Nuclear fast timing: when the speed of light is not fast enough

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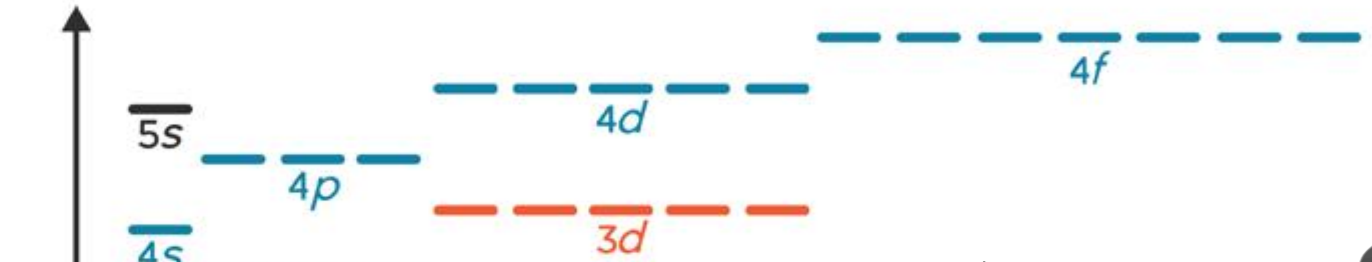
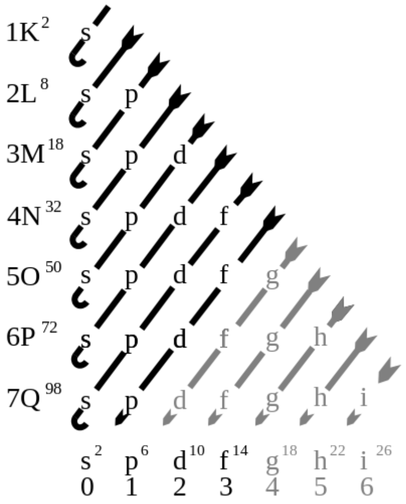
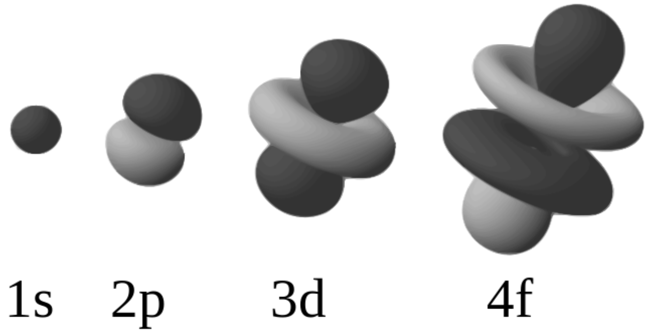
- Nuclear lifetimes and transition strengths
  - Collective motion
  - Weisskopf estimate
- Electronic fast timing
  - Experimental setup
  - Analysis method
- Practical applications:
  - Perturbed Angular Correlations (PACs)
  - Time-of-Flight Positron Emission Tomography (ToF-PET)
  - Proton therapy range verification

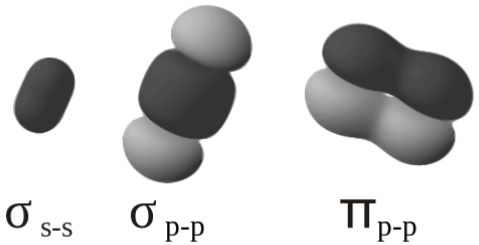
Nuclear lifetimes and transition strengths

