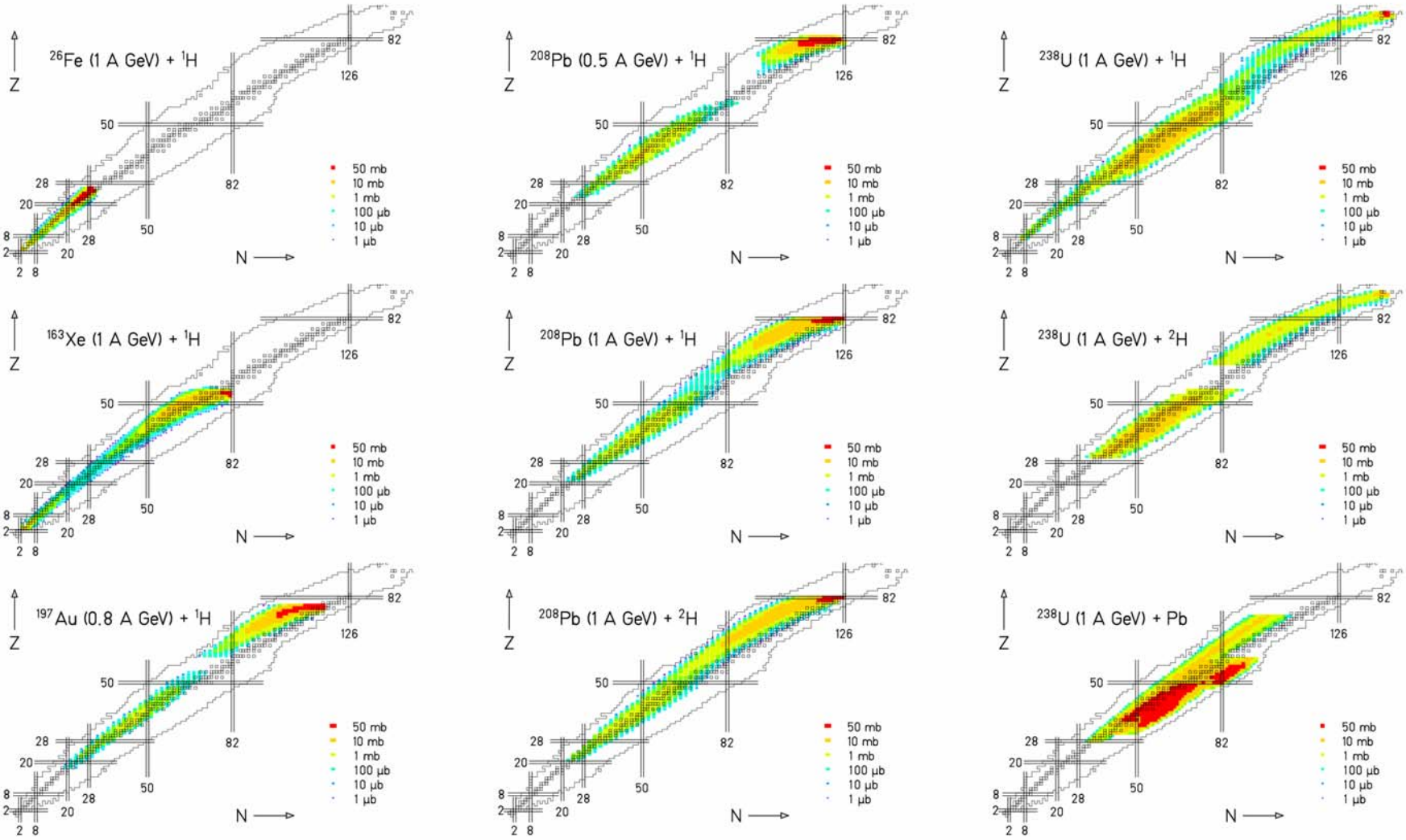
Spallation – energy dependence



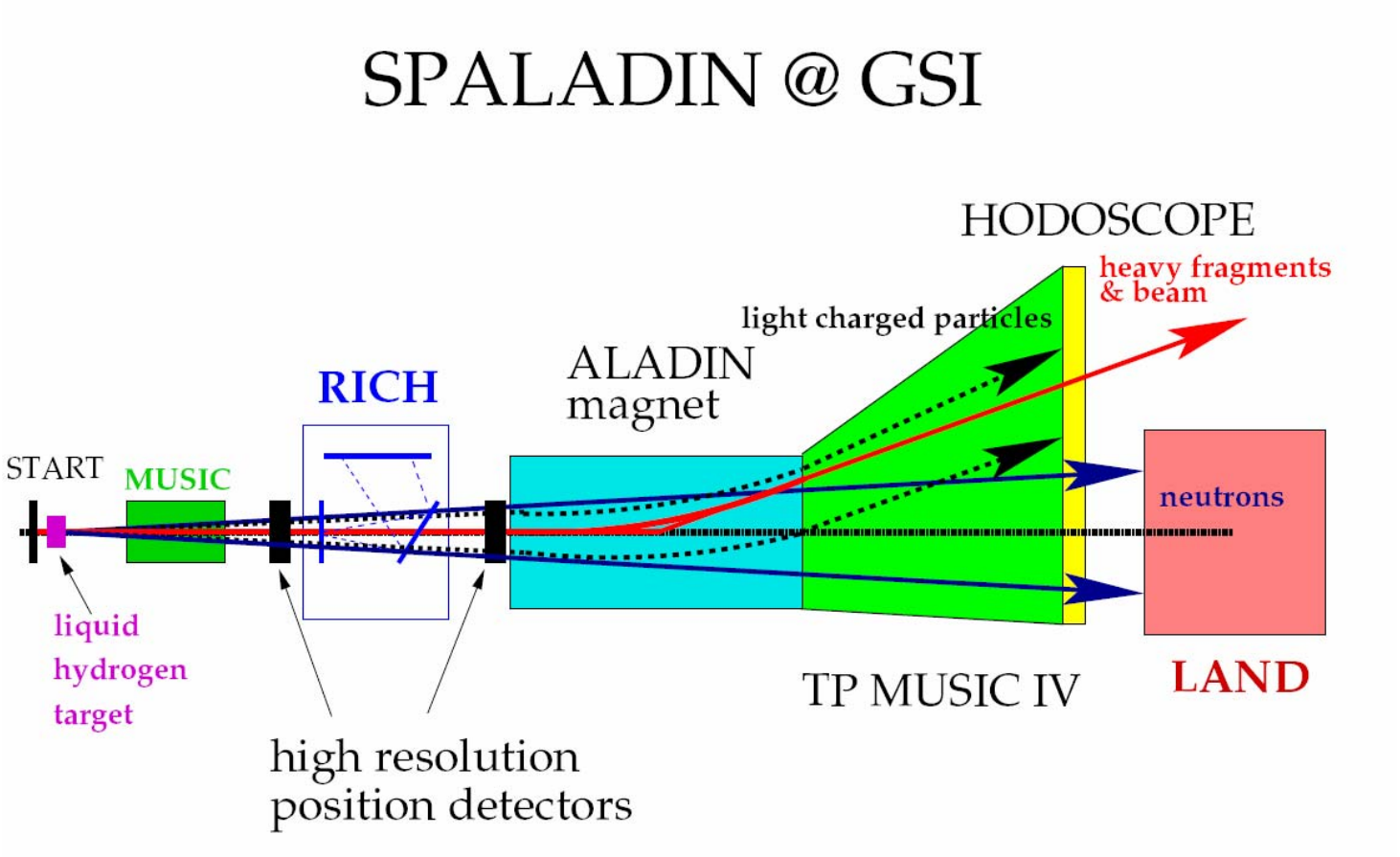
Region on the chart of the nuclides extends with increasing energy available in the system.

