Interesting effects
in well-known reactions and decays

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NEUTRONS FOR SOCIETY

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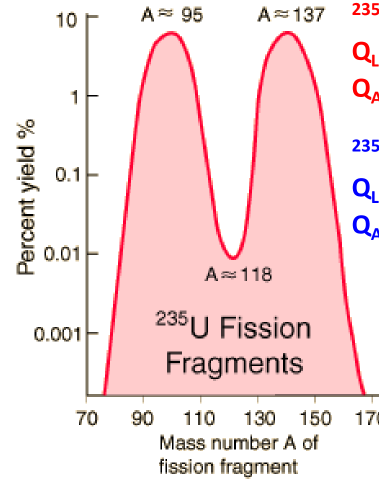
Mass yields of fission fragments



$^{235}U + n \rightarrow {}^{118}Pd + {}^{118}Pd$

Q_{LDM} = +184 MeV

Q_{AME} = +200 MeV

$^{235}U + n \rightarrow {}^{138}Xe + {}^{98}Sr$

Q_{LDM} = +176 MeV

Q_{AME} = +196 MeV

T.G. Wang et al.
J Fluid Mech
354, 43 (1994)
STS-50 flight

Thomson 1910: parabola mass spectrograph

Electric field parallel to magnetic field

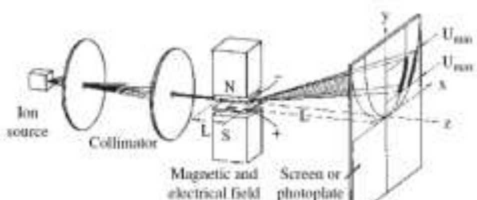


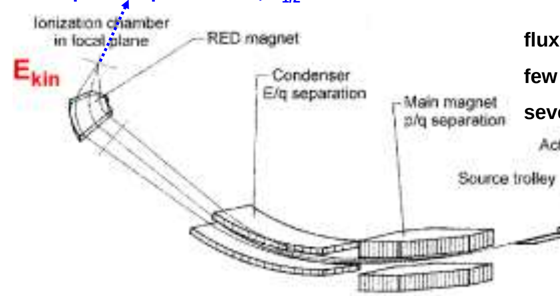
Figure 1.5 Parabola mass spectrograph constructed by J.J. Thomson (1910) with a discharge tube as ion source, a superimposed electrical field and a magnetic field oriented parallel to it for ion separation, and a photoplate for ion detection. (H. Kienitz (ed.), Massenspektrometrie (1968), Verlag Chemie, Weinheim. Reproduced by permission of Wiley-VCH.)

1912: Neon consists of two isotopes with mass 20 and 22

The LOHENGRIN fission fragment separator

mass-separated fission fragments,
up to 10^5 per second, $T_{1/2}$ ≥ microsec.

$\Delta A/A$ = 3E-4 – 3E-3
$\Delta E/E$ = 1E-3 – 1E-2



flux $5\cdot10^{14}$ n./cm²/s

few mg fission target

several 10^{12} fissions/s

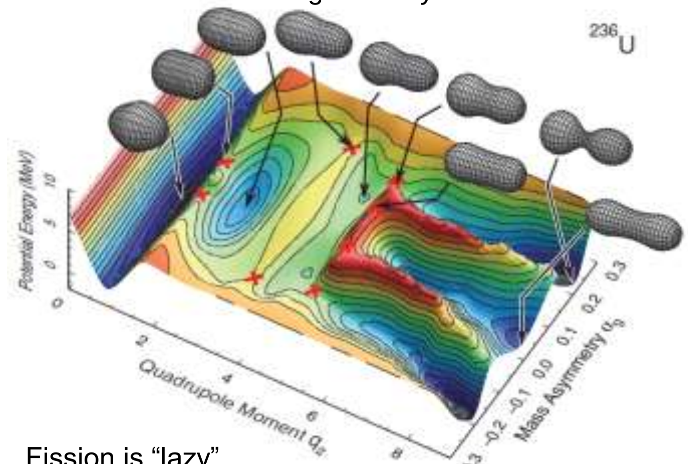
$m v^2 / r_{el} = q E$ $\quad$ $m v^2 / r_{magn} = q v B$

$E_{kin} / q = E / 2 r_{el}$ $\quad$ $m v / q = B r_{magn}$

P. Armbruster et al., Nucl. Instr. Meth. 139 (1976) 213.

The LOHENGRIN fission fragment spectrometer

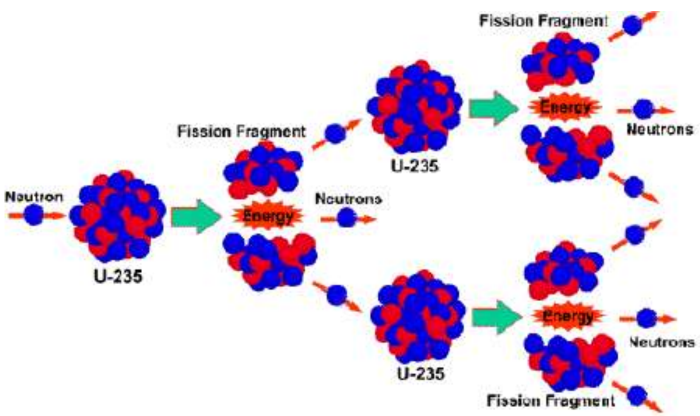
Understanding fission yields of ^{236}U



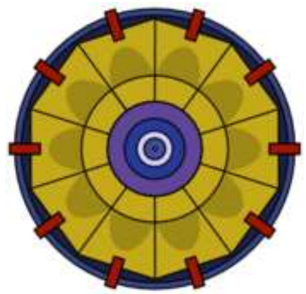
Fission is “lazy”

T. Ichikawa et al. PRC 86, 024610 (2012)

A nuclear chain reaction



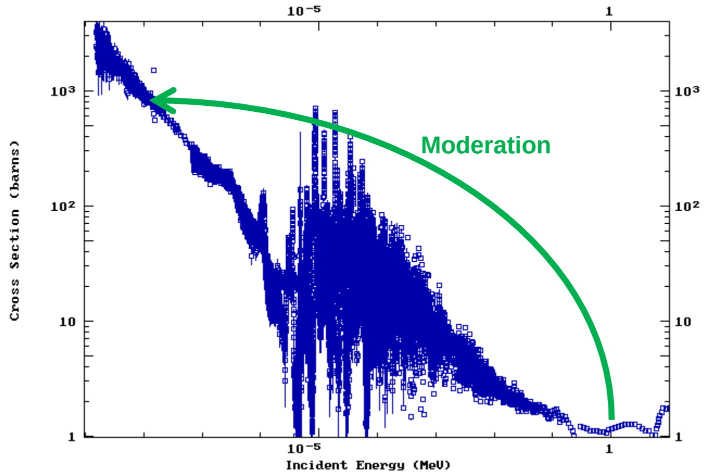
A single-pulse neutron source



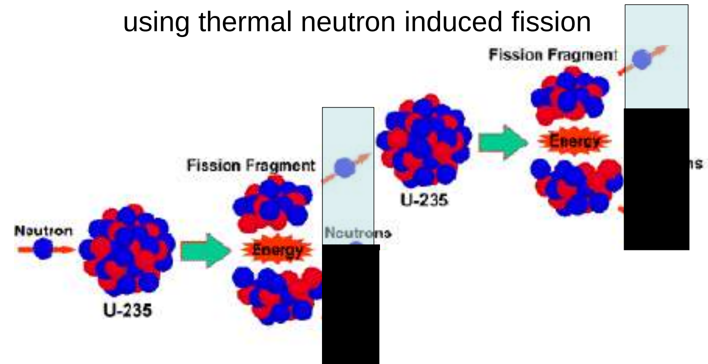
Uncontrolled chain reaction of fast-neutron induced fission

≈25 kg of 93% ^{235}U

^{235}U(n,f) cross-section as function of energy



A controlled nuclear chain reaction using thermal neutron induced fission



1. Moderate neutrons
2. Control neutron losses

k = multiplication factor = (neutrons produced in one generation) / (neutrons produced in previous generation)

Prompt neutron kinetics

Prompt neutron lifetime τ_p is the average time between the birth of prompt fission neutrons and their final absorption.

Assumptions:
-No delayed neutrons
-Infinite reactor, multiplication factor $k_\infty = k$

time	N(t)
0	n
τ_p	kn
$2\tau_p$	k^2n
$3\tau_p$	k^3n

$$\frac{dn}{dt} = \frac{k-1}{\tau_p} n \Rightarrow n(t) = n(0) e^{\frac{(k-1)}{\tau_p} t}$$

Time constant $T = \dfrac{\tau_p}{k-1}$

Exponential decrease (k<1)
or exponential growth (k>1)

cf. demographic projections for Germany
Fertility: 1.5 child/women -> k=0.75
T=25 years / (1-0.75) =100 years

Prompt neutron kinetics

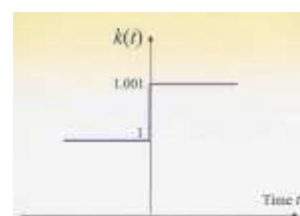
$\tau_p = \tau_s + \tau_d$ = slowing down time + diffusion time

In thermal reactors: $\tau_s << \tau_d$, i.e. $\tau_p \cong \tau_d$

$\tau_d \cong \lambda_a / v \cong$ 10 cm / (2000m/s)

$\tau_p \cong \tau_d \cong 50$ µsec

Example: step of reactivity from k=1.000 to k=1.001



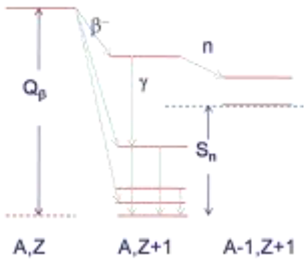
$$T = \frac{\tau_p}{k-1} = \frac{50.10^{-6}}{10^{-3}} = 0.05\text{sec}$$

$$n(t) = n_0 e^{\frac{t}{0.05}}$$

$$\frac{n(1\text{sec})}{n_0} = e^{20} = 5E8$$

"Prompt" control is not possible!

Delayed neutron emission from fission products



Possible precursors
^{87}Br
^{88}Br, ^{137}I, ^{136}Te, ^{134}Sb, ^{141}Cs
^{89}Br, ^{138}I, ^{92,93}Rb, ^{147}La, ^{87}Se, ^{84}As
^{85}As, ^{90}Br, ^{135}Sb, ^{94}Rb, ^{139}I, ^{98,99}Y, ^{142}Cs, ^{80}Ga
^{86,87}As, ^{136}Sb, ^{147,148}Ba, ^{81,82}Ga, ^{140,141}I, ^{92}Br, ^{136}Sn, ^{143}Cs, ^{89}Se
^{83}Ga, ^{146,147}Cs, ^{95,96,97,98,99}Rb, ^{92}Br, ^{91}Se

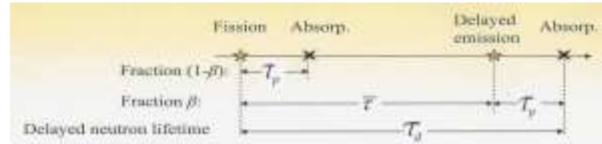
β= percentage of delayed neutrons

^{235}U	β	ν	ν_p	ν_d
Ther	0.0067	2.490	2.473	0.01668

average emission time

$$\tau_{delayed} = \frac{1}{\beta}\sum_i \beta_i \tau_i \approx 12\,\text{sec}$$

Neutron lifetime, taking into account delayed neutrons



$$k = k_{prompt} + k_{delayed} = 1 = (1-\beta) + \beta$$

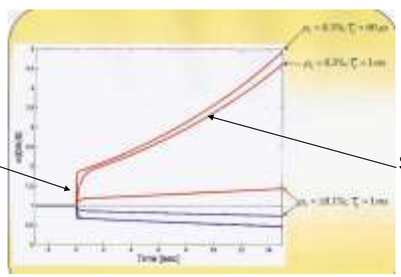
$$\tau = (1-\beta)\tau_p + \beta(\tau_{delayed} + \tau_p) \approx \beta\tau_{delayed} = 0.08\,\text{sec}$$

Now for step from k=1.000 to k=1.001

$T=\beta\tau_{delayed}/(k-1)$= 80 seconds

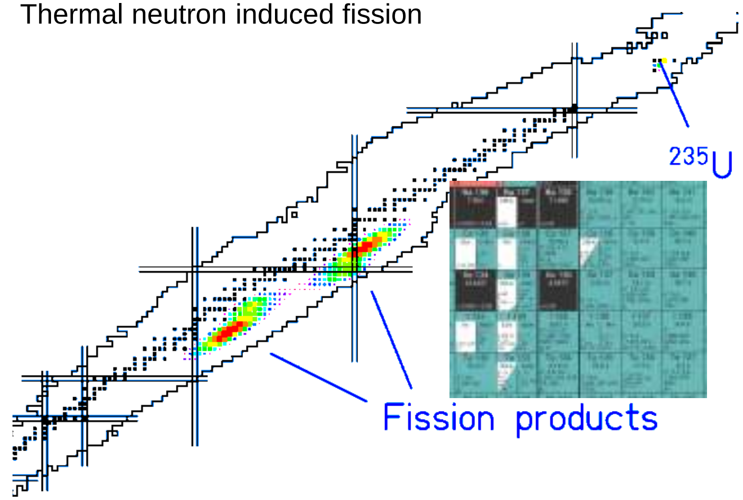
Reactor response to a step of reactivity

Prompt jump

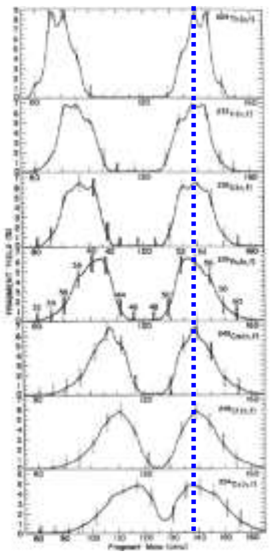


Slow increase due to Delayed neutrons

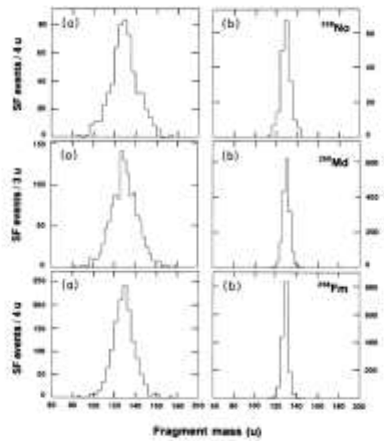
Thermal neutron induced fission



Interesting effects in well-known reactions and decays



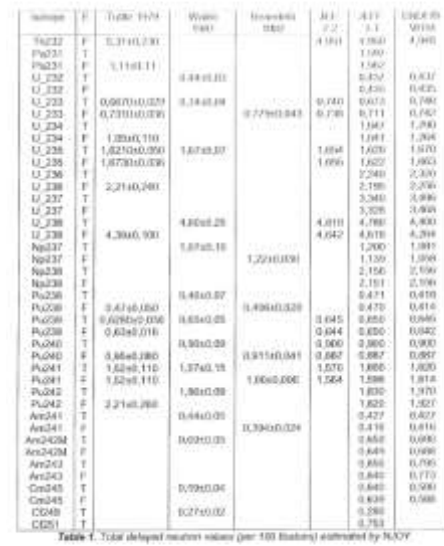
J.P. Unik et al. IAEA (1974)



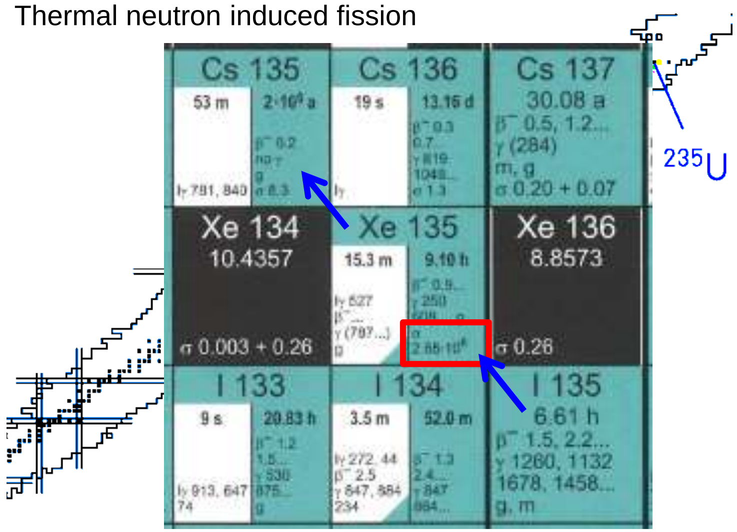
E.K. Hulet et al. PRL 56, 313 (1986)
E.K. Hulet et al. PRC 40, 770 (1989)