Atomic orbitals



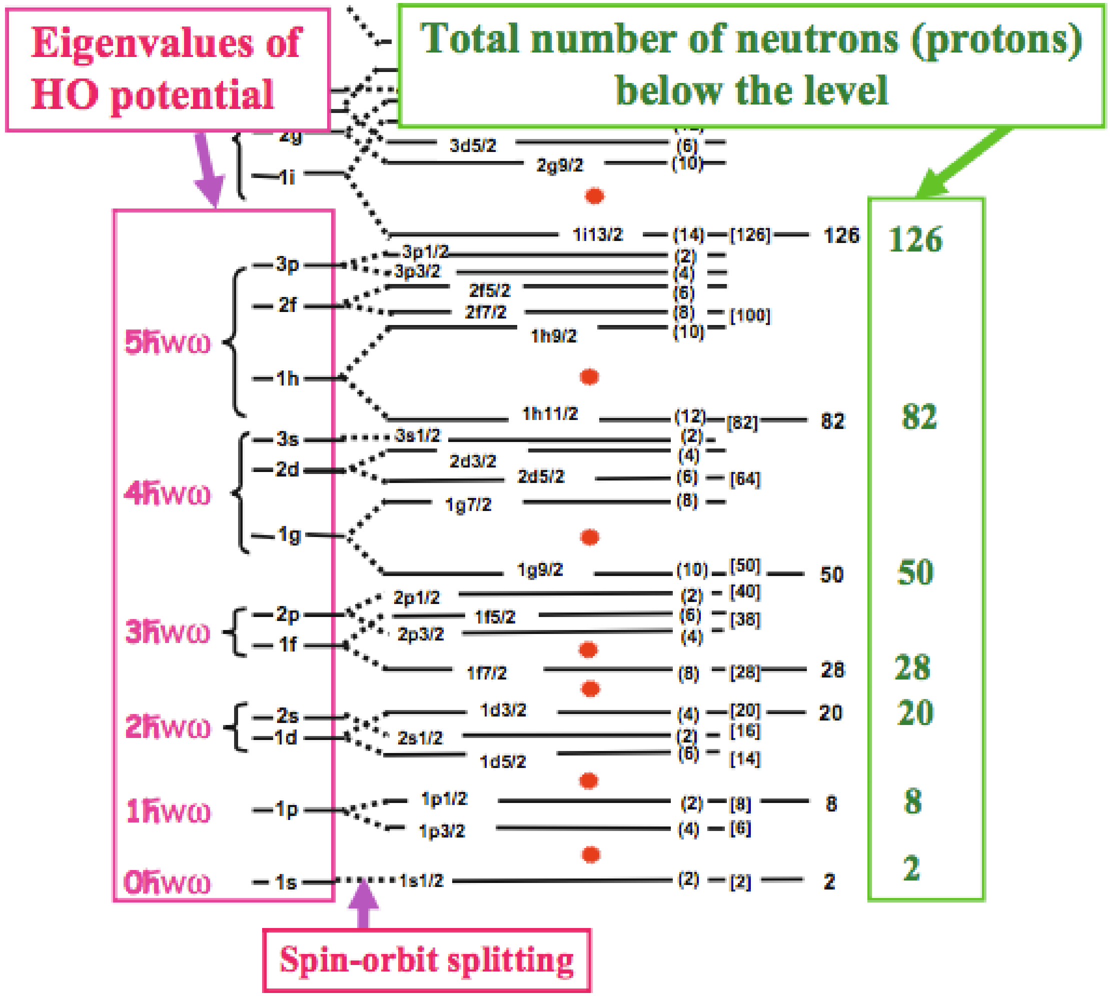


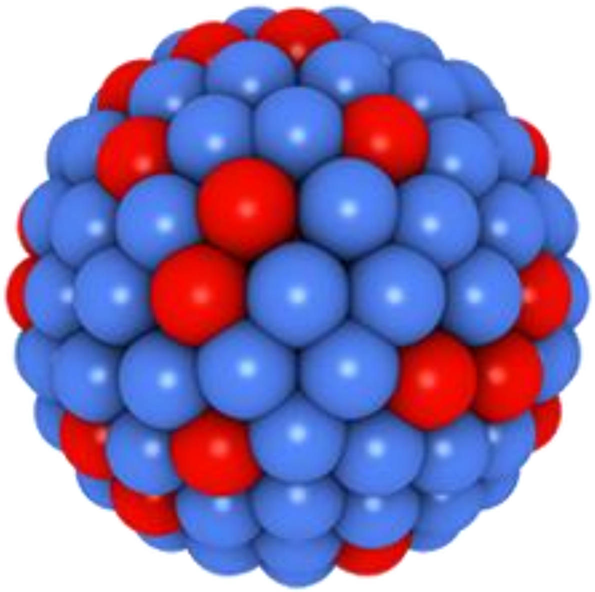




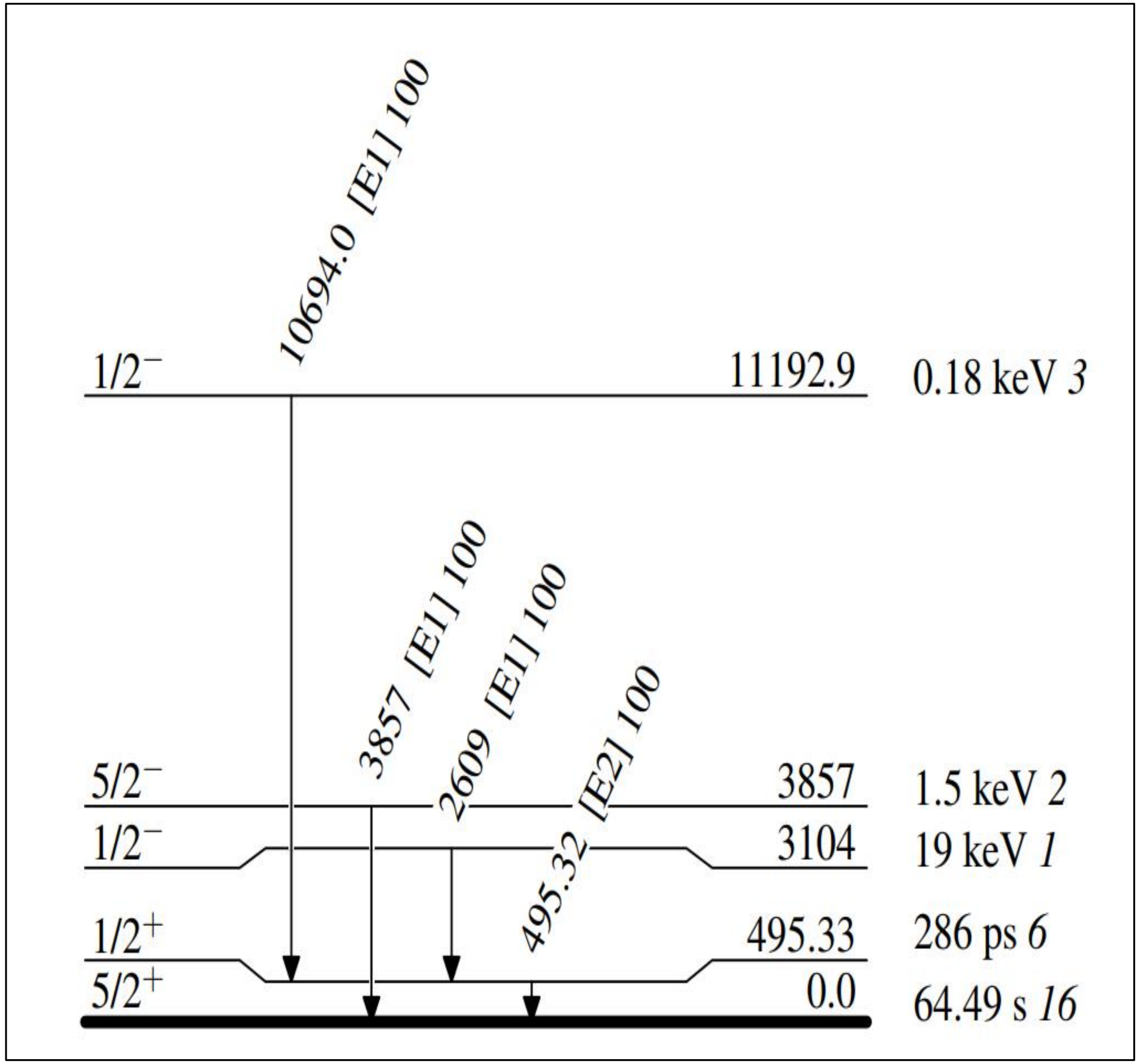
$1s^{2}_{2}\ 2s^{2}_{4}\ 2p^{6}_{10}\ 3s^{2}_{12}\ 3p^{6}_{18}\ 4s^{2}_{20}\ 3d^{10}_{30}\ 4p^{6}_{36}\ 5s^{2}_{38}\ 4d^{10}_{48}\ 5p^{6}_{54}\ 6s^{2}_{56}\ 4f^{14}_{70}\ 5d^{10}_{80}\ 6p^{6}_{86}\ 7s^{2}_{88}\ 5f^{14}_{102}\ 6d^{10}_{112}\ 7p^{6}_{118}$