Experiments performed at GSI

Some systems investigated at GSI (7732 data points)

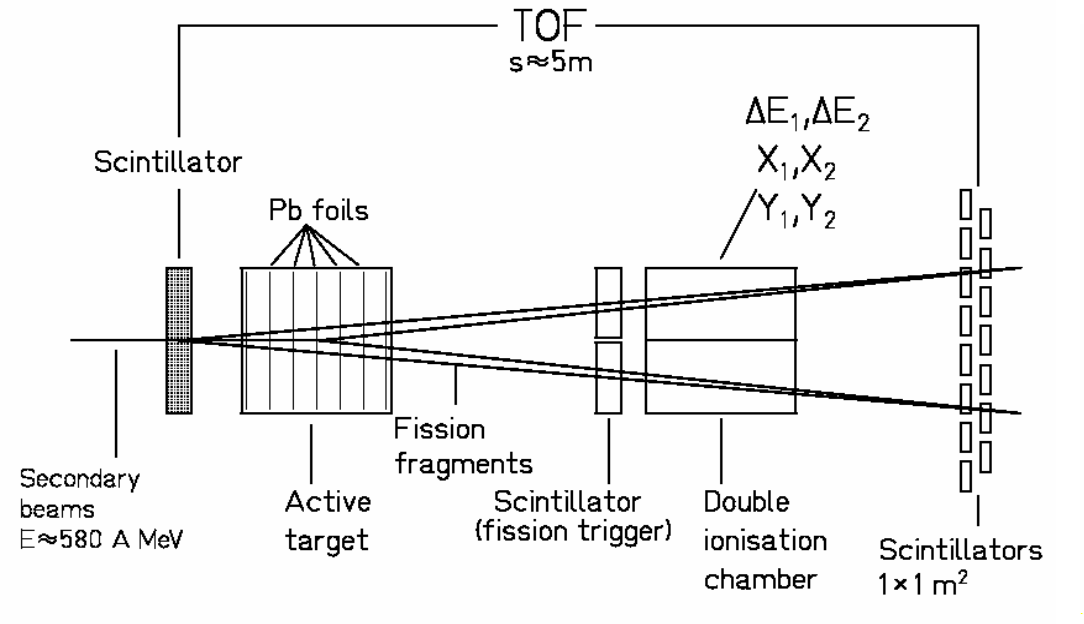


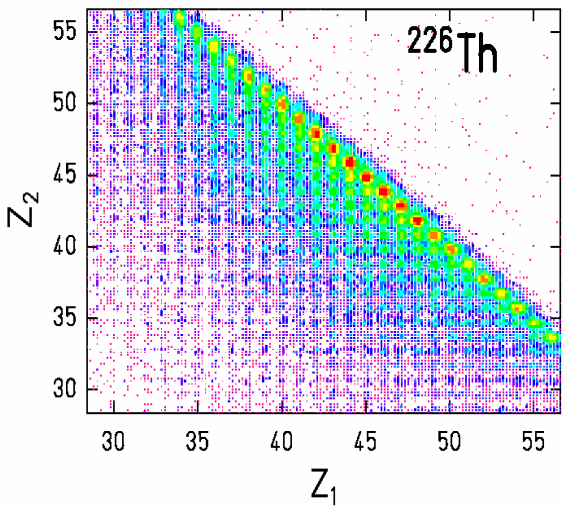
New approach 1: Large-acceptance spectrometer



Detection of neutrons, charged particles and heavy residues in coincidence but limited mass resolution

New approach 2: Double ionisation chamber – ToF wall





Counting projectiles and fission fragments with full coverage:
Developed to measure Z distributions.
Approach to measure also total fission cross sections with high accuray.

The detectors

Identification of both fragments in Z

High-energy nuclear data - Important facts
1. Neutrons

- **Production of neutrons**
  - Strongly forward focussed high-energy neutrons from INC stage (up to E_{beam}).
  - Slightly forward directed, with rather high-energies (more than thermal) from pre-equilibrium decay.
  - Isotropic component, Maxwellian energy spectrum from evaporation.
- **Detection of neutrons**
  - Total yield (also from thick targets) with neutron ball.
    (Precision ≈ 10 % due to efficiency correction.)
  - Double-differential cross sections ($d^2\sigma/dEd\theta$) with kinematical detectors.
- **Behaviour**
  - High-energy neutrons (E > 1 MeV) interact with nuclei with the geometrical cross section.
  - No interaction with electrons (in contrast to protons) → Absorption length 20 cm in lead (independent of energy above the resonance region.
  - Low-energy neutrons (E < 1 MeV) absorption (resonances) or elastic scattering (diffusion)
- **Importance**
  - Neutrons are the main source of secondary reactions in thick targets.