Delayed neutron yields

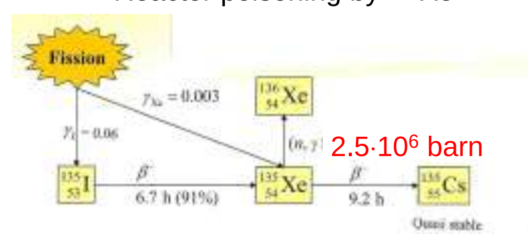
^{235}U(n_{th},f)	1.62%
^{239}Pu(n_{th},f)	0.63%
^{238}U(n_f,f)	4.39%
^{239}Pu(n_f,f)	0.63%



Thermal neutron induced fission



Reactor poisoning by ^{135}Xe



$$\frac{dN_I}{dt} = \gamma_I \Sigma_f \phi_{th} - \lambda_I N_I$$

$$N_I(t) = \frac{\gamma_I \Sigma_f \phi_{th}}{\lambda_I}\left(1 - e^{-\lambda_I t}\right) \to N_I^{eq} = \frac{\gamma_I \Sigma_f \phi_{th}}{\lambda_I}$$

$$\frac{dN_{Xe}}{dt} = \lambda_I N_I + Y_{Xe} \Sigma_f \Phi - \lambda_{Xe} N_{Xe} - \sigma_c^{Xe} N_{Xe} \Phi \quad \longrightarrow \quad N_{Xe}^{eq} = \frac{(Y_I + Y_{Xe}) \Sigma_f \Phi}{\lambda_{Xe} + \sigma_c^{Xe} \Phi}$$

Xenon equilibrium concentration

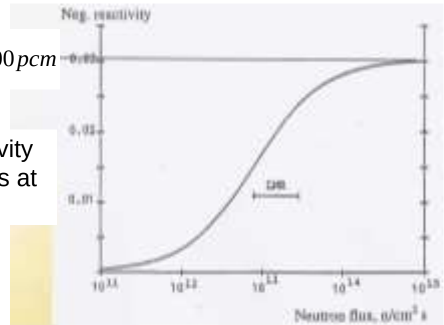
Reactivity due to Xe poisoning

$$\Delta\rho = -\frac{\Sigma_{xénon}}{\nu\Sigma_f} = -\frac{\sigma_X X}{\nu\Sigma_f} \longrightarrow \Delta\rho^{xénon} = -\frac{Y_T}{(\lambda_X+\sigma_X\Phi)}\frac{\Phi\sigma_X}{\nu}$$

High flux limit

$$\Delta\rho = -\frac{Y_T}{\nu} = -\frac{0.063}{2.45} \approx -2600\,pcm$$

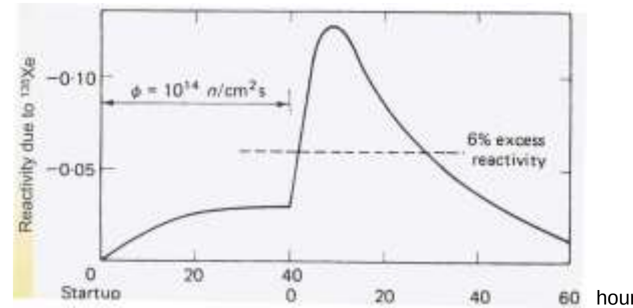
The maximum anti-reactivity is reached after 1 - 2 days at full power



Xenon transient following a shutdown

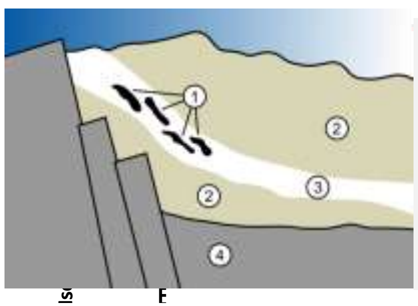
$$\frac{dN_{Xe}}{dt} = \lambda_I N_I + Y_{Xe}\Sigma_f\Phi - \lambda_{Xe}N_{Xe} - \sigma_c^{Xe}N_{Xe}\Phi$$

No more neutrons to destroy ^{135}Xe



The anti reactivity increase prevents restarting of the reactor if a large enough positive reactivity reserve is not available!

The first nuclear reactor



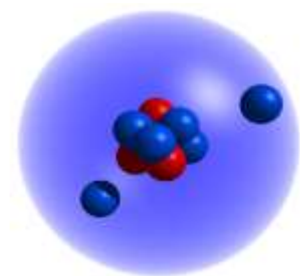
≈ 0.5 h on: boiling water
≈ 2.5 h off: cooling down

Operating self-regulated for few 100 ka

Fission products ("waste") did not move more than cm over 2 Ga

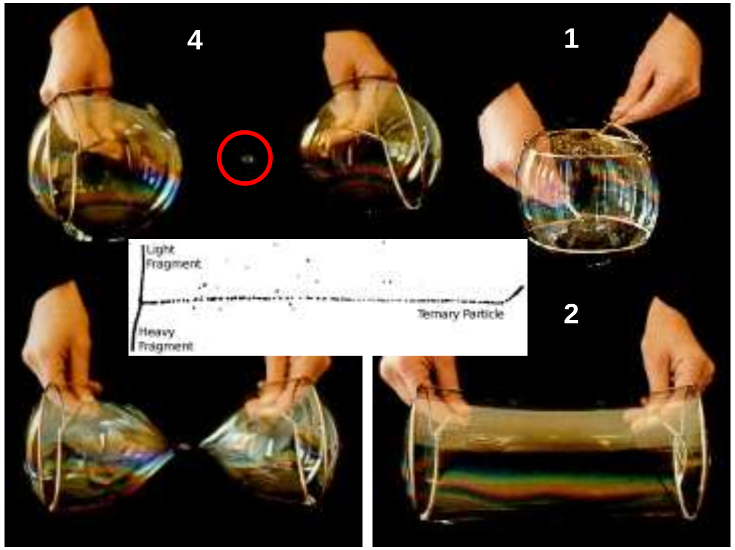
In nuclear reactors a NEGATIVE FEEDBACK is very POSITIVE (=good/useful)!

^{11}Li production in thermal neutron induced fission?



	^{235}U	+	n	⇒	^{11}Li	+	^{225}Ac
A	235	+	1	=	11	+	225
Z	92	+	0	=	3	+	89
ΔM (MeV)	40.919		8.071		40.728		21.639

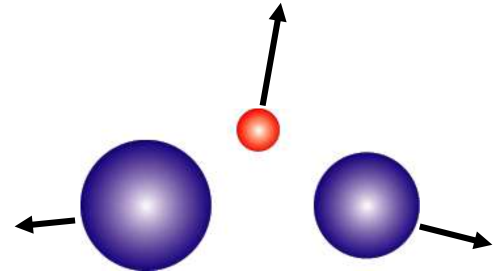
Q (MeV) = (40.919 + 8.071) – (40.728 + 21.639) = -13.4



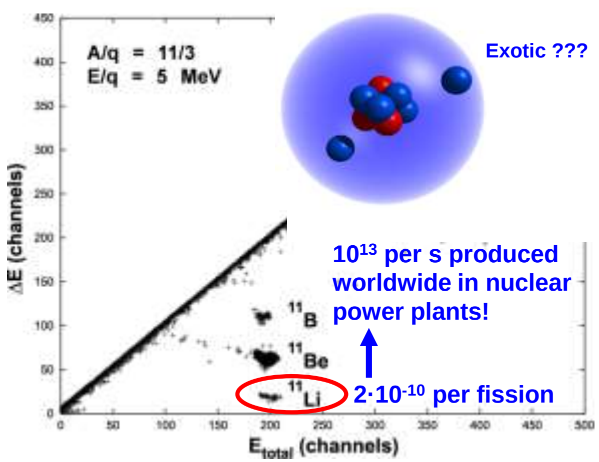
^{11}Li production in thermal neutron induced fission?

	^{235}U	+	n	⇒	^{11}Li	+	^{134}Te	+	^{91}Rb
A	235	+	1	=	11	+	134	+	91
Z	92	+	0	=	3	+	52	+	37
ΔM (MeV)	40.919		8.071		40.728		-82.536		-77.745

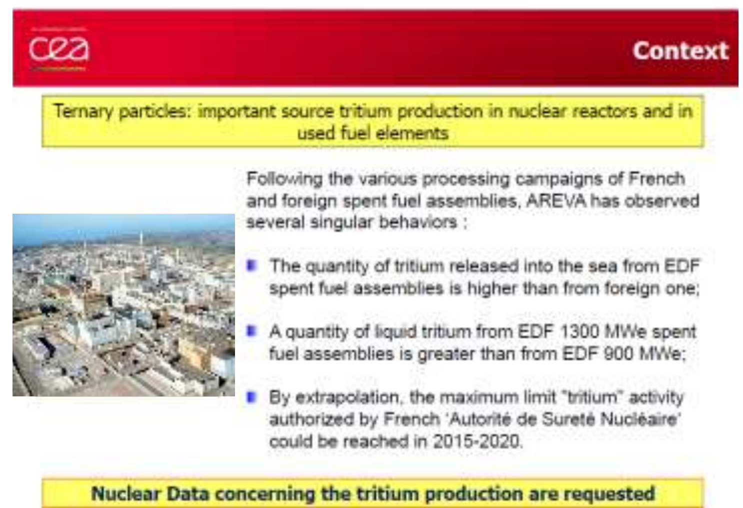
Q (MeV) = (40.919 + 8.071) – (40.728 – 82.536 – 77.745) = **+168.5**



Detection of rare ternary particles



Nucl. Phys. A652, 371.



cea Context

Ternary particles: important source tritium production in nuclear reactors and in used fuel elements

Following the various processing campaigns of French and foreign spent fuel assemblies, AREVA has observed several singular behaviors :

- The quantity of tritium released into the sea from EDF spent fuel assemblies is higher than from foreign one;
- A quantity of liquid tritium from EDF 1300 MWe spent fuel assemblies is greater than from EDF 900 MWe;
- By extrapolation, the maximum limit "tritium" activity authorized by French 'Autorité de Sureté Nucléaire' could be reached in 2015-2020.

Nuclear Data concerning the tritium production are requested

O. Serot. JEFF 2014.

Context

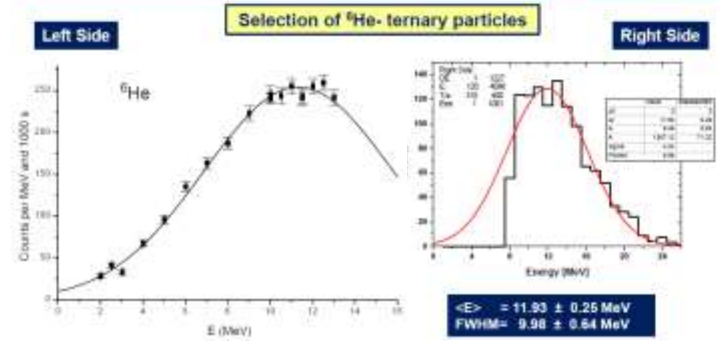
Calculations performed for UOx (3.25%) at 33GWd/t (J. Pavageau, private communication)

Tritium Formation in UOx fuel element	Contribution
Direct production from Ternary Fission	81.94 %
Indirect Production from ^{6}He Ternary Fission: • $^{6}He \rightarrow {}^{6}Li + \beta^-$ • $^{6}Li + n \rightarrow {}^{4}He + {}^{3}H$ (σ=941.3b)	11.50 %
Indirect ^{3}H production • $^{3}H \rightarrow {}^{3}He + \beta^-$ • $^{3}He + n \rightarrow p + {}^{3}H$ (σ=5317b)	4.82 %
Reaction (n,^{3}H) on ^{16}O: $n + {}^{16}O \rightarrow {}^{14}N + {}^{3}H$	0.96 %
Reaction (n,^{3}He) on ^{16}O: • $n + {}^{16}O \rightarrow {}^{14}C + {}^{3}He$ • $^{3}He + n \rightarrow p + {}^{3}H$	0.05 %

The accuracy of the tritium production could be improved by improving our knowledge on **^{6}He ternary fission yields**

O. Serot. JEFF 2014.

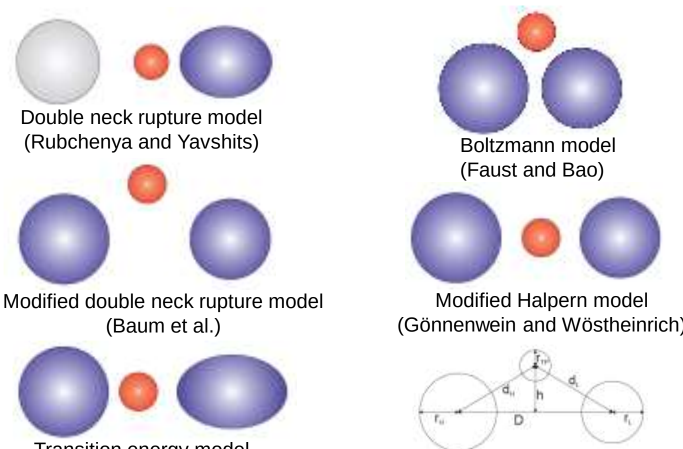
Measurement of ternary fission yields from $^{241}Pu(n_{th},f)$ reaction



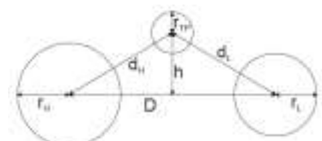
^{6}He ternary fission yield for ^{241}Pu(n,f): $(4.48 \pm 0.30) \times 10^{-5}$

O. Serot. JEFF 2014.

Ternary fission models

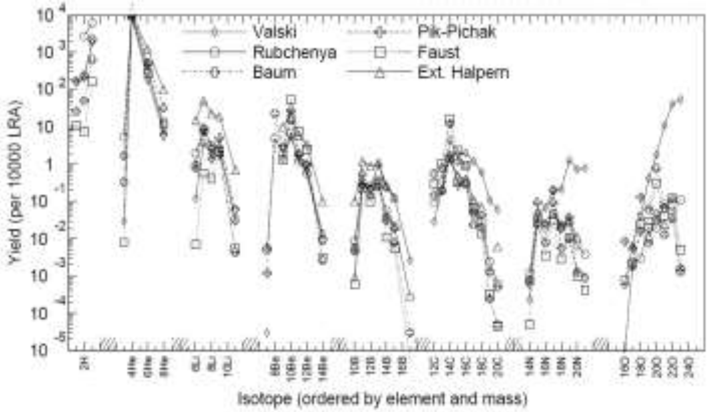


Ternary fission models

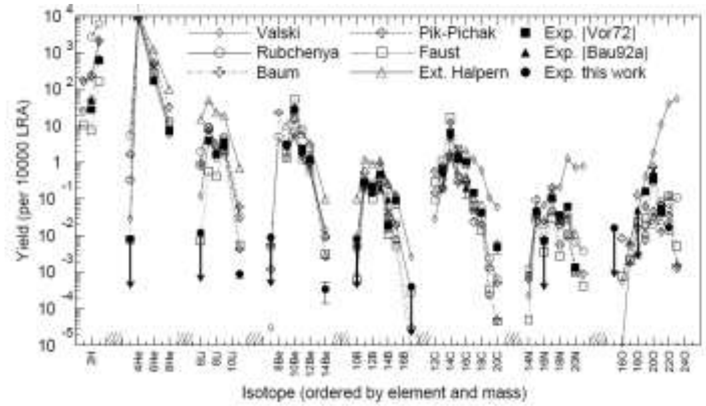


Model (ref.)	(A_H, Z_H)	(A_L, Z_L)	D fm	d_L fm	d_H fm	h fm	T MeV
[Rub88]	^{140}Cs	^{86}As$^{(11}$	≈ 25	11.2	≈ 13.8	0	2.1
[Rub94, Rubch]	^{132}Sn	^{94}Sr	23.6	11.3	12.3	0	2.19
[Bau92b]	^{132}Sn	^{94}Sr	22.6	13.6	14.2	7.5	1.91
[PP94]	^{132}Sn	^{94}Sr	22.9	12.8	10.1	0	2.1$^{(12}$
[Fau95b]	^{132}Sn	^{94}Sr	13.4	9.9	10.5	7.7	1.3
[Wös96]	^{132}Sn$^{(13}$	^{94}Sr	22.4	10.8	11.6	0	(3.1)$^{(14}$