Nuclear shell model

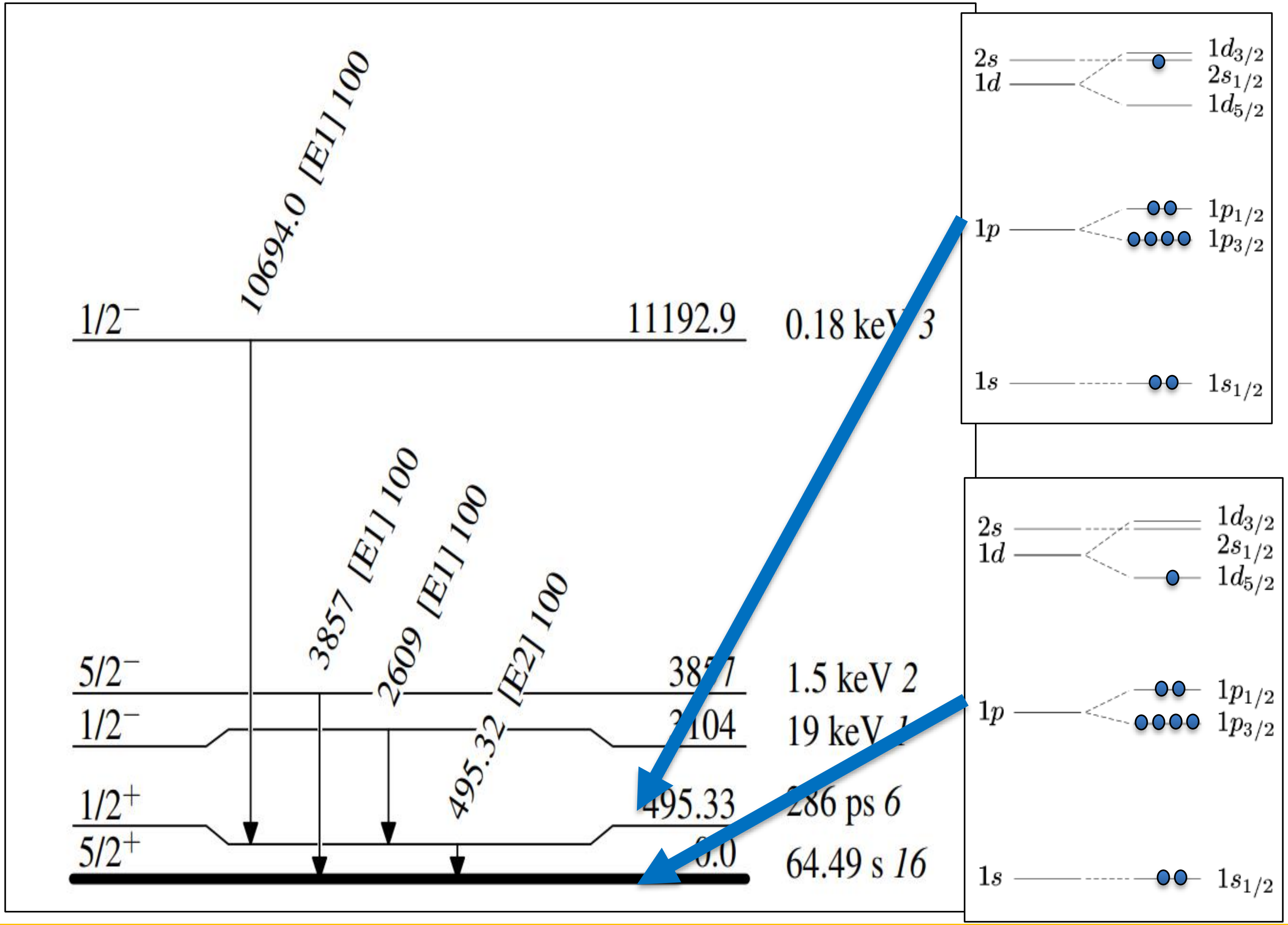




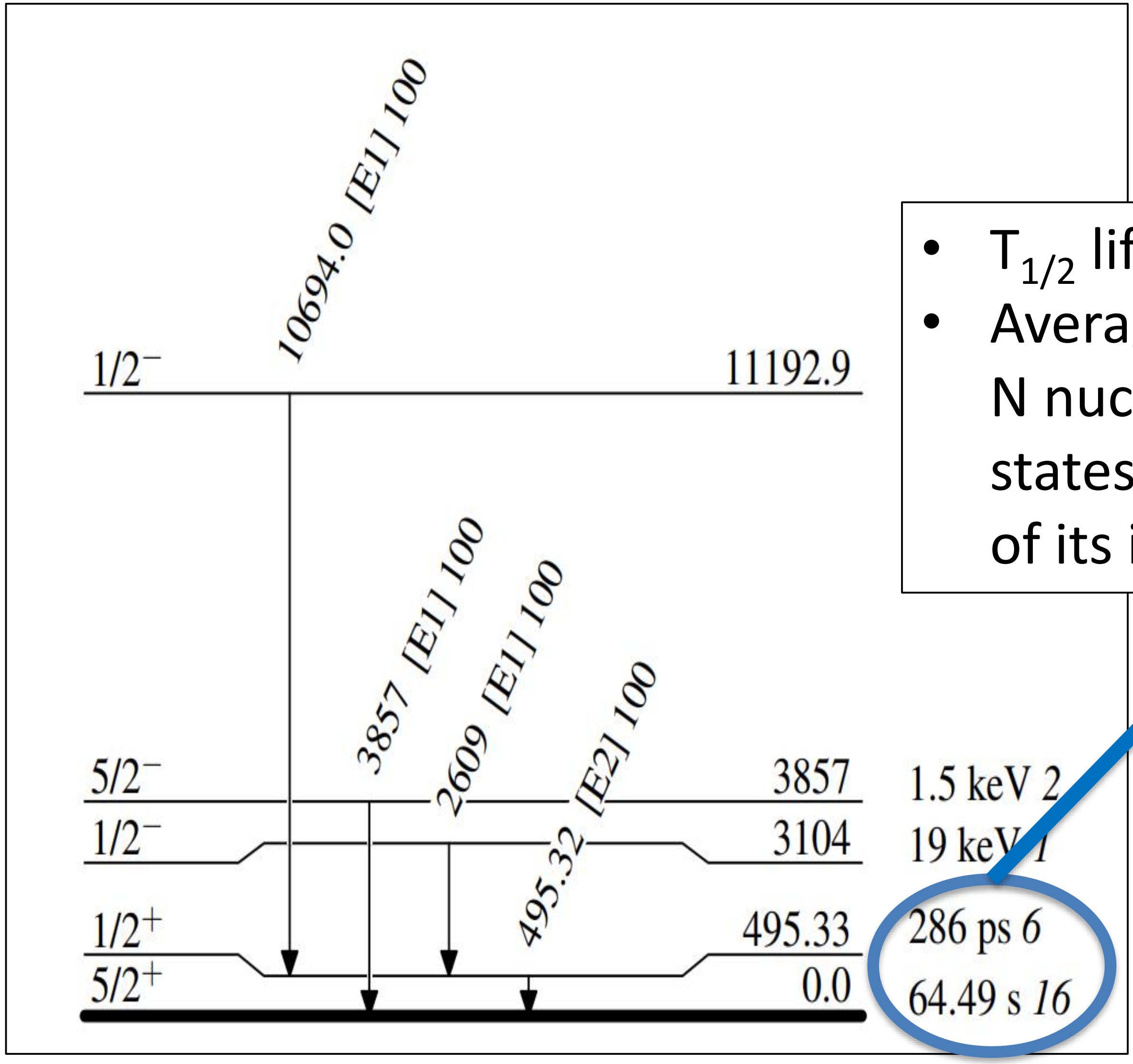
Nuclear level scheme



Nuclear level scheme

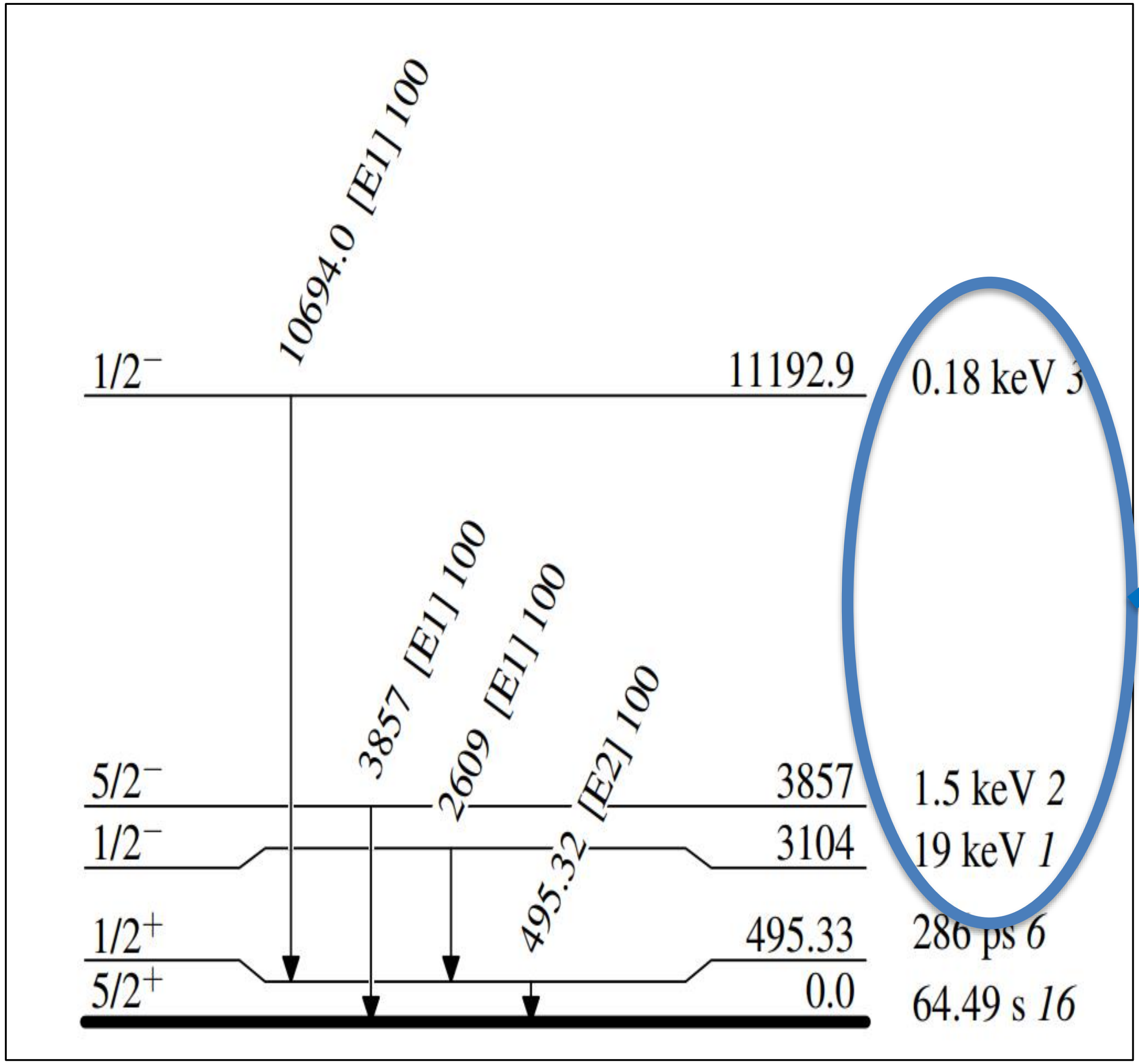