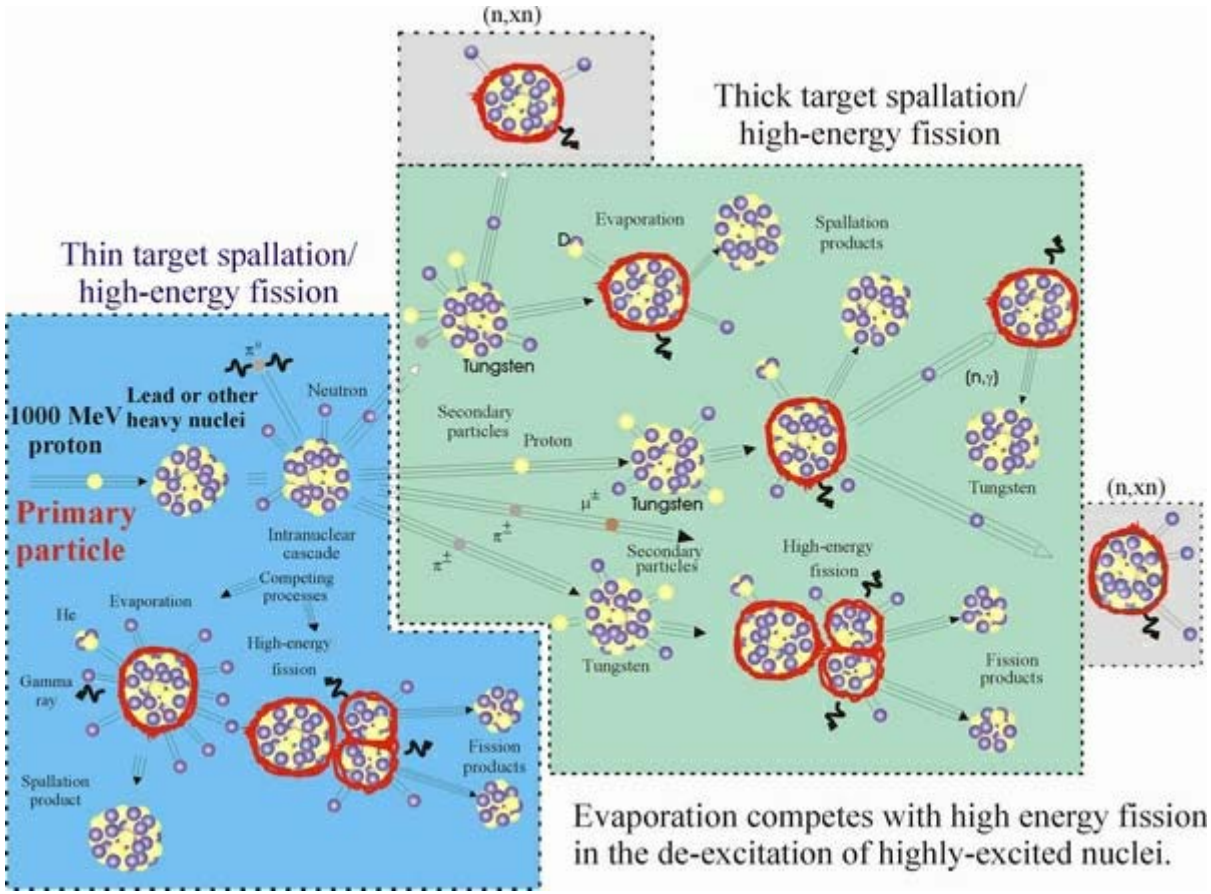
High-energy nuclear data - Important facts
2. Protons

- **Production of protons**
  - Strongly forward focussed high-energy protons from INC stage (up to E_{beam}) – like neutrons.
  - Slightly forward directed, with rather high-energies (more than thermal) from pre-equilibrium decay – similar to neutrons.
  - Isotropic component, Maxwellian-like energy spectrum from evaporation – less frequent and higher energies than neutrons due to Coulomb barrier.
- **Detection of protons**
  - Double-differential cross sections ($d^2\sigma/dEd\theta$) with kinematical detectors.
- **Behaviour**
  - High-energy protons (E > 10 MeV) interact with nuclei with the geometrical cross section.
  - Small range values (15 mm at 100 MeV, 0.15 mm at 10 MeV) due to interactions with electrons.
- **Importance**
  - Protons are a minor source of secondary reactions in thick targets.

High-energy nuclear data - Important facts
3. Heavy residues

- **Production of heavy residues**
  - Production in INC stage weak, mostly nucleons (limited to A < 5).
  - Production in -equilibrium decay weak, mostly nucleons.
  - Production of IMFs by multifragmentation.
  - Production of heavy residues close to the target nucleus by spallation evaporation.
  - Production of mid-mass nuclei from fission (strong for actinides).
- **Detection of heavy residues**
  - Normal kinematics: activation and gamma spectroscopy / accelerator mass spectrometry → only few "independent yiels" (before beta decay) – systematic excitation functions.
  - Inverse kinematics: In-flight identification of all residues before beta decay.
- **Behaviour**
  - Very low energies – very small ranges – no secondary reactions.
- **Importance**
  - Production of many radioactive species → activation – shielding.
  - Main source of DPAs → radiation damages of construction material.
  - Production of gases → damage of construction material.
  - Production of wide range of elements – possible importance for catalytic reactions.