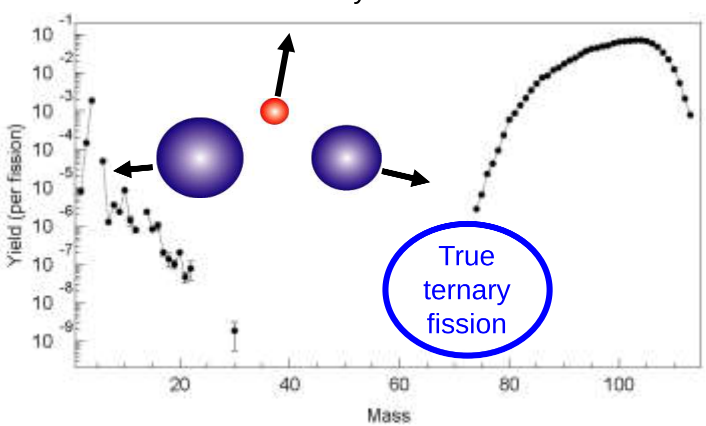
Ternary fission models



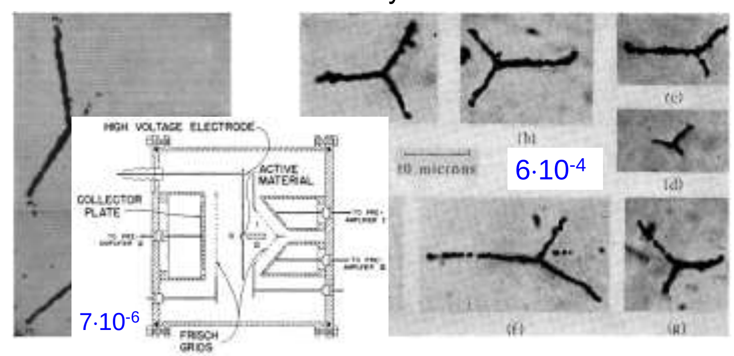
Comparison with measurement



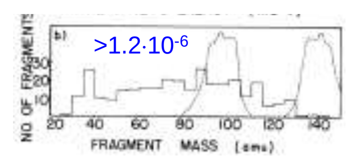
Ternary fission



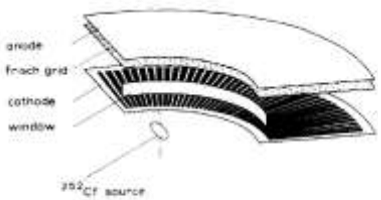
"True ternary fission"

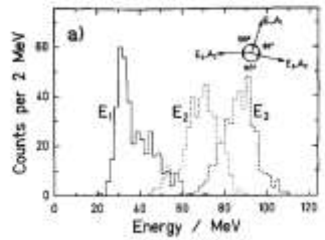


T. San-Tsiang et al. Nature 159, 773 (1947)
L. Rosen and A.M. Hudson, PR 78, 533 (1950)
M.L. Muga et al. PR 121, 270 (1961)
M.L. Muga, PRL 11, 129 (1963)



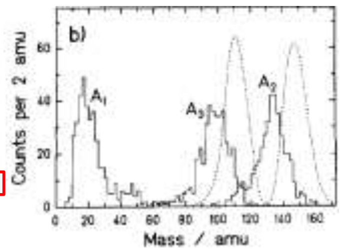
DIOGENES: upper limit for true ternary fission



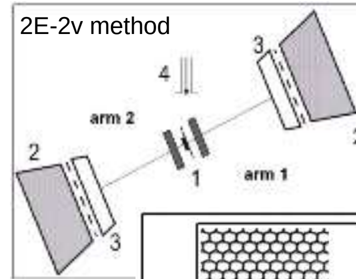


P. Schall et al. PLB 191, 339 (1987)

The mass spectrum (fig. 3b) does not exclude ternary fragmentations with lightest masses in the range $30 \leqslant A \leqslant 70$. These events are, however, preferably measured in those chamber sectors which face the Ni backing indicating an interference with fragment scattering on Ni. For a possible true ternary fission we deduce an upper limit of 8×10^{-8} per binary fission, which is about one order of magnitude lower than observed by Muga et al.

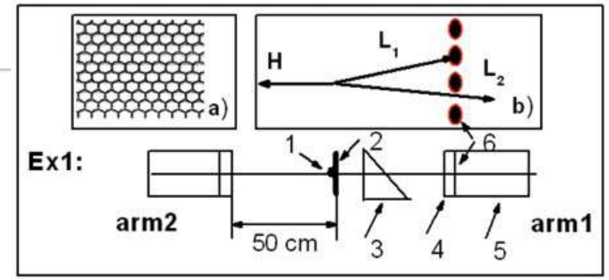


FOBOS

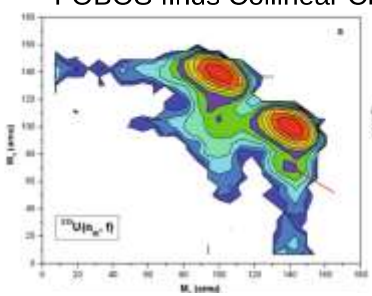


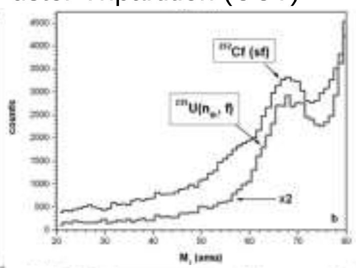
In the present work "true ternary fission" is discovered as collinear cluster decay. The discovery relies on the fact that two of the fragments emitted into the same direction become separated by an angle of ~1° on their way to the detector while passing through material. One fragment may then be lost by hitting a grid at the detector entry. In the experiments masses and velocities are

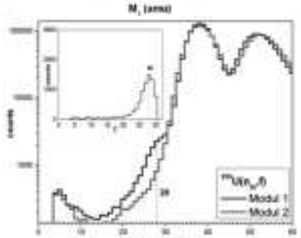
Yu.V. Pyatkov et al. EPJA 45, 29 (2010)
Yu.V. Pyatkov et al. EPN 41, 14 (2010)



FOBOS finds Collinear Cluster Tripartition (CCT)



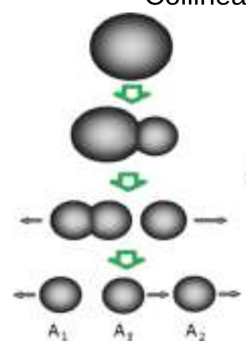


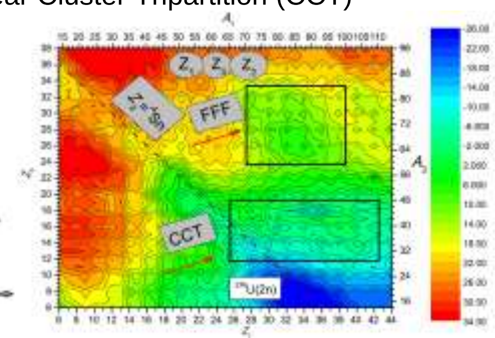


Ternary ≈^{70}Ni particles
0.51(4)% per binary fission

Yu.V. Pyatkov et al. EPJA 45, 29 (2010)
Yu.V. Pyatkov et al. EPN 41, 14 (2010)
Yu.V. Pyatkov et al. Phys At Nucl 73, 1309 (2010)
Yu.V. Pyatkov et al. EPJA 48, 94 (2012)

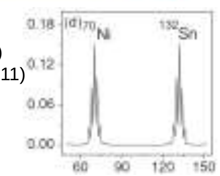
Collinear Cluster Tripartition (CCT)



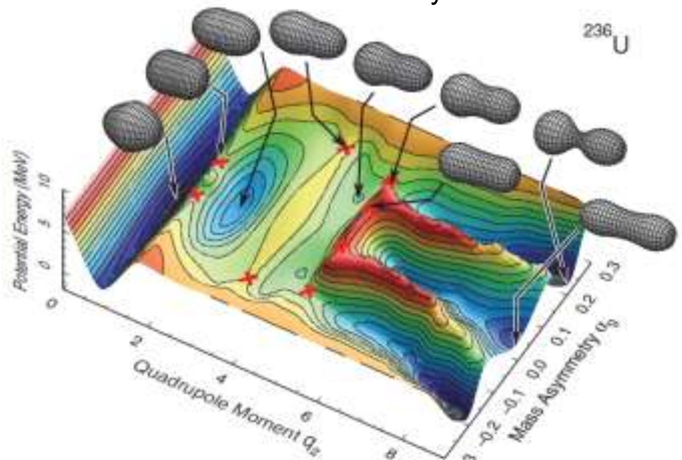


^{70}Ni + ^{34}Si + ^{132}Sn

K. Manimaran and M. Balasubramaniam. EPJA 45, 293 (2010)
K. Manimaran and M. Balasubramaniam. PRC 83, 034609 (2011)
R.B. Tashkhodjaev et al. EPJA 47, 136 (2011)
W. von Oertzen, A.K. Nasirov. PLB 734, 234 (2014)
K. R. Vijayaraghavan et al. PRC 91, 044616 (2015)
R. B. Tashkhodjaev et al. PRC 91, 054612 (2015)

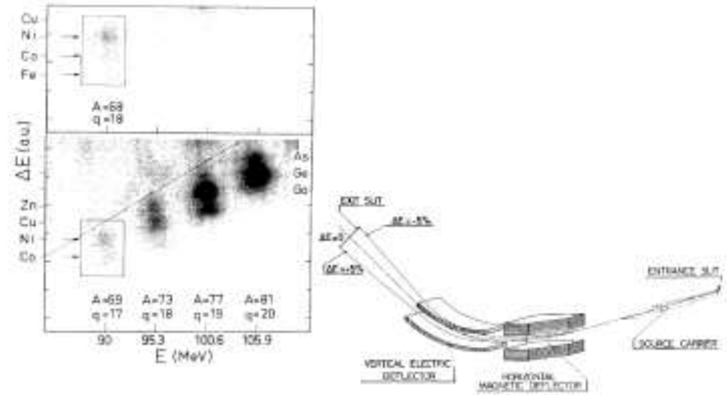


Fission is “lazy”



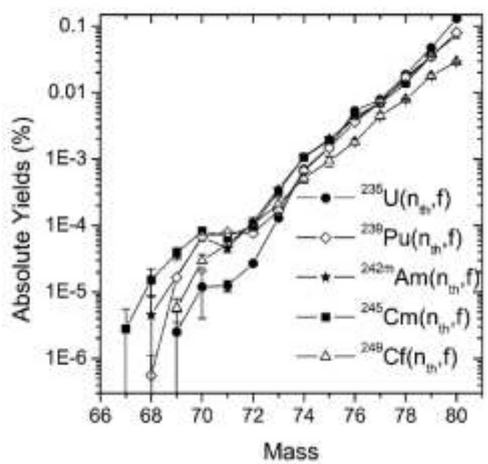
T. Ichikawa et al. PRC 86, 024610 (2012)

Discovery of ^{68}Fe and $^{68,69}Co$



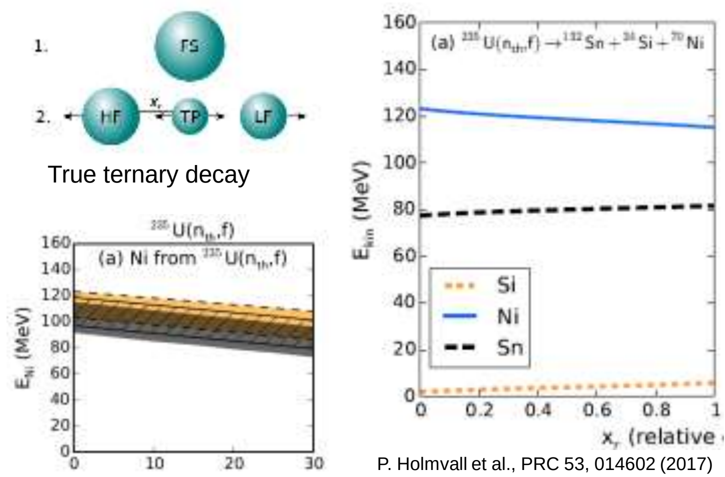
M. Bernas et al. PRL 67, 3661 (1991)

Far asymmetric fission studied at LOHENGRIN



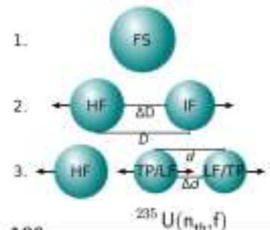
D. Rochman et al. NPA 735, 3 (2004)

Calculation of allowed kinetic energy range

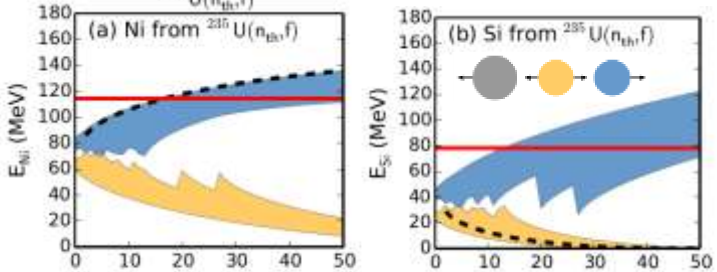


P. Holmvall et al., PRC 53, 014602 (2017)

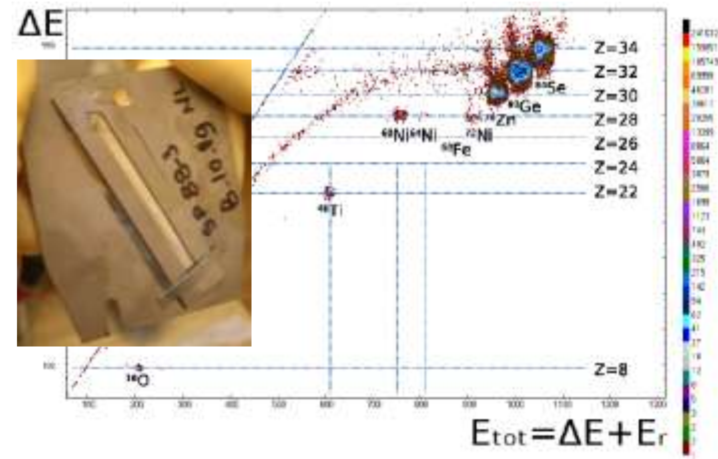
Allowed kinetic energies in sequential model



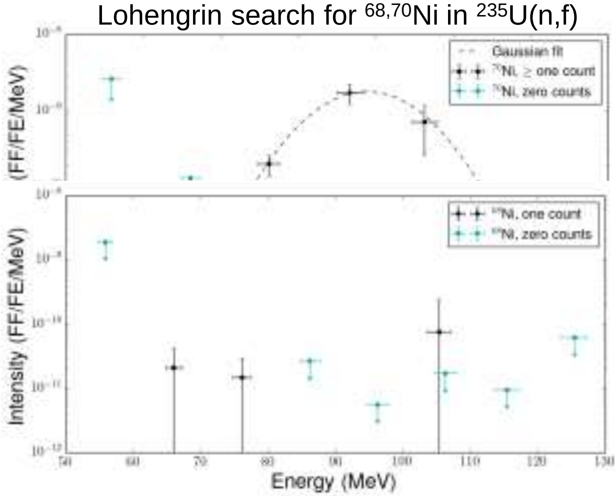
P. Holmvall et al., PRC 53, 014602 (2017)



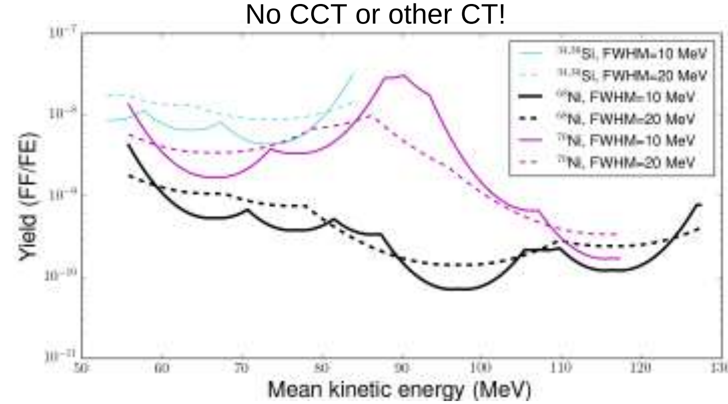
ΔE/E identification plot: A/q = 4



Lohengrin search for ^{68,70}Ni in ^{235}U(n,f)

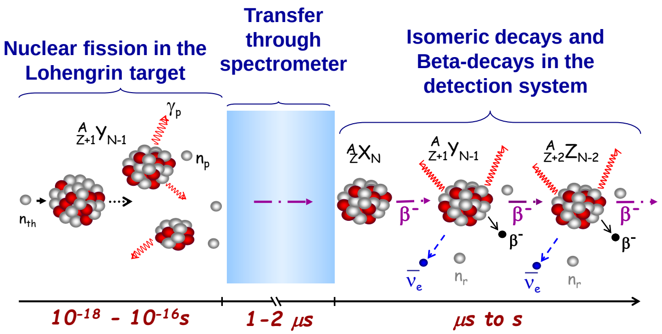


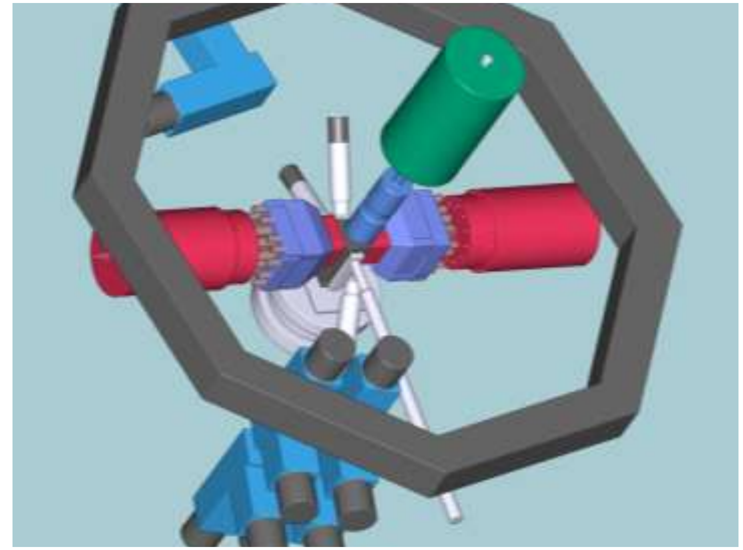
No CCT or other CT!