Nuclear level scheme

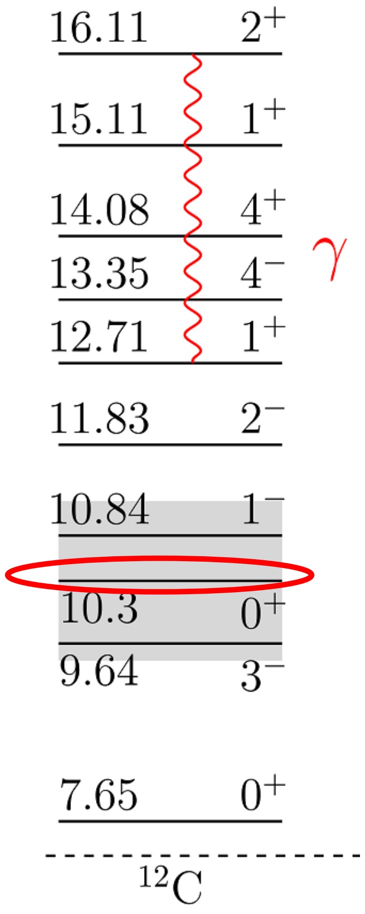


- $T_{1/2}$ lifetime or half-life.
- Average time it takes for N nuclei (or excited states) to decay to 50% of its initial value