Processes in a thick target - Internuclear cascade

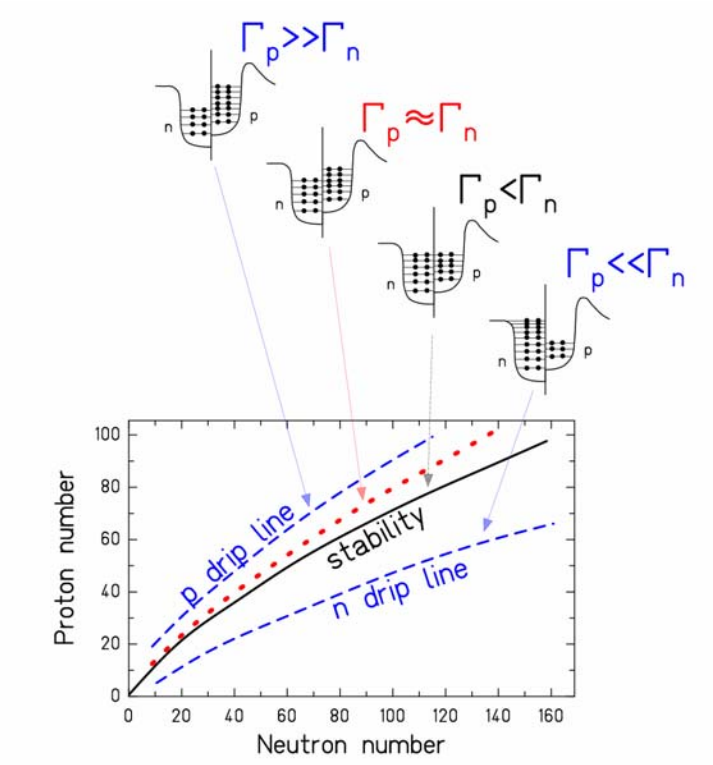


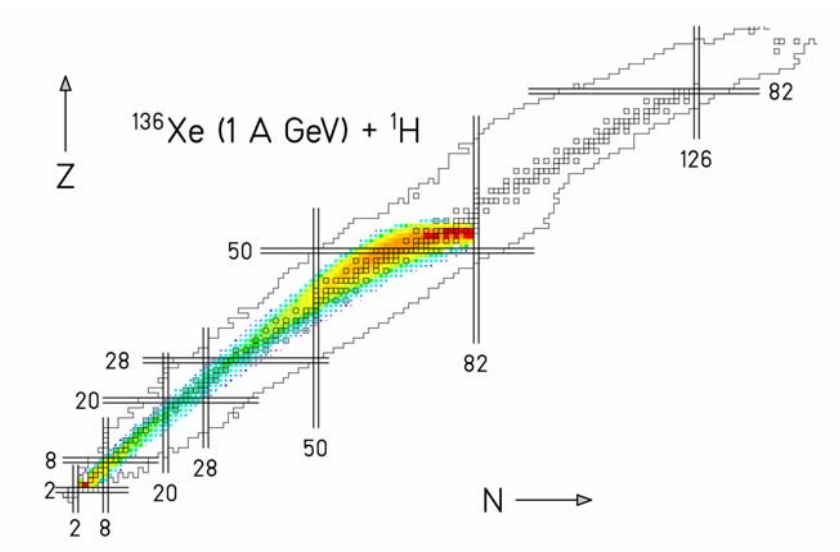
Evaporation competes with high energy fission in the de-excitation of highly-excited nuclei.

Supplement

Detailed considerations on the production of heavy residues

Evaporation process – the evaporation corridor

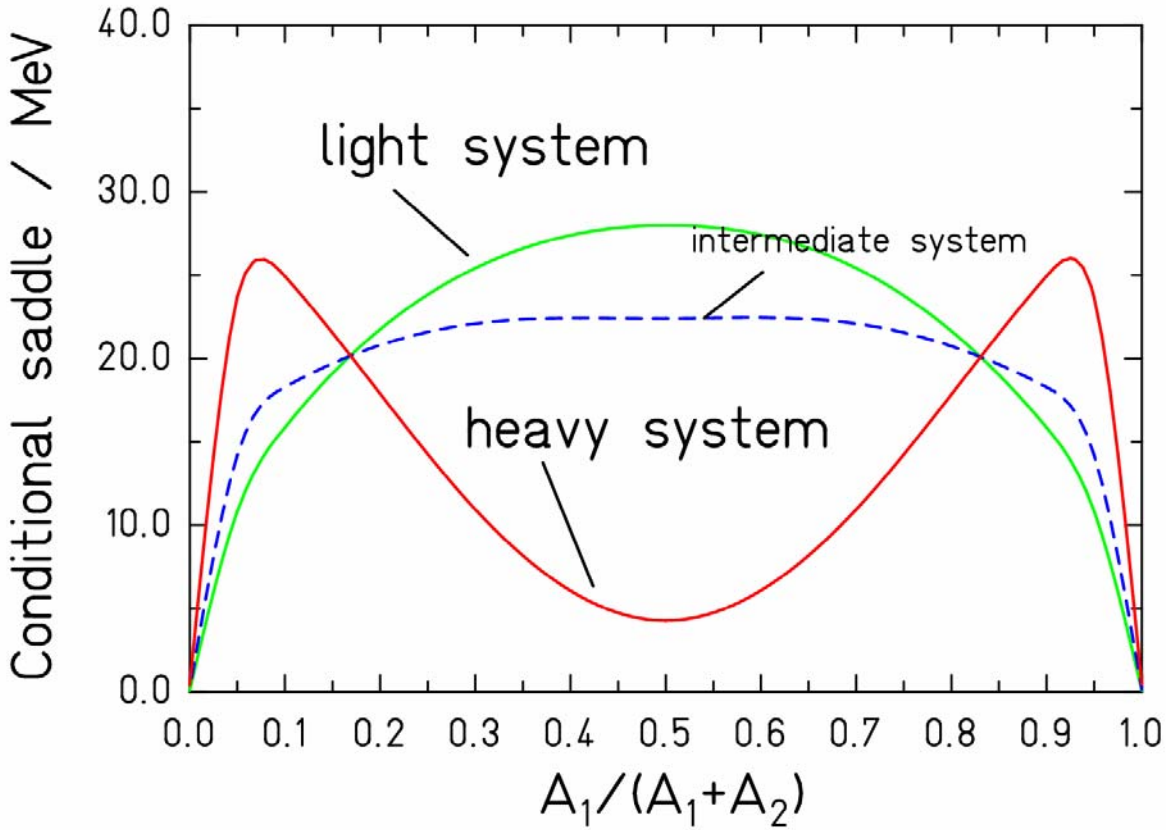




Evaporation is like a diffusion process on the chart of the nuclides.
→ Residues concentrate on an "evaporation corridor".