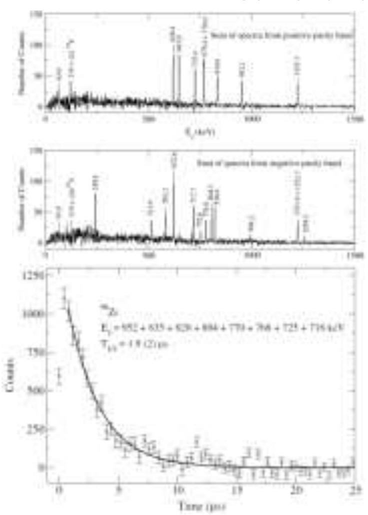
Cluster tripartition into ^{68,70}Ni refuted at 10^{-8} level

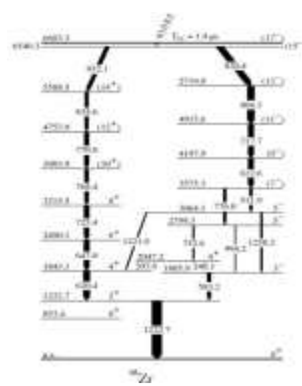
Fission product spectroscopy





17⁻ isomer at 6.6 MeV in ^{98}Zr

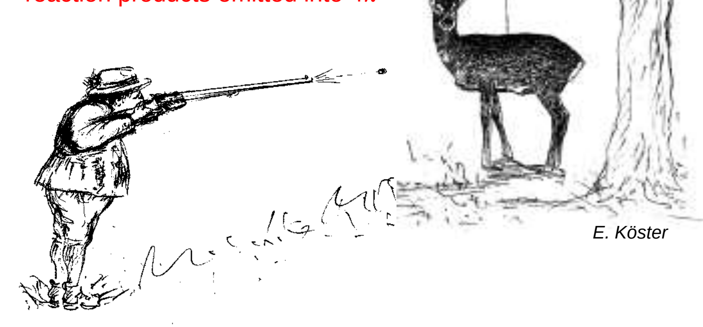




Over 70 microsecond isomers studied so far at LOHENGRIN: $T_{1/2} \geq 0.5$ μs

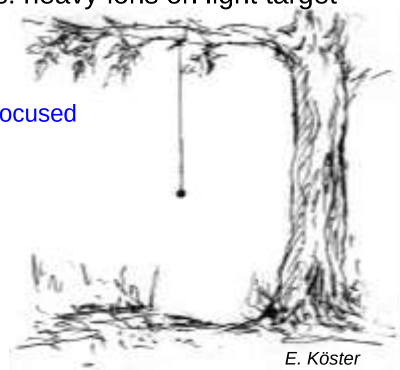
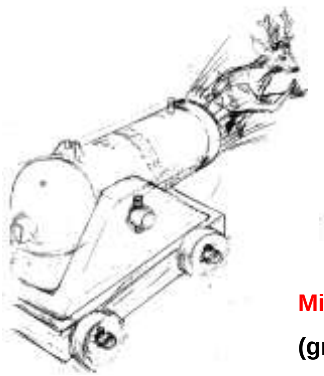
Normal kinematics: n, p or light ions on heavy target

- "conventional" accelerator providing "easily" high beam intensity
- dedicated target required
- reaction products emitted into 4π



Inverse kinematics: heavy ions on light target

- complex and expensive accelerator
- reaction products forward focused

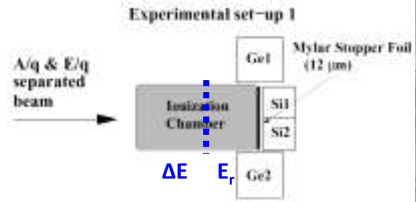



E. Köster

Mind the losses during stopping!

(graphical representation censored)

Detection setup



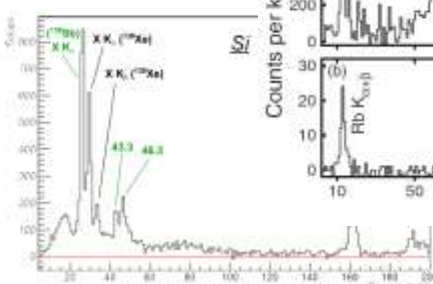
Gamma ray detection
Conversion-electron detection

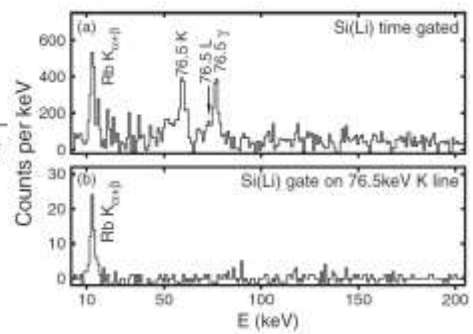
Z identification **via energy loss (Bethe Bloch)**

$$\frac{dE}{dx} \sim \frac{Z^2}{E}$$

Intrinsic advantage:
< 1 µm longitudinal straggling
cf. BigRIPS 1100 µm, FRS 4000 µm

Conversion electron spectroscopy at LOHENGRIN





M. Rudigier et al.
Phys Rev C 87, 064317 (2013)

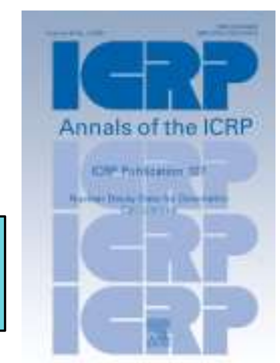
$T_{1/2}$ > 500 ns, E > 12 keV

G. Gey, PhD LPSC Grenoble (2014)

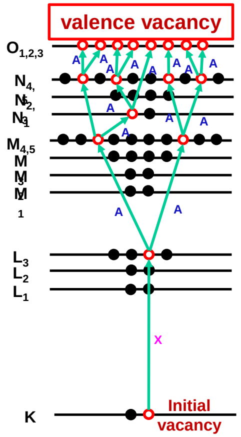
Which nuclear data do we need for nuclear medicine?

1. Half-life
2. Gamma ray energies (roughly!)
3. Particle spectra (electrons, alpha)

Lu 177
6.7 d
β^- 0.5…
γ 208, 113…

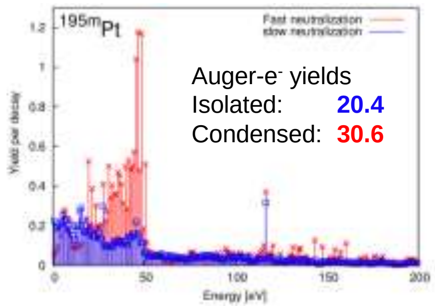


BrIccEmis: Simulation of Auger electron spectra



Two approaches:

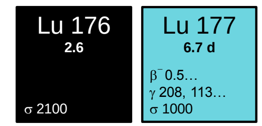
1. No neutralization during Auger cascade **(slow)** isolated atom
2. Valence vacancy immediately neutralized **(fast)** condensed phase



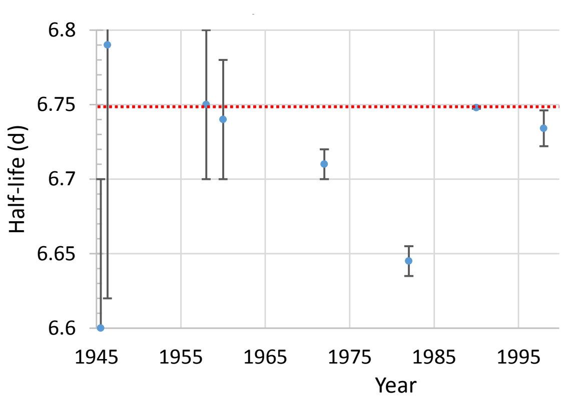
B.Q. Lee et al., Int J Radiat Biol 2016 & Radiother Oncol 2016;118 Suppl 1:S66.

Which nuclear data do we need for nuclear medicine?

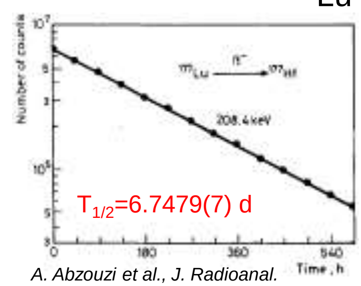
1. Half-life
2. Gamma ray energies (roughly!)
3. Particle spectra (electrons, alpha)
4. Cross-sections



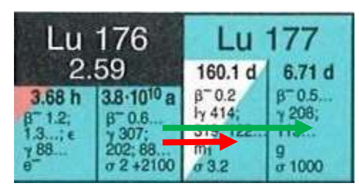
"Time dependence" of the ^{177}Lu half-life

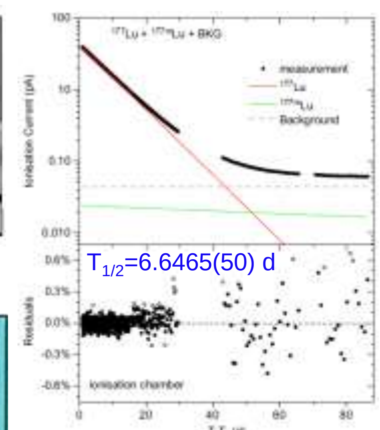


^{177}Lu half-life



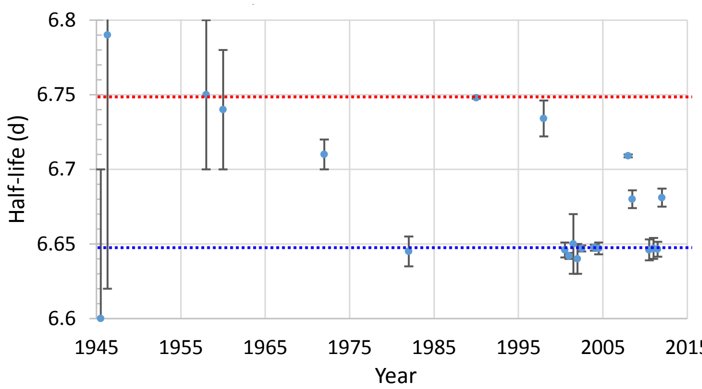
A. Abzouzi et al., J. Radioanal. Nucl. Chem. Lett. 144 (1990) 359.



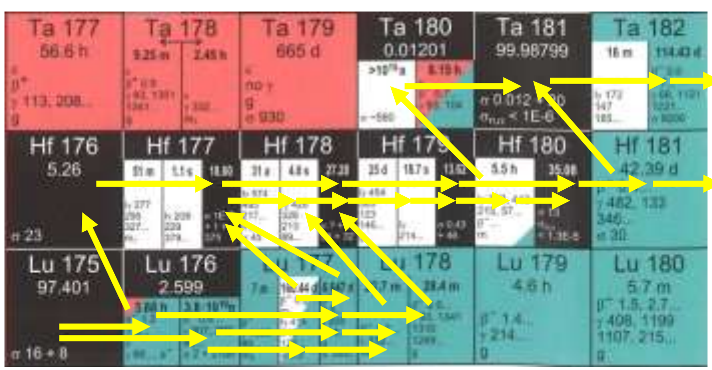


S. Pommé et al., Appl. Radiat. Isot. 69 (2011) 1267.

“Time dependence” of the ^{177}Lu half-life

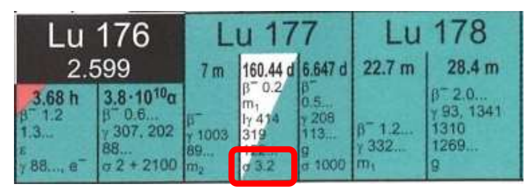


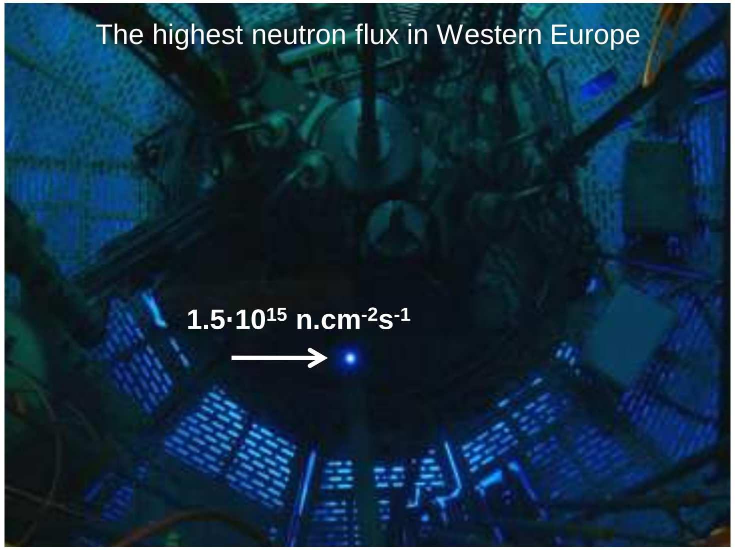
Bateman’s nightmare – Phil Walker’s dream



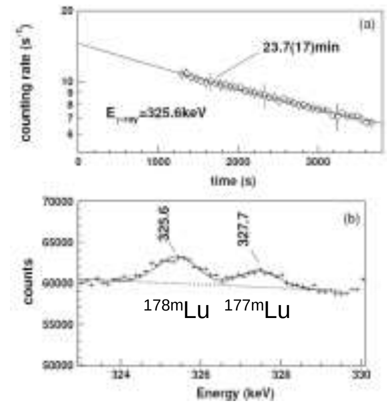
Thermal neutron capture cross-section σ_{th}
Resonance integral I_{γ}
In addition: Fast neutron cross-sections for (n,p), (n,α), (n,n’)