Nuclear level scheme



- Level width, Γ
- $\Gamma = \hbar/\tau$



K L Laursen *et al* 2014 *J. Phys.: Conf. Ser.* **569** 012073

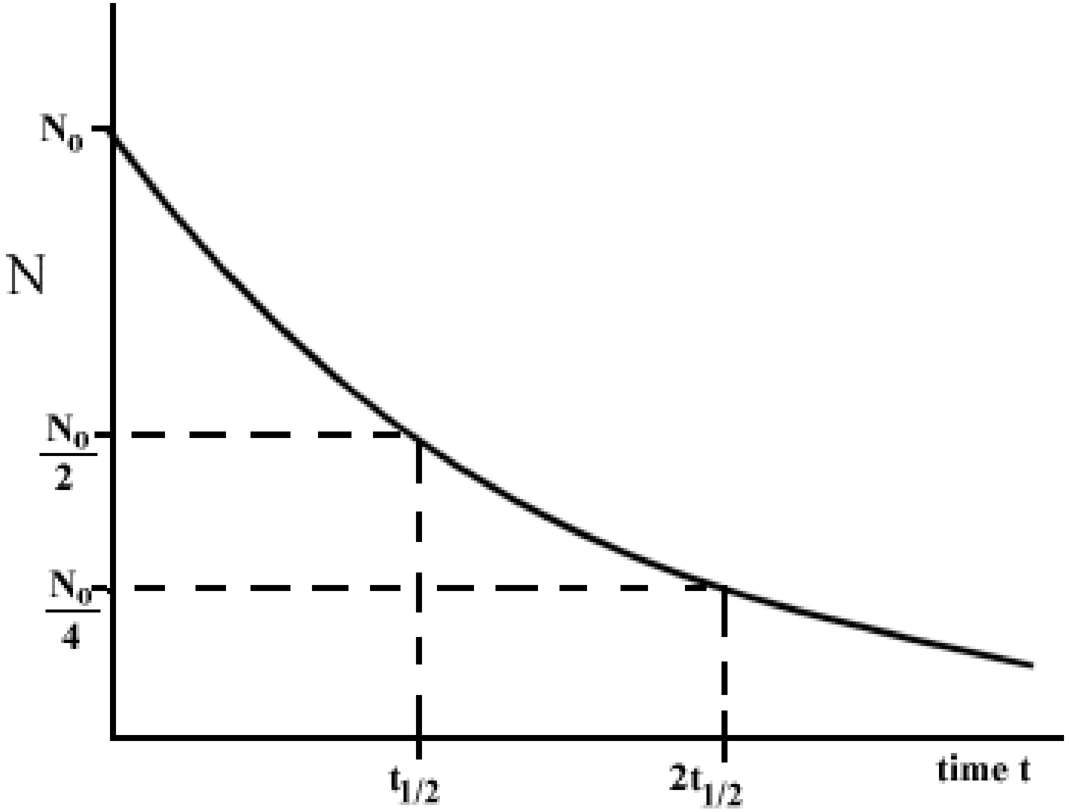
Nuclear half-life

- The activity of the sample changes as: $A(t) = A_0\, e^{-\lambda t}$
- From λ = decay constant, one can define $\tau = 1/\lambda$, the mean lifetime
- The time for half of the nuclei to decay is called the half-life:

$$t_{1/2} = \ln 2 / \lambda = \tau \ln 2$$

$$N(t_{1/2}) = N_0\, e^{-\lambda t} = N_0 e^{-\ln 2} = N_0/2$$

- Nuclear lifetime span over 35 orders of magnitude (from fs to Gy)



Transition strength

What does a state lifetime tell us?

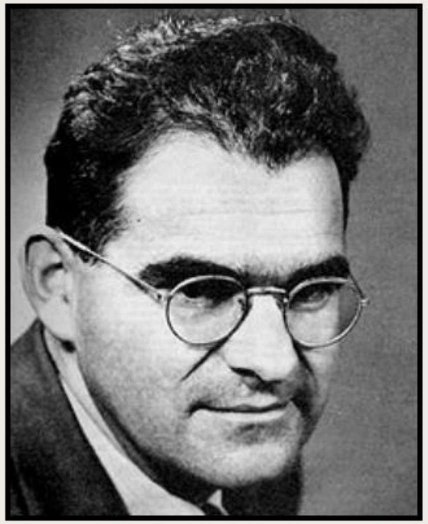
Internal transitions are electromagnetic, and the transition probability can be defined as

$$T_{if}(\lambda L) = \frac{8\pi(L+1)}{\hbar L((2L+1)!!)^2}\left(\frac{E_\gamma}{\hbar c}\right)^{2L+1} B(\lambda L; J_i \to J_f)$$

Which relates the transition probability (and therefore the lifetime) to the reduced transition matrix element, B(λL)

$$B\left({}^{E}_{M}\lambda, L_i \to L_f\right) = \frac{1}{2L_i+1}\left|\langle L_f \| M({}^{E}_{M}\lambda) \| L_i\rangle\right|^2$$

Weisskopf estimates



So what?