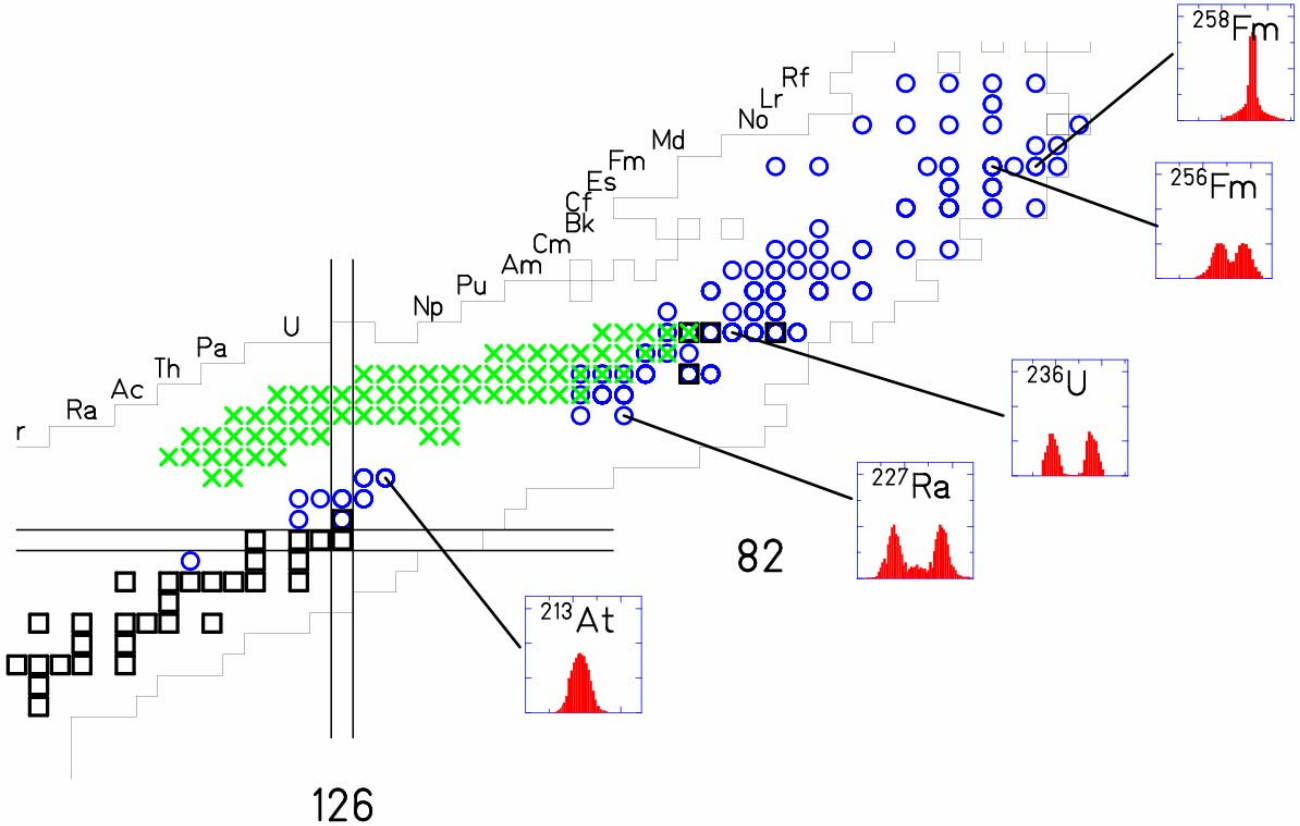
Proton evaporation is hindered by the Coulomb barrier.
→ The evaporation corridor is neutron-deficient.

Fission, macroscopic features



Fission barrier decreases with increasing Coulomb force for heavy nuclei.
Symmetric fission favoured for $Z^2/A > 22$ (Z around 50), disregarding shell effects.

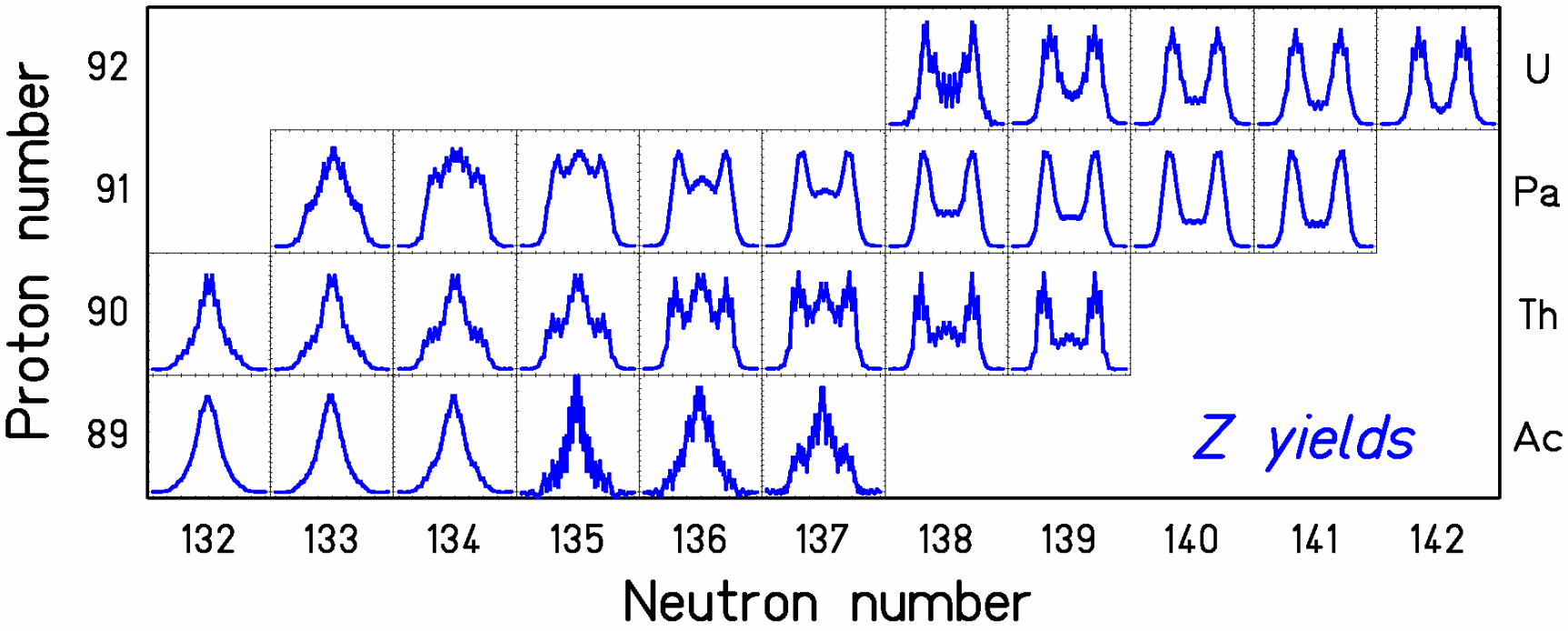
General features of the mass division in low-energy fission