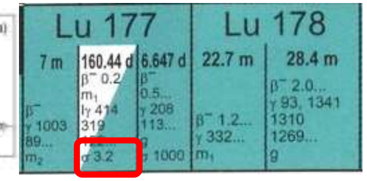
^{177m}Lu(n,γ)^{178m}Lu

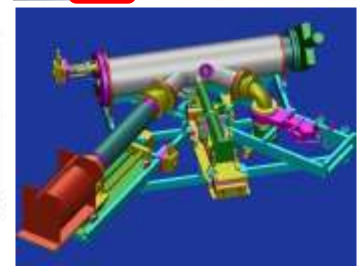




^{177m}Lu(n,γ)^{178m}Lu



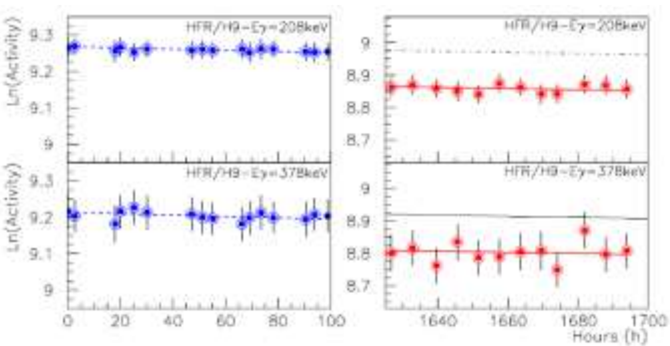




σ[^{177m}Lu(n,γ)] = 368(26) b

G. Bélier et al.
Phys Rev C71, 014603 (2006)

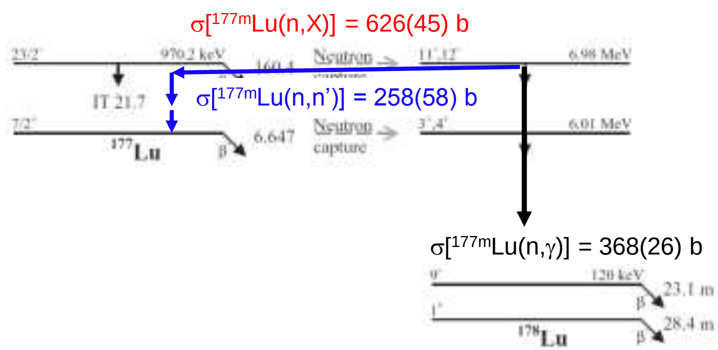
Measurement of burnup cross-section



σ[^{177m}Lu(n,X)] = 626(45) b

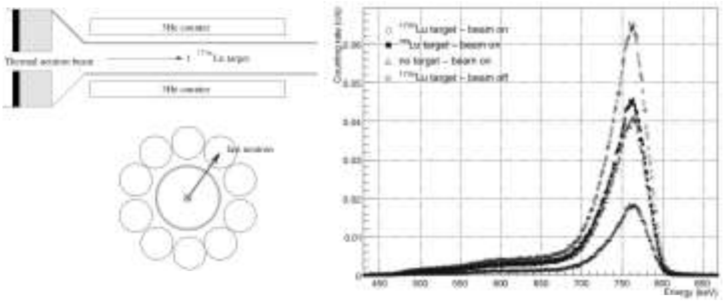
O. Roig et al. Phys Rev C71, 014603 (2006)

Inelastic neutron acceleration (INNA)



O. Roig et al. Phys Rev C71, 014603 (2006)

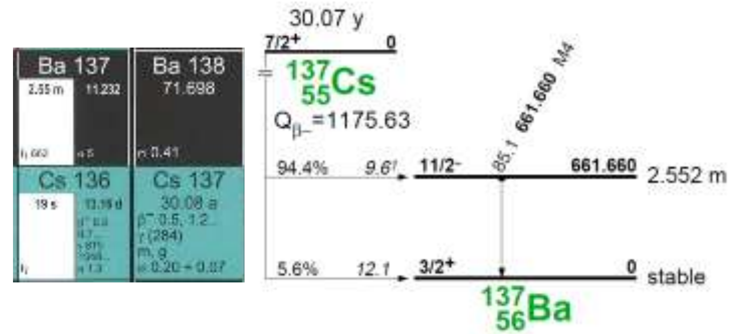
Direct observation of INNA



σ_{INNA} (cold) = 363(48) b
<< 642(151) b
(1/v scaling from thermal)

E. Bauge et al. Eur. Phys. J A48, 113 (2012)

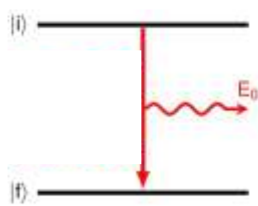
Having fun with a nasty fission product: ^{137}Cs



The double-gamma decay

First discussed by Maria Göppert-Mayer in her doctoral thesis in 1930

M. Göppert-Mayer, Über Elementarakte mit zwei Quantensprüngen (1930)



C Walz et al. Nature 526, 406 (2015)

The double-gamma decay

First discussed by Maria Göppert-Mayer in her doctoral thesis in 1930

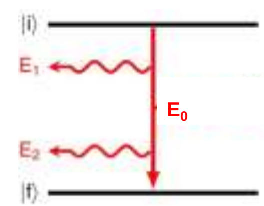
M. Göppert-Mayer, Über Elementarakte mit zwei Quantensprüngen (1930)

Second order process (10^{-6} weaker)

- $E_0 = E_1 + E_2$
- E_1, E_2 are continuous

well studied in atomic physics

M. Lipes et al., PRL 15, 690 (1965)
P.H. Mokler et al., Phys Scr 69, C1 (2004)
K. Ilakovac et al., Rad Phys Chem 75, 1451 (2006)



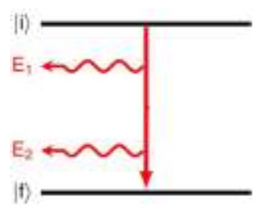
Unit of 2-photon-absorption: 1 GM = 10^{-50} cm^4 s $photon^{-1}$

C Walz et al. Nature 526, 406 (2015)

The double-gamma decay in nuclear physics

γγ-decay only known in a special case:
$0^+ \rightarrow 0^+$ (^{90}Zr, ^{40}Ca, ^{16}O)
J. Schirmer et al., PRL 53, 1897 (1984)
J. Kramp et al., NPA 474, 412 (1987)

never observed in competition to allowed single γ-transition
W. Beusch et al., Helv Phys. Acta 33, 363 (1960)
J. Kramp et al., NPA 474, 412 (1987)
V.K. Basenko et al., Bull. Russ. Acad. 56, 94 (1992)
C.J. Lister et al., Bull. Am. Phys. Soc. 58(13), DNP.CE.3 (2013)

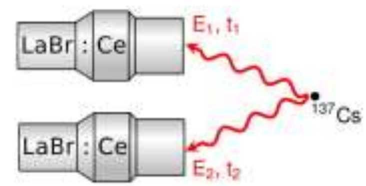


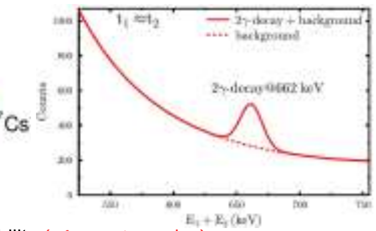
main experimental obstacle:
presence of the one-photon decay

C Walz et al. Nature 526, 406 (2015)

Basic principle of the experiment

•use radioactive ^{137}Cs -source: 16.3(5)μCi



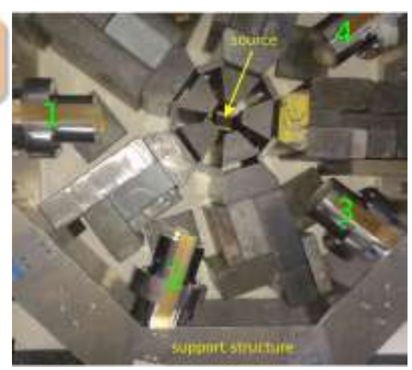


• background ↔ small decay probability (~1 event per day)
  • direct Compton scattering
  • random coincidences
  • cosmic rays, sequential Compton scattering, internal radioactivity

C Walz et al. Nature 526, 406 (2015)

The experimental setup & direct Comption scattering

- 72°: 5 detector pairs
- 144°: 5 detector pairs

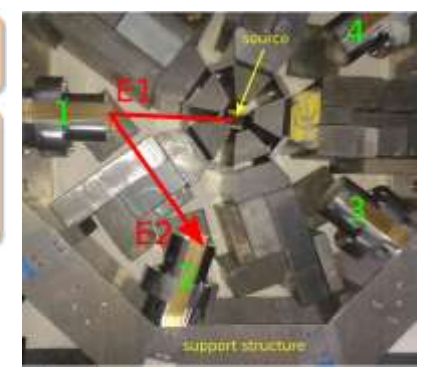


C Walz et al. Nature 526, 406 (2015)

The experimental setup & direct Comption scattering

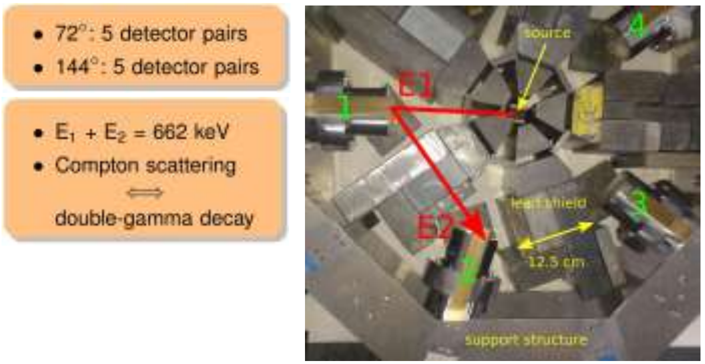
- 72°: 5 detector pairs
- 144°: 5 detector pairs

- $E_1 + E_2 = 662$ keV
- Compton scattering ⟺ double-gamma decay



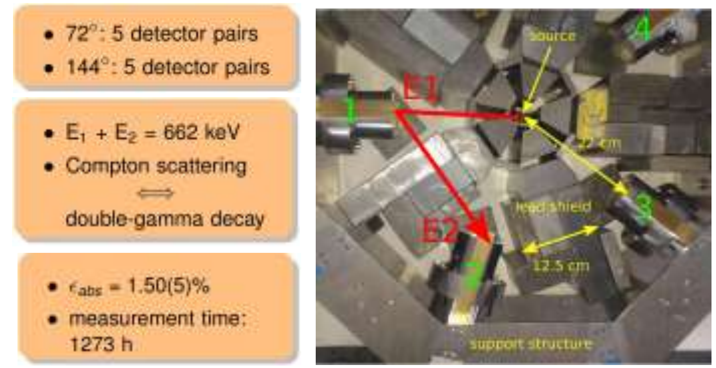
C Walz et al. Nature 526, 406 (2015)

The experimental setup & direct Comption scattering



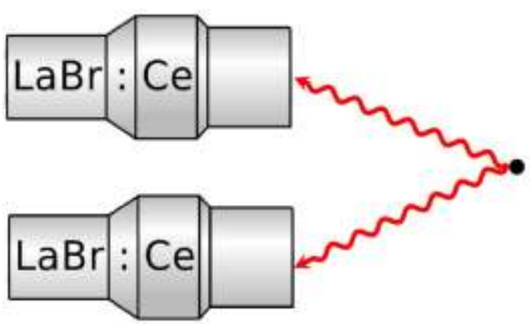
C Walz et al. Nature 526, 406 (2015)

The experimental setup & direct Comption scattering



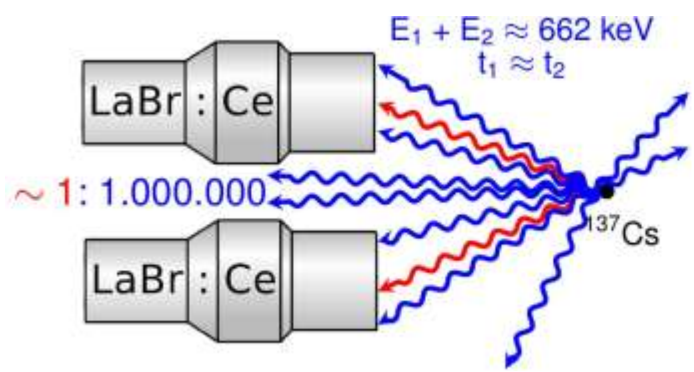
C Walz et al. Nature 526, 406 (2015)

Timing spectrum & random coincidences



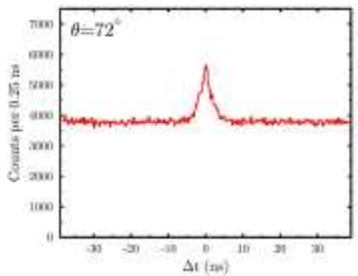
C Walz et al. Nature 526, 406 (2015)

Time spectrum & random coincidences



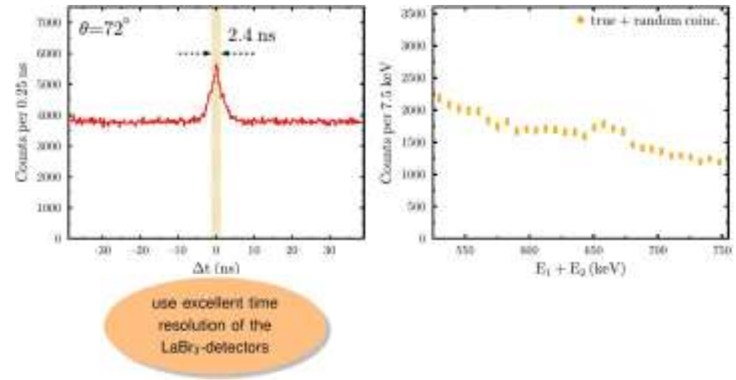
C Walz et al. Nature 526, 406 (2015)

Time spectrum & random coincidences



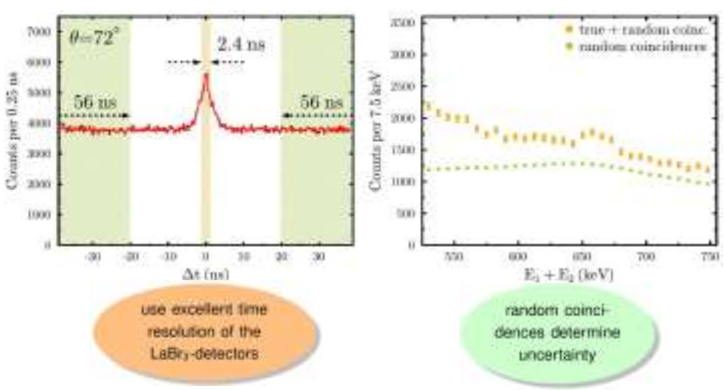
C Walz et al. Nature 526, 406 (2015)

Time spectrum & random coincidences



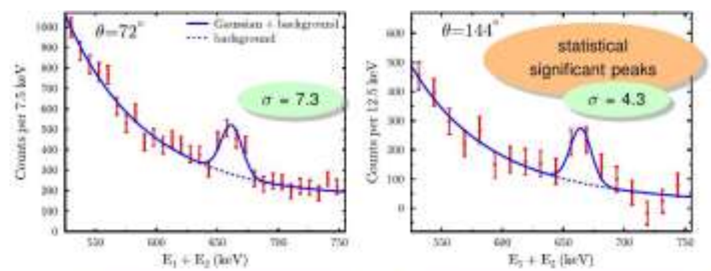
C Walz et al. Nature 526, 406 (2015)

Time spectrum & random coincidences



C Walz et al. Nature 526, 406 (2015)

Results



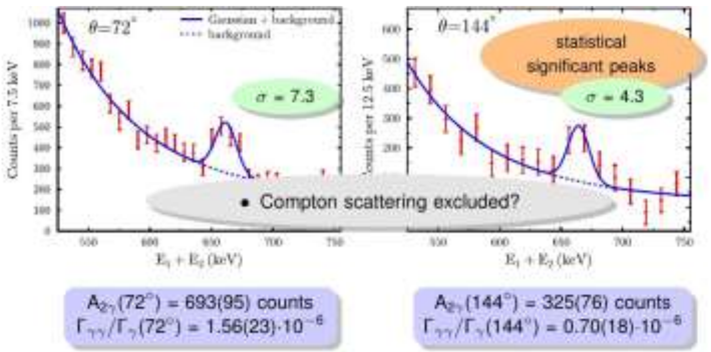
$A_{2\gamma}(72^\circ)$ = 693(95) counts
$\Gamma_{\gamma\gamma}/\Gamma_\gamma(72^\circ) = 1.56(23)\cdot 10^{-6}$

$A_{2\gamma}(144^\circ)$ = 325(76) counts
$\Gamma_{\gamma\gamma}/\Gamma_\gamma(144^\circ) = 0.70(18)\cdot 10^{-6}$

► Result: Successful observation of the competitive double-gamma decay

C Walz et al. Nature 526, 406 (2015)

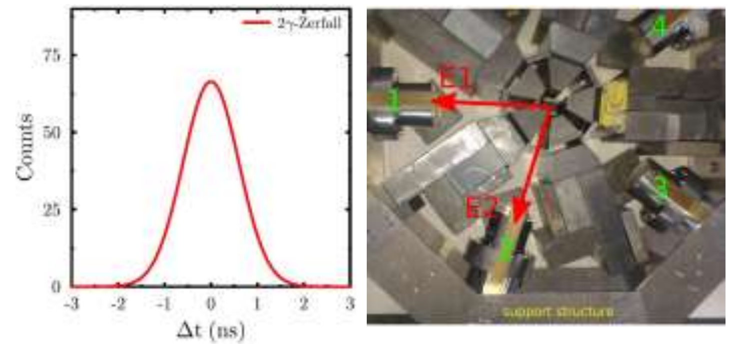
Results



► Result: Successful observation of the competitive double-gamma decay

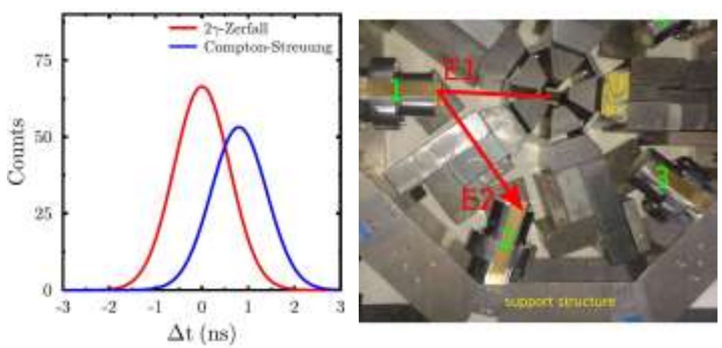
C Walz et al. Nature 526, 406 (2015)