We can use this reduced matrix element to determine whether the transition is "single-particle" like

$$B(Wu:EL) = \frac{1.2^{2L}}{4\pi}\left(\frac{3}{L+3}\right)^2 A^{2L/3}e^2 fm^{2L}$$

$$B(Wu:ML) = \frac{10}{\pi}1.2^{2L-2}\left(\frac{3}{L+3}\right)^2 A^{2L-2}2\left(\frac{e\hbar}{2Mc}\right)^2 fm^{2L-2}$$

Weisskopf estimates

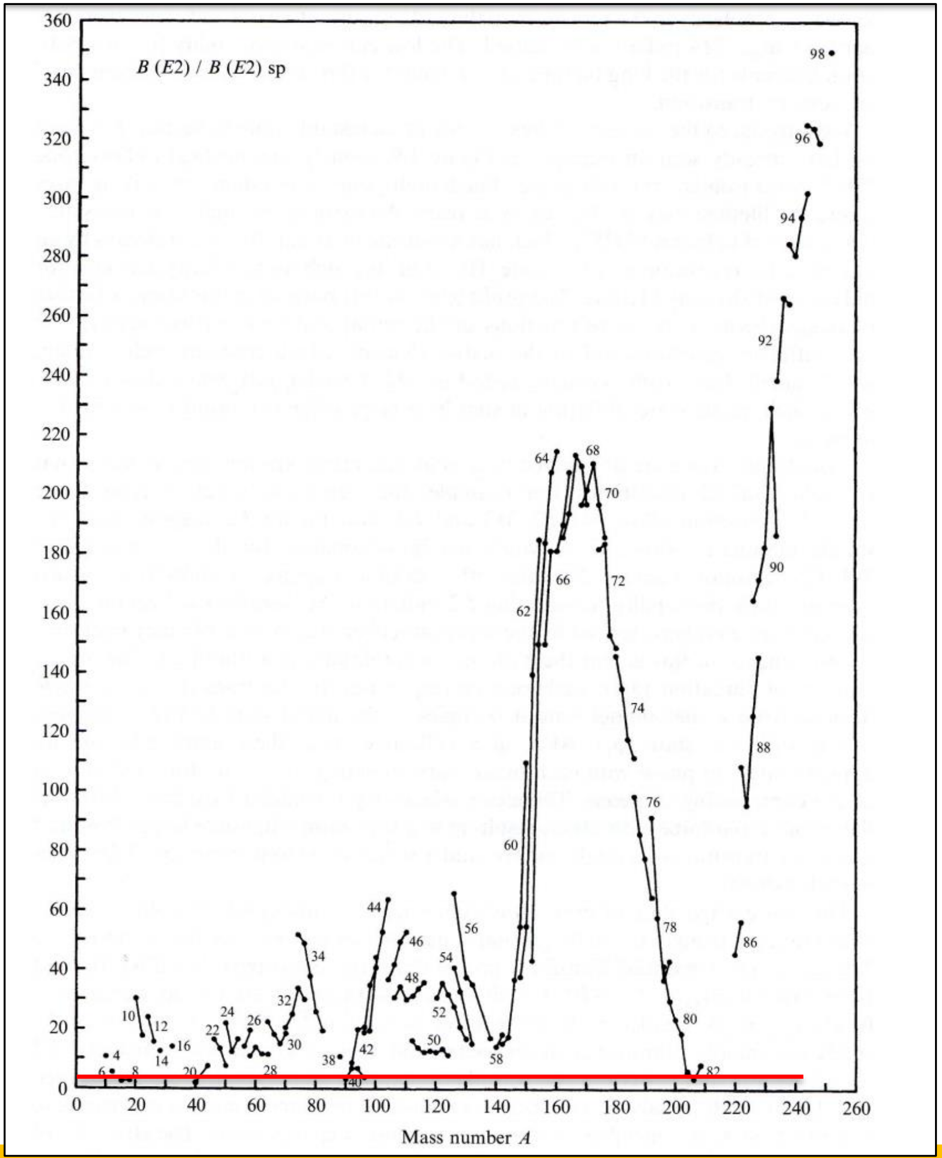
Table 1. Formulae for single-particle transition half-lives, corrected for internal conversion.

Electric	$t^{\gamma}_{1/2}$ (s)	Magnetic	$t^{\gamma}_{1/2}$ (s)
E1	$\frac{6.76\times10^{-6}}{E_\gamma^3 A^{2/3}}$	M1	$\frac{2.20\times10^{-5}}{E_\gamma^3}$
E2	$\frac{9.52\times10^{6}}{E_\gamma^5 A^{4/3}}$	M2	$\frac{3.10\times10^{7}}{E_\gamma^5 A^{2/3}}$
E3	$\frac{2.04\times10^{19}}{E_\gamma^7 A^{2}}$	M3	$\frac{6.66\times10^{19}}{E_\gamma^7 A^{4/3}}$
E4	$\frac{6.50\times10^{31}}{E_\gamma^9 A^{8/3}}$	M4	$\frac{2.12\times10^{32}}{E_\gamma^9 A^{2}}$
E5	$\frac{2.89\times10^{44}}{E_\gamma^{11} A^{10/3}}$	M5	$\frac{9.42\times10^{44}}{E_\gamma^{11} A^{8/3}}$