o: Mass distributions, x: Z distributions from GSI experiment

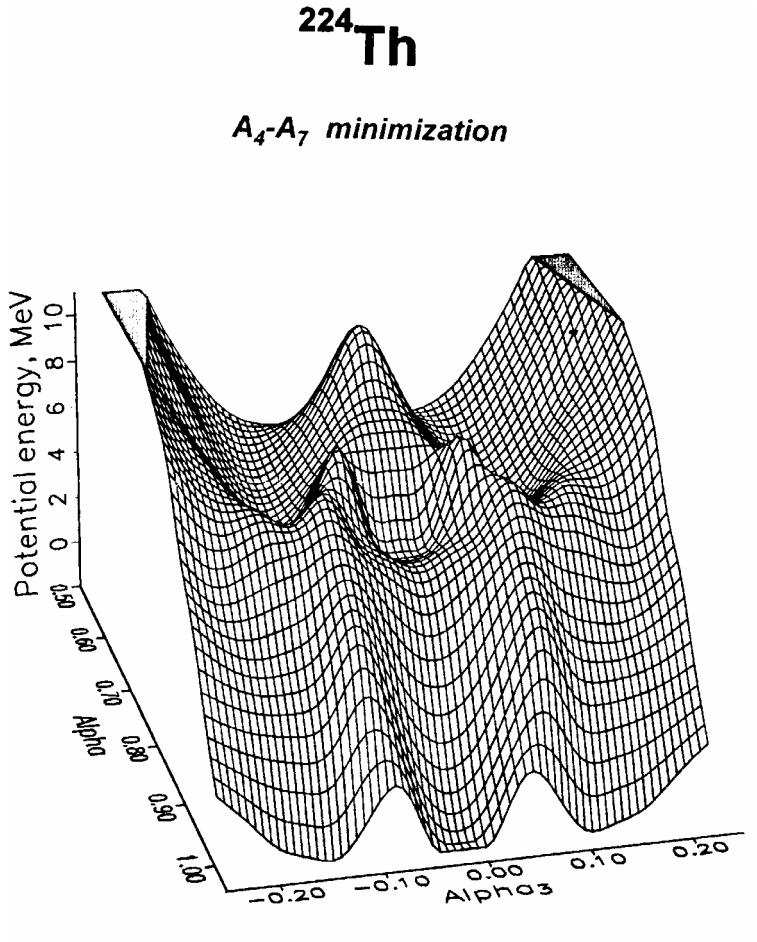
Compilation of general experimental knowledge on multi-model fission.

Systematic measurement of Z distributions with secondary beams (GSI experiment)



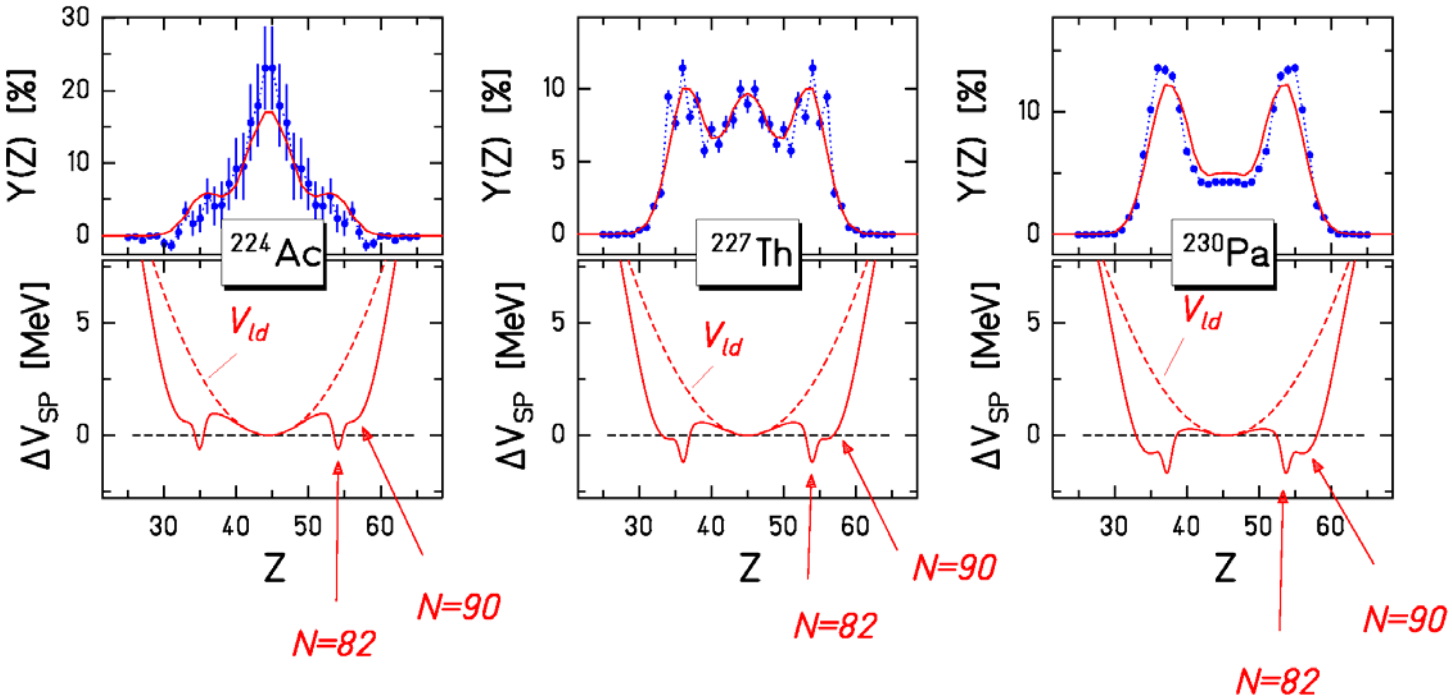
Fission induced by electromagnetic excitations, E* ≈ 11 MeV