Critical analysis (1)



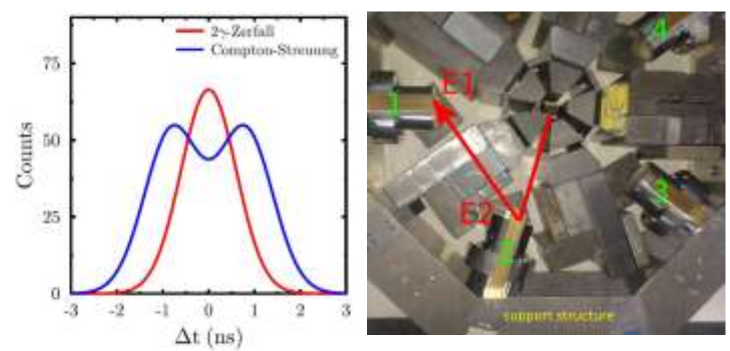
C Walz et al. Nature 526, 406 (2015)

Critical analysis (1)



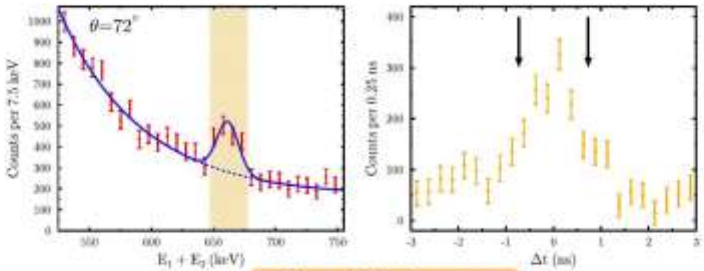
C Walz et al. Nature 526, 406 (2015)

Critical analysis (1)



C Walz et al. Nature 526, 406 (2015)

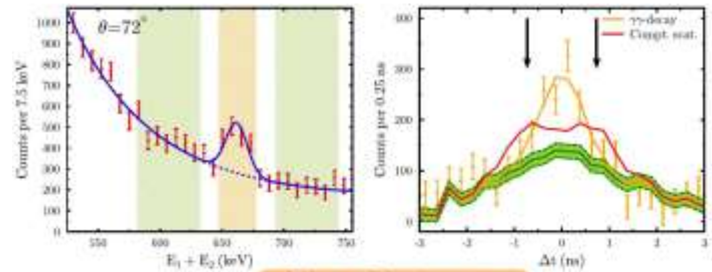
Critical analysis (2)



Shape of the time spectrum proofs that Compton scattering is not the origin of the peak at 662 keV

C Walz et al. Nature 526, 406 (2015)

Critical analysis (2)



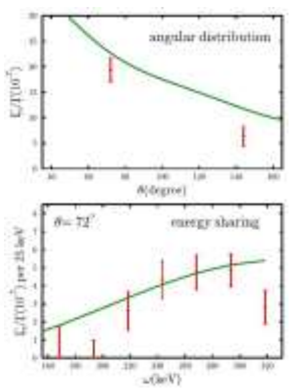
Shape of the time spectrum proofs that Compton scattering is not the origin of the peak at 662 keV

C Walz et al. Nature 526, 406 (2015)

Results & comparison to QPM

	exp	QPM
$\Gamma_{\gamma\gamma}/\Gamma_{\gamma}(10^{-6})$	2.1(3)	2.69
$\alpha_{M2E2}(\frac{e^2 fm^4}{MeV})$	+38.2(36)	+42.6
$\alpha_{E3M1}(\frac{e^2 fm^4}{MeV})$	+7.4(38)	+9.5

- α_{M2E2} dominates
- relative sign between α_{M2E2} and α_{E3M1} is positive
- good description in the framework of the QPM



C Walz et al. Nature 526, 406 (2015)

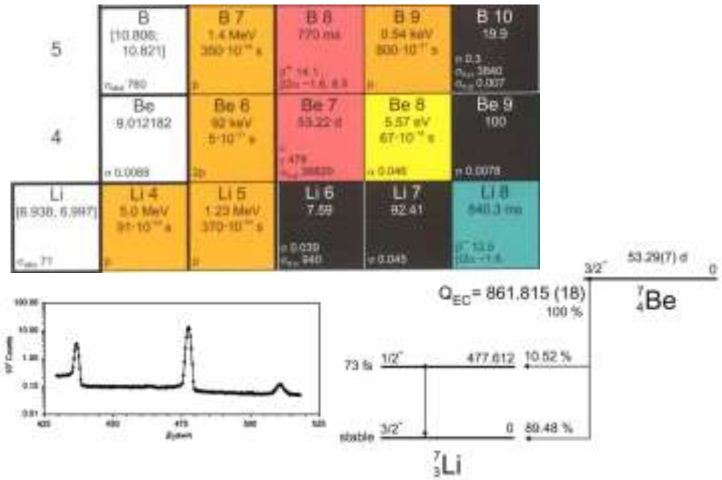
Dependence of radioactive decay on external conditions?

The nuclear decay constant is a fundamental constant which cannot be changed by external, non-nuclear processes.
E Rutherford and F Soddy, J Chem Soc Trans 81, 837 (1902)

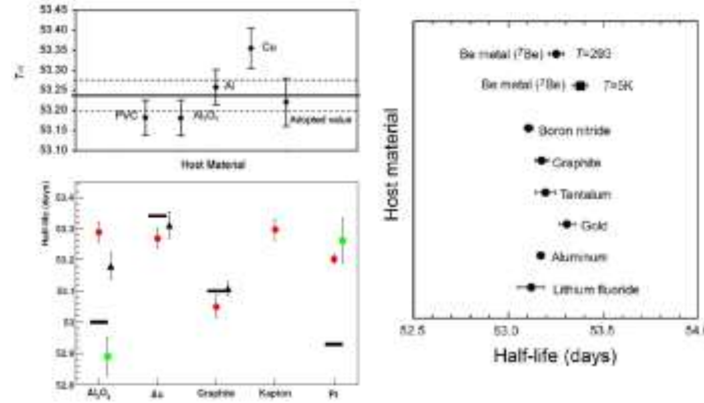
Effects of temperature, pressure, electromagnetic fields, chemistry, etc. less than 1%.
GT Emery, Ann Rev Nucl Sci 22, 165 (1972)

No dependence on season, moon phase, government, etc.
However,

^{7}Be: the lightest EC decaying isotope

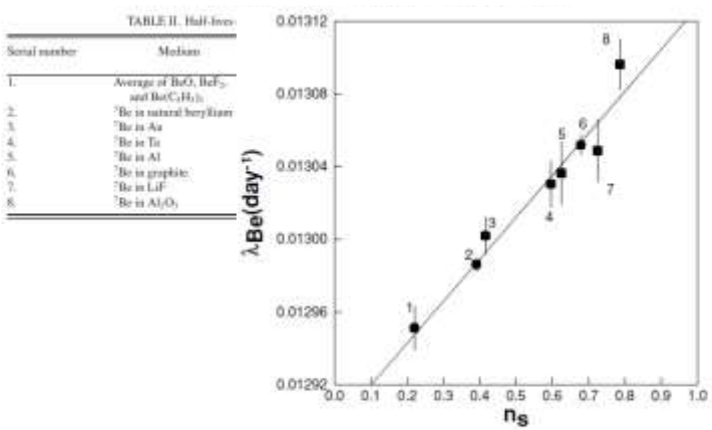


Different implantation conditions



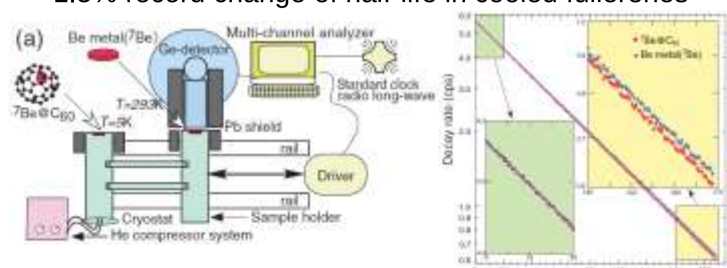
Y. Nir-El et al., Phys Rev C 75, 012801 (2012).
C. Mazzocchi et al., Acta Phys Pol B 43, 279 (2012).
T. Ohtsuki et al., Proc Radiochim Acta 1, 101 (2011).

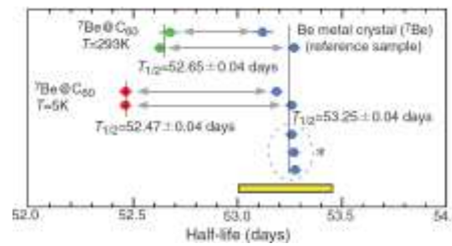
Systematics of ^{7}Be decay rate measurements



P. Das and A. Ray, Phys Rev C 71, 025801 (2005).

1.5% record change of half-life in cooled fullerenes





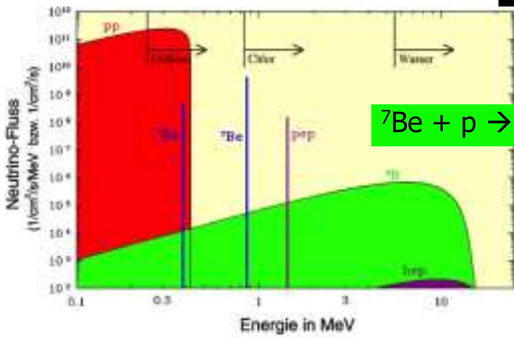
T. Ohtsuki et al., PRL 71, 025801 (2005).

^{7}Be decay in the Sun

Ionization of ^{7}Be in the Sun can be ~20-30 %

AV Gruzinov, JN Bahcall, Astroph J 490, 437 (1997)

$^{7}\mathrm{Be} + e^{-} \rightarrow {}^{7}\mathrm{Li} + \nu_e$

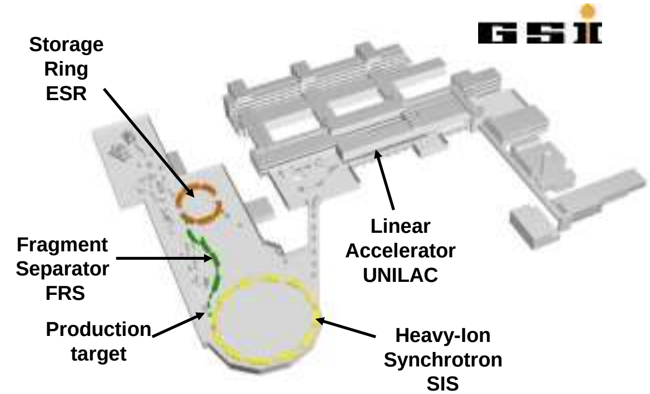


$^{7}\mathrm{Be} + p \rightarrow {}^{8}\mathrm{B} + \gamma$

Y. Litvinov, GSI

Change the electron density more radically ?

⇒ highly charged ions



Y. Litvinov, GSI

Nuclear beta decay

Nuclear weak decay in general form:

$$n + \nu_e \leftrightarrow p + e^{-}$$

i) continuum beta decay:

$n \rightarrow p + e^{-} + \bar{\nu}_e$ $\quad \beta^{-}$ – decay

$p \rightarrow n + e^{+} + \nu_e$ $\quad \beta^{+}$ – decay

ii) two-body beta decay:

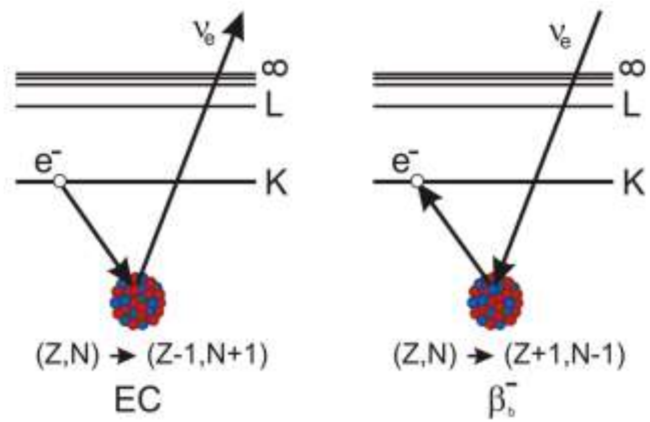
$p + e_b^{-} \rightarrow n + \nu_e$ $\quad$ Orbital electron capture (EC)

$n \rightarrow p + e_b^{-} + \bar{\nu}_e$ $\quad$ Bound state beta decay (β_b^{-})

$p + e^{-} \rightarrow n + \nu_e$ $\quad$ Free electron capture

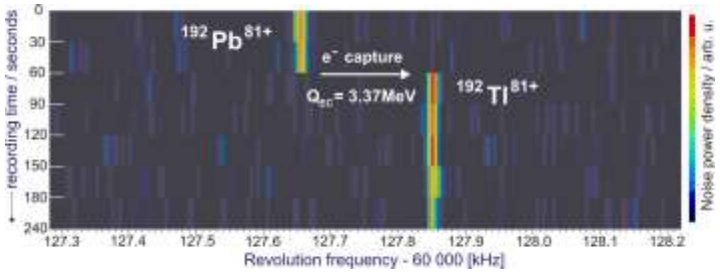
Y. Litvinov, GSI

Two-body beta decay



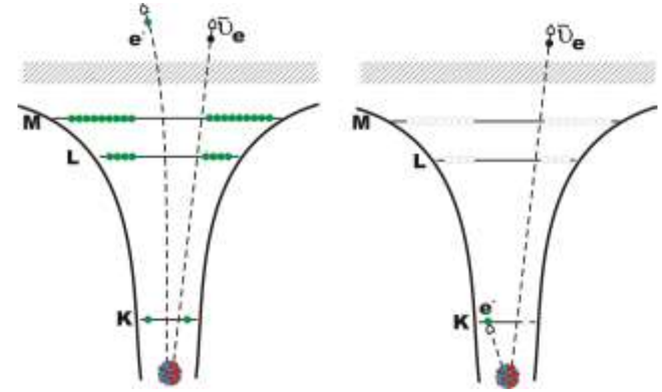
Y. Litvinov, GSI

Observation in Schottky frequency spectra



Y. Litvinov, GSI

Bound-state beta decay

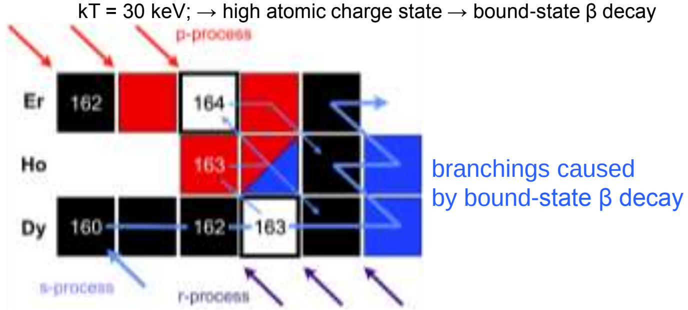


Q_β $\quad$ $Q_\beta + E_K$

Y. Litvinov, GSI

Bound state beta decay of ^{163}Dy

s process: slow neutron capture and β^- decay near valley of β stability at kT = 30 keV; → high atomic charge state → bound-state β decay