Potential energy landscape on fission path



Strutinsky-type calculation of Pashkevich

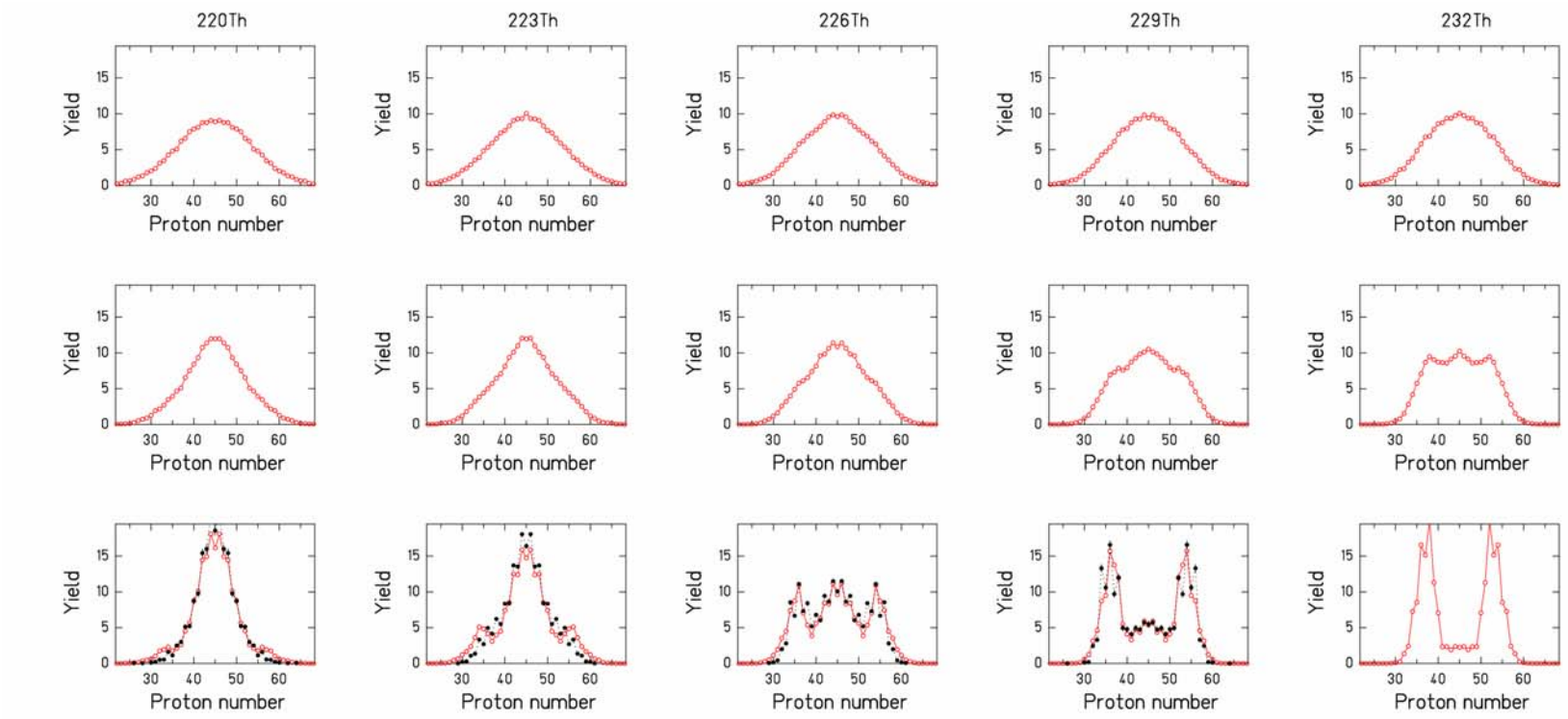
Modelling of multi-modal fission in ABLA



Transition from single-humped to double-humped distributions investigated experimentally and modelled by macroscopic (CN) and microscopic (nascent fragments) properties of the potential-energy landscape near outer saddle

Essential ingredient: Vanishing of shell effects with increasing excitation energy.

Multimodal fission around A = 226



E = 60 MeV

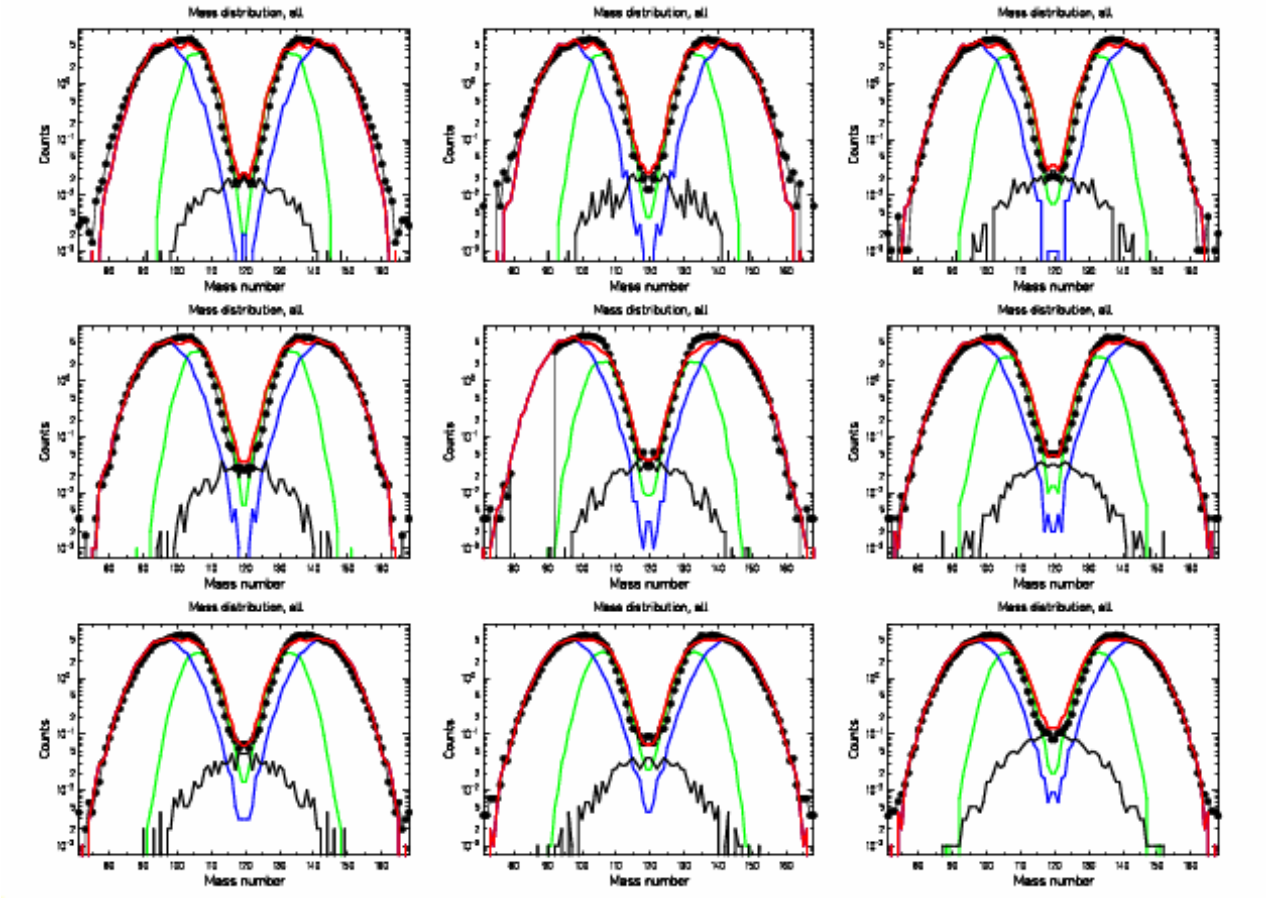
E = 20 MeV

E = 10 MeV

black: experiment

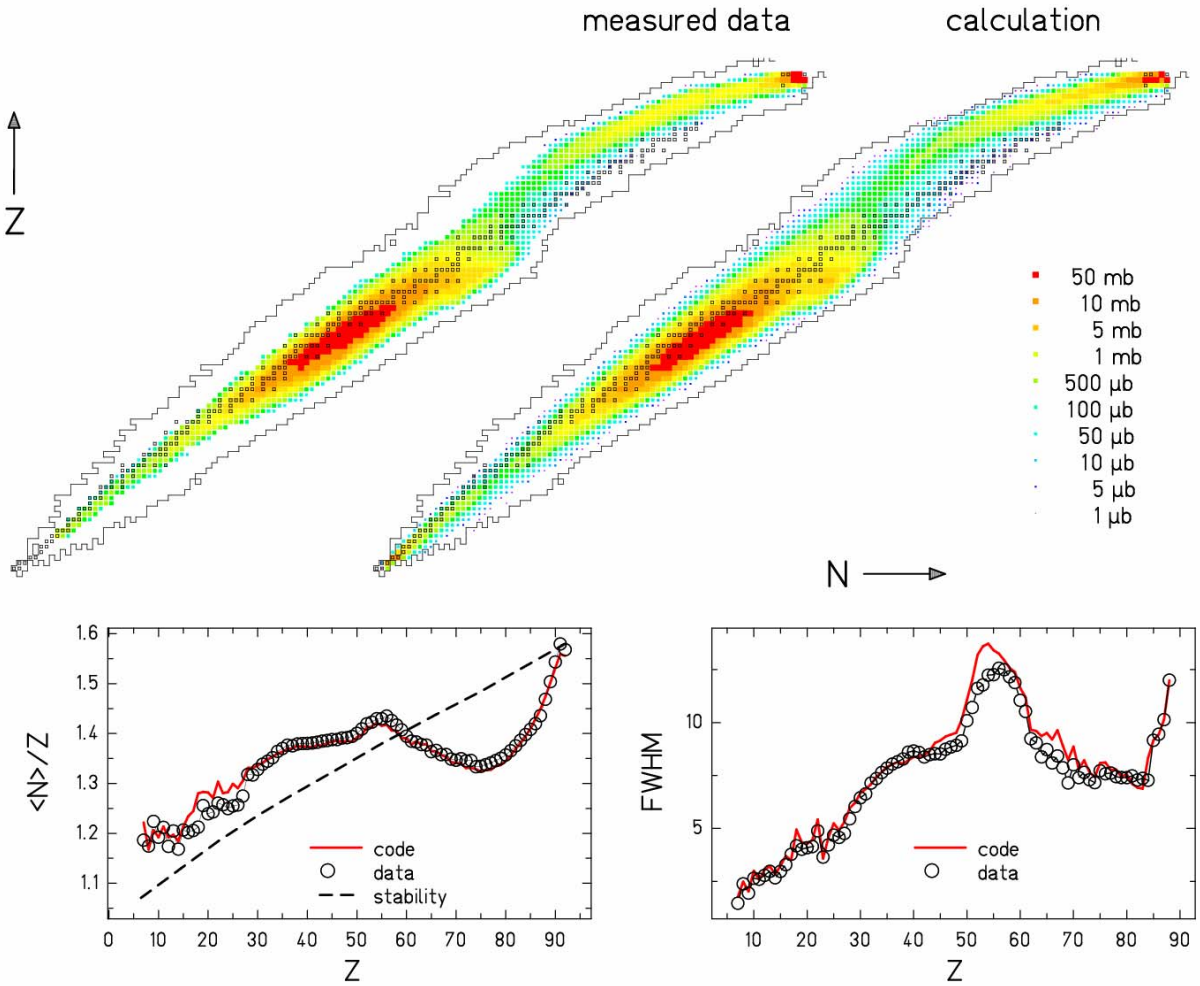
red: calculations with ABRABLA

$^{238}U + n, E_n = 1.2$ to 5.8 MeV



Data (Vives et al) compared to ABLA

Development of realistic codes (example: ^{238}U + p, 1 GeV)



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