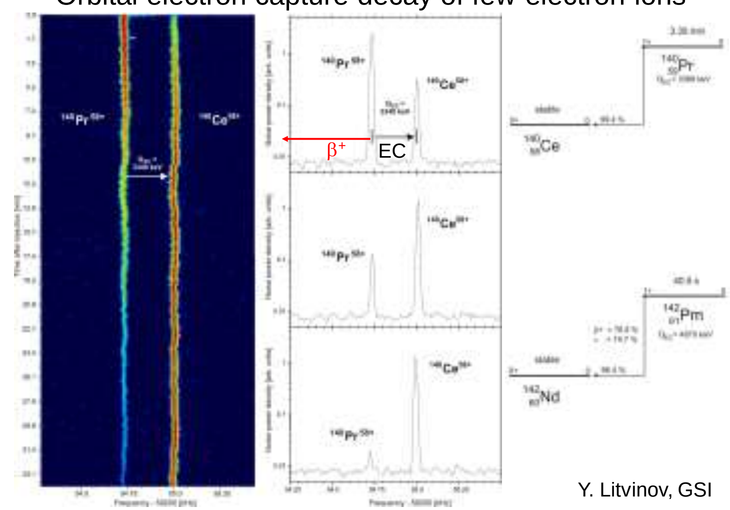
^{163}Dy $\quad Q_\beta = -2.8$ keV $\quad$ stable

$^{163}Dy^{66+}$ $\quad Q_\beta + E_K = +50$ keV $\quad T_{1/2} = 47(5)$ d

(L capture also energetically allowed but negligible due to Q^5)

M. Jung et al., Phys. Rev. Lett. 69 (1992) 2164 $\quad$ Y. Litvinov, GSI

Orbital electron capture decay of few-electron ions



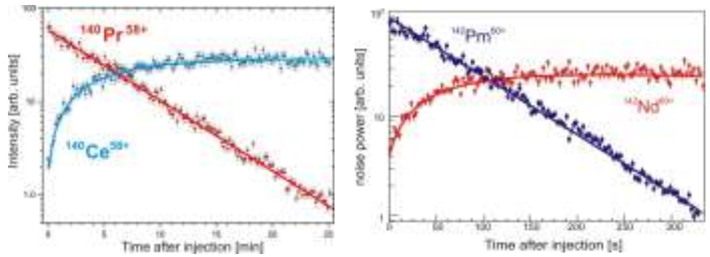
Y. Litvinov, GSI

Orbital electron capture decay of few-electron ions

Expectations: λ_{EC}(H-like) / λ_{EC}(He-like) ≈ 0.5

λ_{EC}(H-like) / λ_{EC}(He-like) = 1.49(8)

λ_{EC}(H-like) / λ_{EC}(He-like) = 1.44(6)

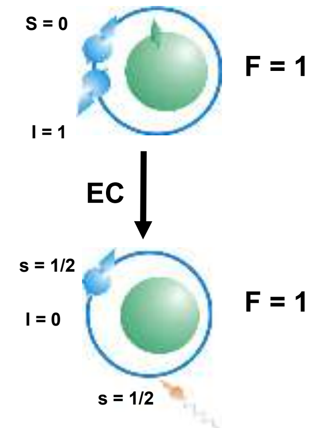


Yu.A. Litvinov et al., Phys. Rev. Lett. 99 (2007) 262501

N. Winckler et al., Phys. Lett. B579 (2009) 36

Y. Litvinov, GSI

Selection rule



$$\lambda_{EC}^{H} = \lambda_{EC}^{He} \cdot \frac{2I_i + 1}{2F_i + 1}$$

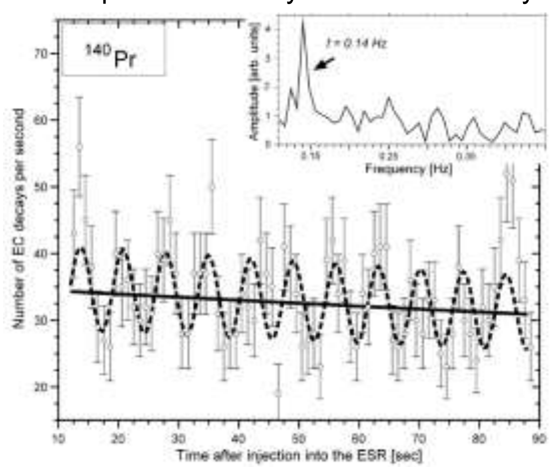
$$I_i = 1$$

$$F_i = I_i - s = 1/2$$

~~$F_i = I_i + s = 3/2$~~

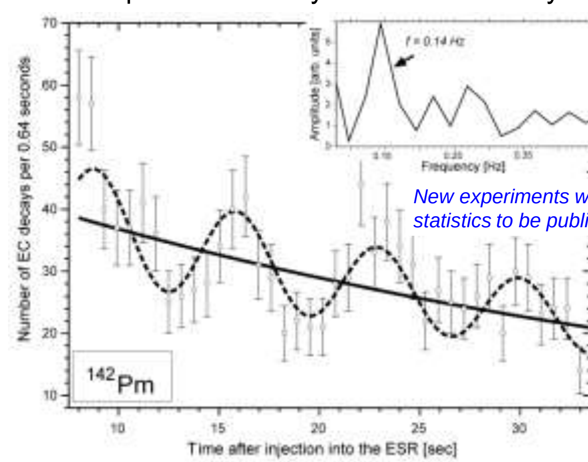
Y. Litvinov, GSI

Non-exponential decay: the "GSI anomaly"



Y. Litvinov et al., Phys. Lett. B664 (2008) 162

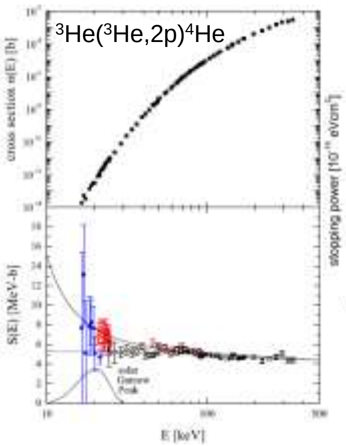
Non-exponential decay: the "GSI anomaly"

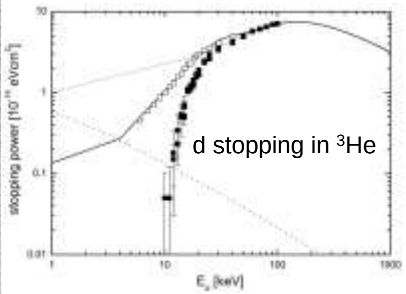


New experiments with high statistics to be published...

Y. Litvinov et al., Phys. Lett. B664 (2008) 162

Nuclear astrophysics: fusion well below Coulomb barrier





H. Costatini et al., Rep Prog Phys 72, 086301 (2009)

Electron screening in low-energy fusion reactions

cross-sections below Coulomb barrier significantly enhanced by electron screening:

$$f_{\text{lab}}(E) = E(E+U_e)^{-1}\exp(\pi\eta U_e/E)$$

U_e screening energy ≈300 eV for d(d,p)t reaction when deuterium is embedded in metals

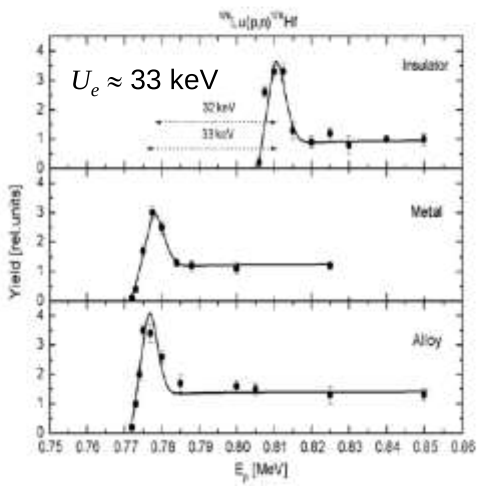
U_e screening energy ≈ U_D from Debye model should scale with nuclear charge of target.

$$U_D = 2.09\times10^{-11}(Z_t(Z_t+1))^{1/2}(n_{\text{eff}}\rho_a/T)^{1/2}\,(\text{eV})$$

U_D = 21(6) keV predicted for Lu and 36(4) keV for $PdLu_{0.1}$ alloy

K.U. Kettner et al., J Phys G 32, 489 (2006)

Electron screening in low-energy fusion reactions



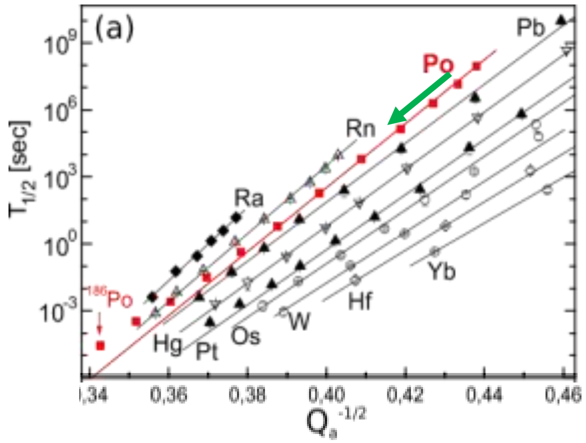
K.U. Kettner et al.,
J Phys G 32, 489 (2006)

Proposed scaling to low temperatures

One major reason of the present work, i.e. to extend screening tests up to $Z_t = 71$, was another prediction of the Debye model concerning radioactive decay of transuranic nuclides ($Z_t \geqslant 82$) in a metallic environment [5]. In general, for the α-decay and β^+-decay one expects a shorter half-life due to the acceleration mechanism of the Debye electrons for these positively charged particles similar as for the protons, deuterons or ^3He in the fusion reactions, while for the β^--decay and e-capture process one predicts a longer half-life (here, deceleration for the negatively charged particles). For example, if the α-decay $^{210}Po \rightarrow \alpha + ^{206}Pb$ with E_α = 5.30 MeV and $T_{1/2}$ = 138 days occurs in a metal cooled to $T = 4$ K, one arrives at $U_D = Z_\alpha Z_t U_e(\text{d+d})(290/4)^{1/2} = 2\times82\times300\ \text{eV}\times8.5 = 420$ keV, where we used again a typical value of $U_e = 300$ eV for the d+d fusion reaction in metals at $T = 290$ K. The enhancement factor then gives $f_{\text{lab}} = 265$, and thus the half-life is shortened to 0.5 days. For the biologically dangerous transuranic waste [12] $^{226}Ra \rightarrow \alpha + ^{222}Rn$ (E_α = 4.78 MeV, $T_{1/2}$ = 1600 years) an analogous calculation leads to $T_{1/2}$ = 1.3 years. Experiments are in progress to test these predictions. If these predictions of the Debye model should also be verified, one may have a cheap solution to remove the transuranic waste (involving all an α-decay) of used-up rods of fission reactors in a time period of a few years. Finally, a reduced half-life of α-emitters such

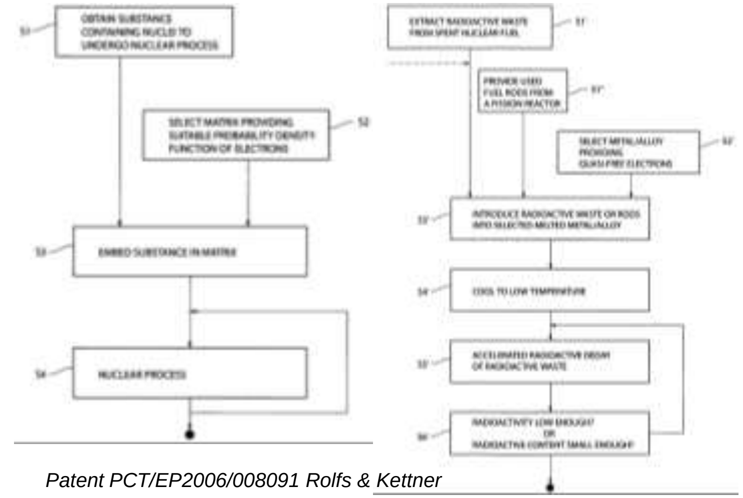
K.U. Kettner et al., J Phys G 32, 489 (2006)

Geiger Nuttall rule for alpha-decay



C. Qi *et al.* Phys.Lett. B734 (2014) 203.

"Cool" solution for nuclear waste ?



Patent PCT/EP2006/008091 Rolfs & Kettner

Experiments invalidate application of Debye model !

^{22}Na in Pd at 12 K: 1.2(2)% faster β^+ decay vs. 11% predicted
B Limata et al., Eur Phys J A 28, 251 (2006)

^{210}Po in Cu at 12 K: 6.3(14)% faster α decay vs. 1000% predicted
F Raiola et al., Eur Phys J A (2007)

^{253}Es in Fe at 4 K: 0.4(3)% faster α decay vs. 10^2 predicted
^{253}Es in Fe at 50 mK: 1.4(6)% faster α decay vs. >>10^6 predicted
N Severijns et al., Phys Rev C 76, 024304 (2007)

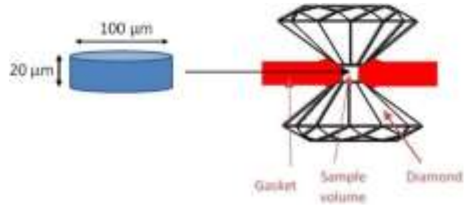
^{224}Ra in Fe, at 1 K and 20 mK: <1% effect on $T_{1/2}$
^{225}Ra in Fe, at 1 K and 20 mK: <0.5% effect on $T_{1/2}$
^{227}Ac in Fe, at 1 K and 20 mK: <1% effect on $T_{1/2}$
vs. effects of 10^4 to 10^{10} predicted by Debye model
NJ Stone et al., Nucl Phys A 793, 1 (2007)

Theoretical arguments

Correct treatment of screening effects in tunnelling of alpha particles gives for ^{210}Po and ^{226}Ra at 4 K a predicted half-life increase of <0.1% with the Debye model and 0.9% and 1.3% with the Thomas-Fermi model.

N.T. Zinner, Nucl. Phys. A 781, 81 (2007)

Effect of pressure on alpha decay ?

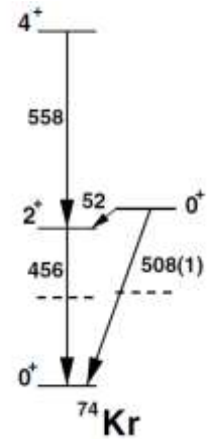


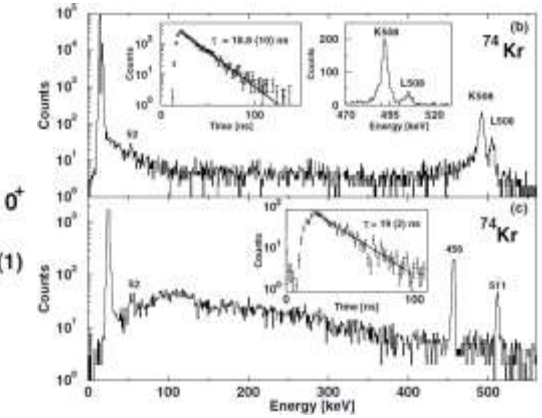
Diamond anvil cell

Expected effect: -0.02% half-life change for ^{241}Am at 0.5 MBar

N. Nissim et al. PRC 94, 014601 (2016)

Isomeric decay





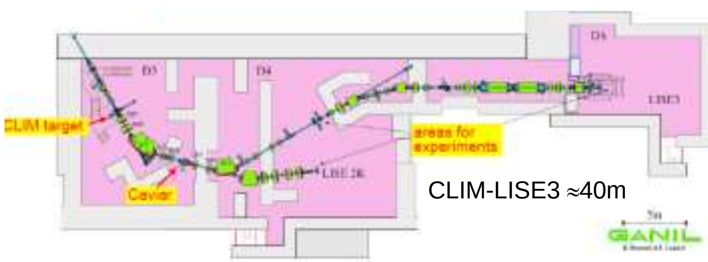
E. Bouchez et al. PRL 90, 082502 (2003)

Decay losses in flight ?

TOF ≈ 400 ns

τ = 18.8 ns, i.e. $T_{1/2}$ = 13.0 ns

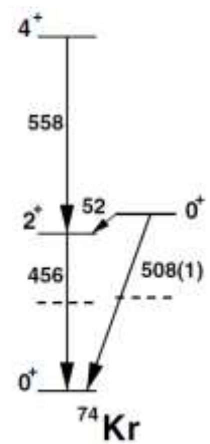
$\exp(-400/18.8) \approx 6\cdot 10^{-10}$



CLIM-LISE3 ≈40m

R. Anne et al., Nucl. Instr. Meth. A257 (1987) 215.
R. Anne et al., Nucl. Instr. Meth. B70 (1992) 276.

Isomeric decay of fully stripped $^{74}Kr^{36+}$



τ = 18.8 ns = $1/\lambda(E2) + 1/\lambda(E0)$

$\lambda(E2) / \lambda(E0) = 1.2$

$\tau(E2)$ = 34.4 ns   ~~$\tau(E0)$ = 41.5 ns~~

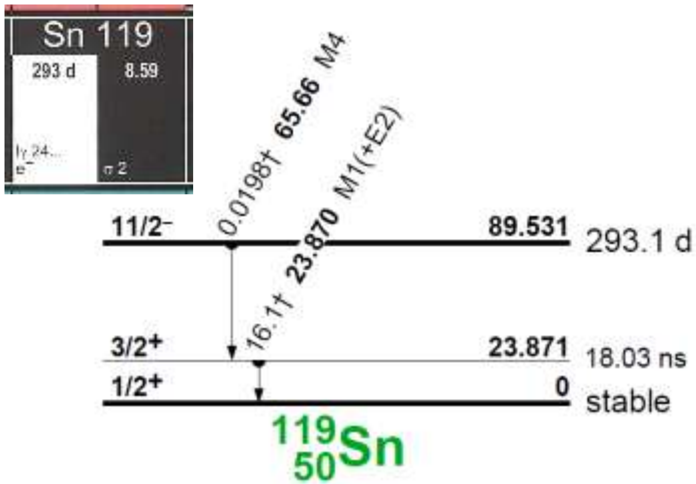
$\alpha(E2)$ = 8.7 = [decays by CE] / [all decays]

$\tau(E2\text{-}\gamma) = (\alpha(E2)+1)\cdot \tau(E2)$ = 334 ns

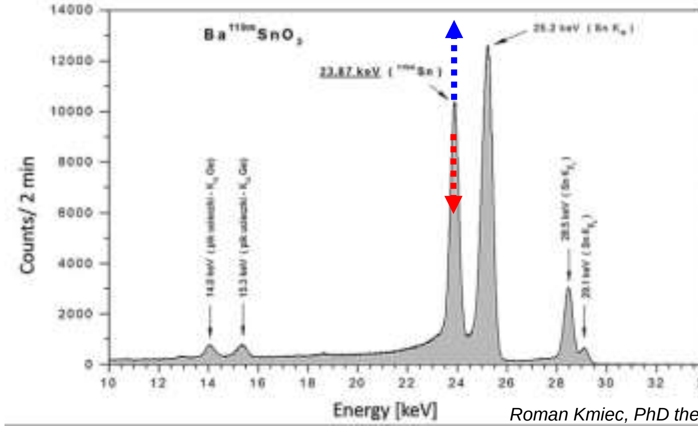
$\exp(-400/334) = 0.3$

E. Bouchez et al. PRL 90, 082502 (2003)

Isomeric decay of ^{119m}Sn

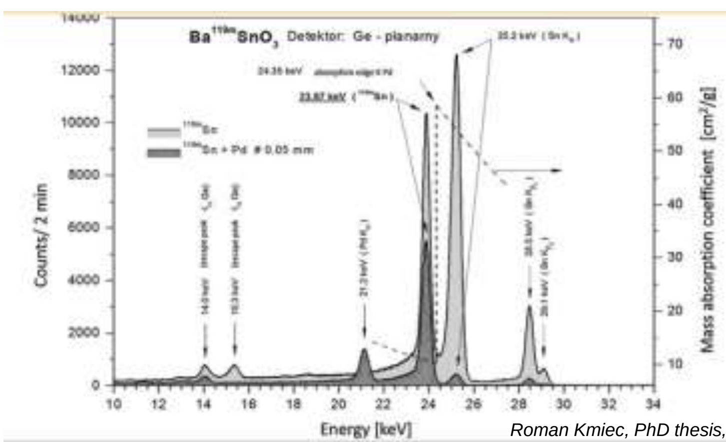


^{119m}Sn spectrum with planar Ge detector



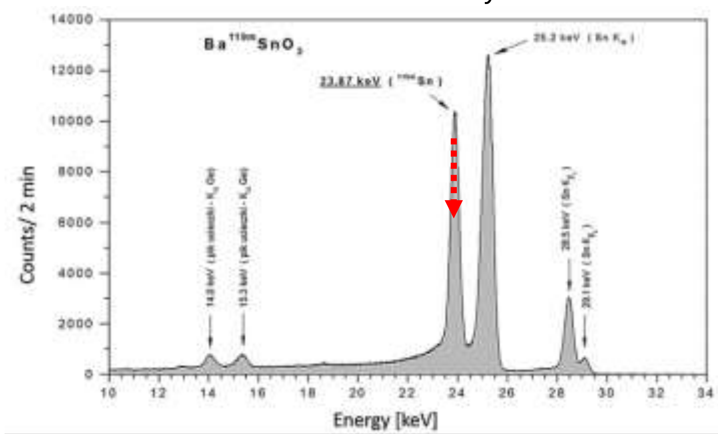
Roman Kmiec, PhD thesis, IFJ Krakow, 2005.

Attenuation with Pd layer



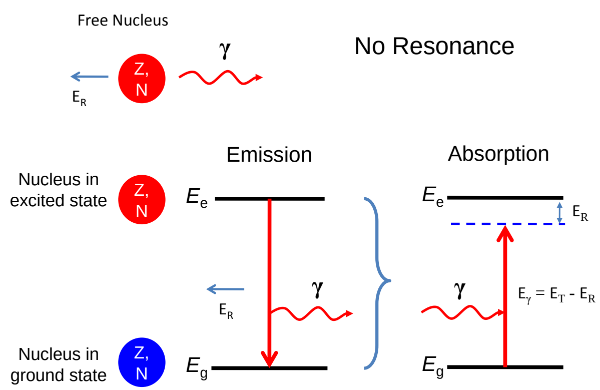
Roman Kmiec, PhD thesis, IFJ Krakow, 2005.

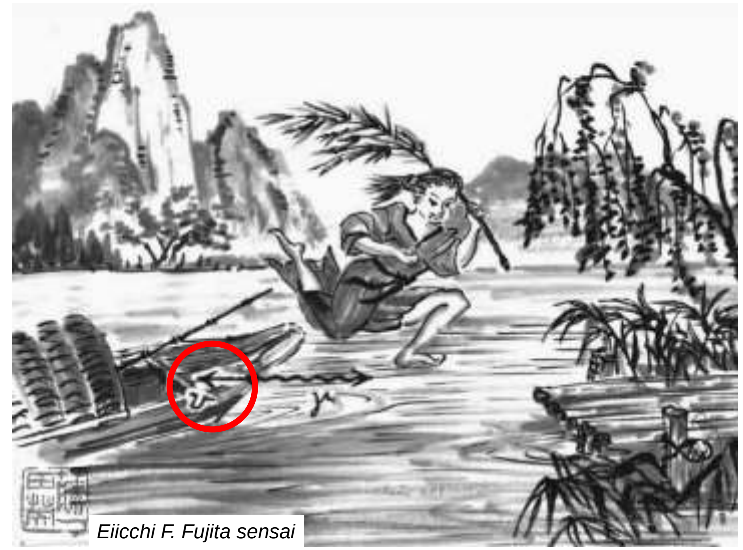
Attenuation with Sn layer



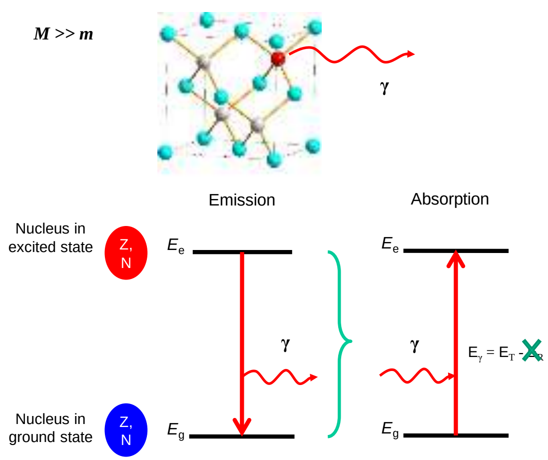
Resonant nuclear attenuation ≈ 1000 · non-resonant attenuation

Nuclear resonance fluorescence





Recoilless nuclear resonance fluorescence



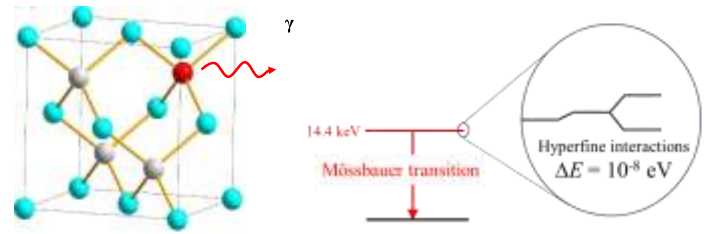
Mössbauer effect

1957 discovery of recoilless nuclear resonance

1961 Nobel Prize in Physics

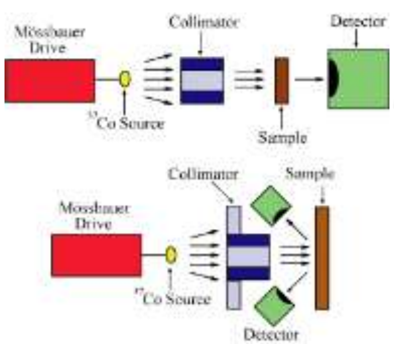
Application of the Mössbauer effect

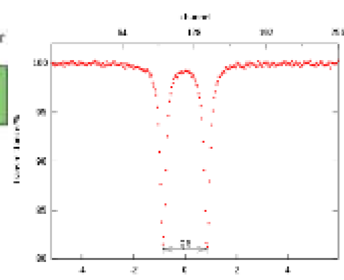
Interactions between the nucleus and its surrounding electrons…



…causing changes in the nuclear (and electronic) energy levels.

Mössbauer spectroscopy





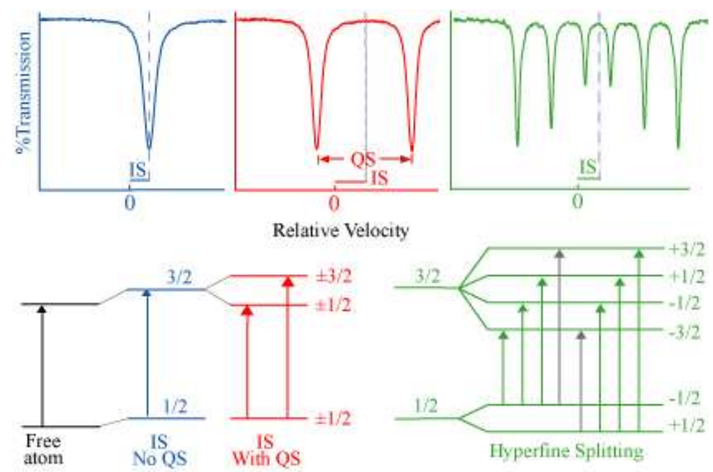
1 mm/s ≈ 4 neV

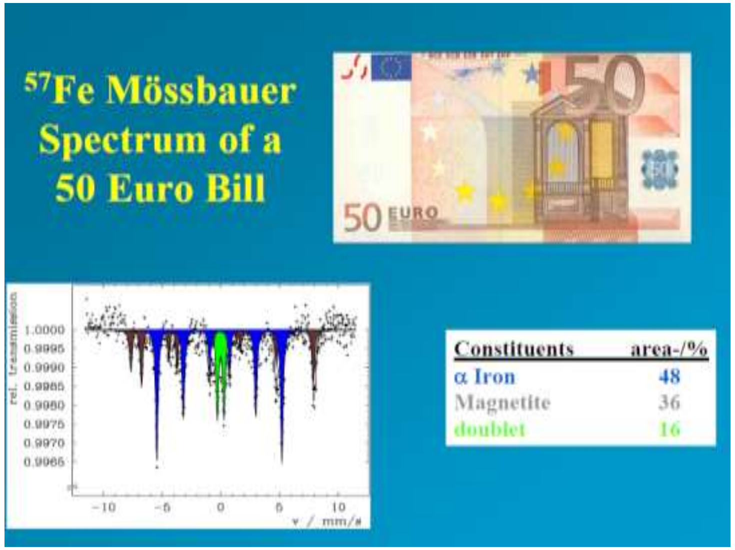
$\Delta E = \hbar/\tau$

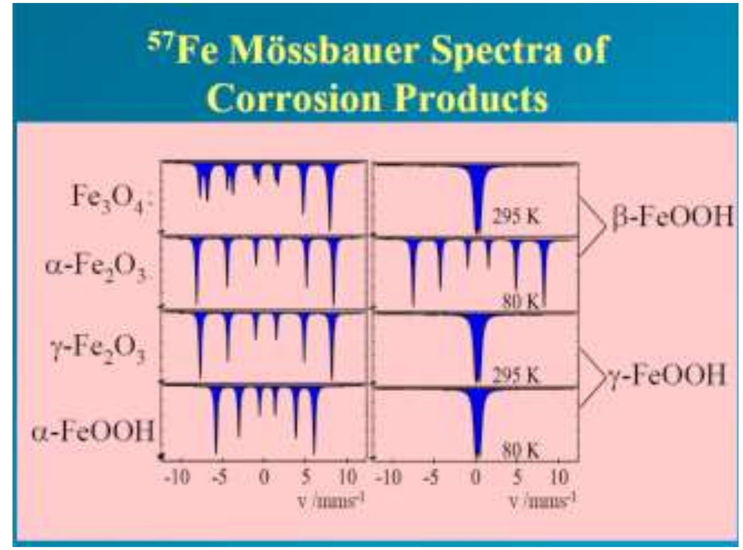
^{57}Fe: $\tau = 140$ ns $\Rightarrow \Delta E = 5$ neV

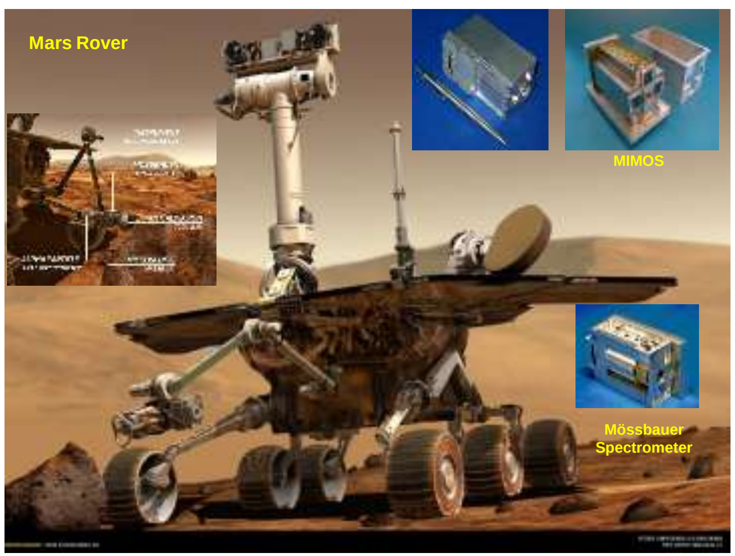
$\Delta E/E = 3\cdot10^{-13}$

Hyperfine interactions

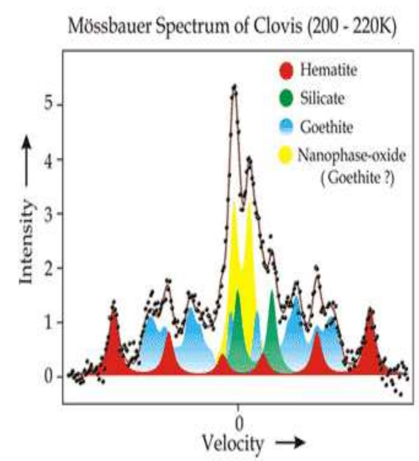






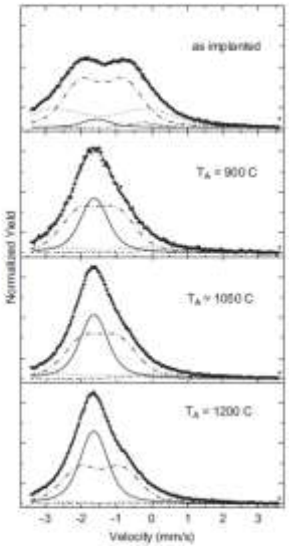


Nuclear method proofs water was on Mars

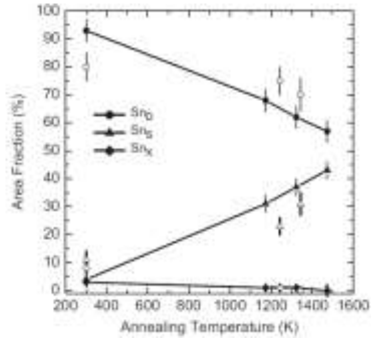


Goethite contains hydroxyl (OH^-) as a part of its structure.

→ water



^{119m}Sn implanted into diamond



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Summary

Physics close to stability remains exciting !

Very high resolution and/or sensitivity and good control of systematics are essential to study subtle effects.

New, astonishing effects may either indicate new physics, or they will disappear with better statistics and/or higher resolution.

Develop your own judgement !

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