- Single particle estimates
- Depend on $T_{1/2}$, branching ratio, E and A (actually radius)
- All observables that can be measured
- Traditionally, $T_{1/2}$ is the hardest
- Strong dependence with E
  - The lower the energy → the much longer the lifetime
    - E_1= 100 keV → τ_1= 500 ps
    - E_2= 200 keV → τ_2= 15 ps

Single particles are not enough

- A transition can be described as a single particle (de)excitation when B(XL)~1 W.u.
- Very few nuclei follow this rule
- This means that we need more than one nucleon excitation to explain what is happening
- Collective motions and deformation



Recommended upper limits

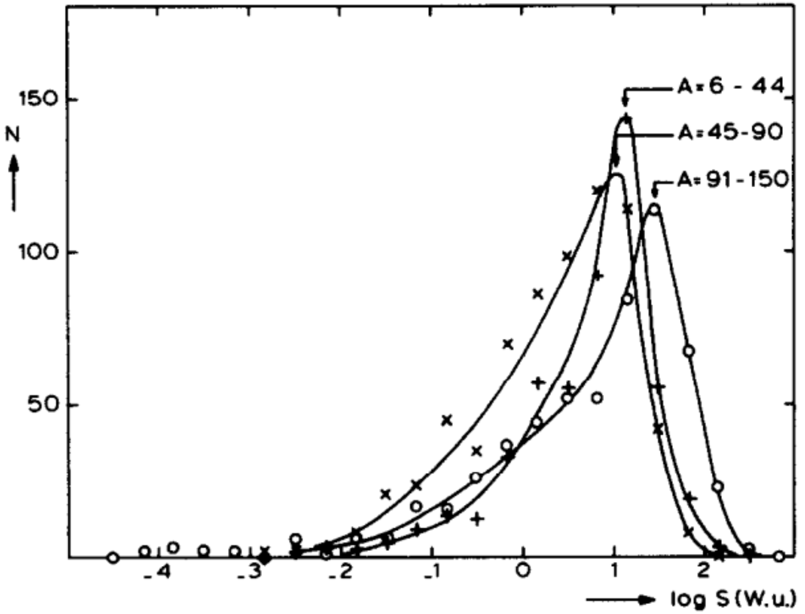


Fig. 5. Comparison of *E*2 strength distributions for different *A*-regions. Data for *A* = 6–44 and *A* = 45–90 are from Refs. 1 and 2, respectively.

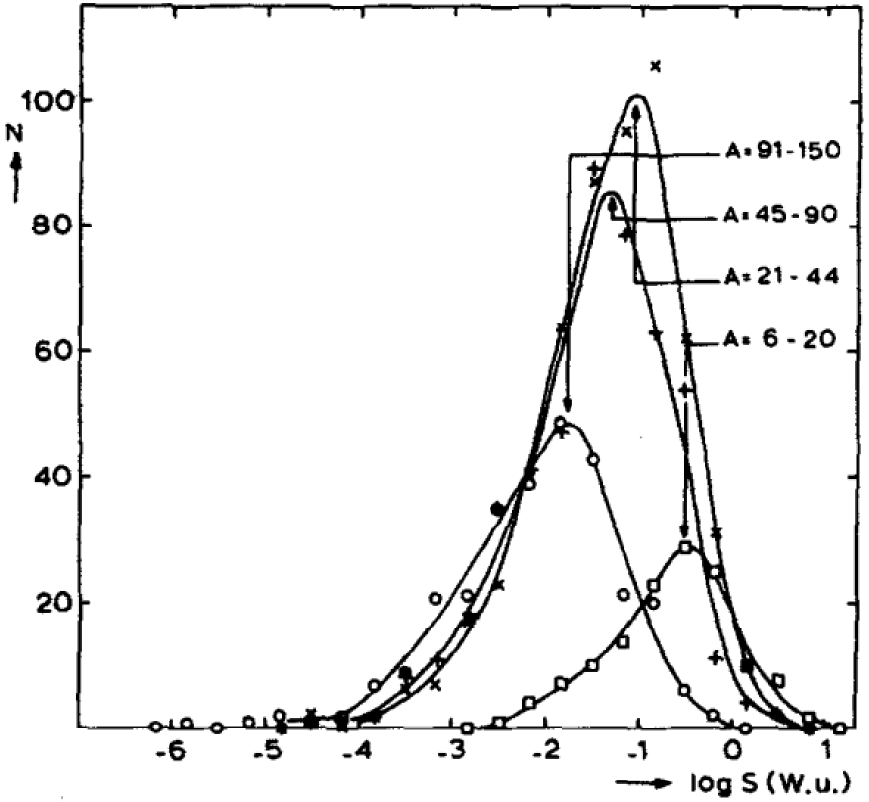


Fig. 6. Comparison of *M*1 strength distributions for different *A*-regions. Data for *A* = 6–44 and *A* = 45–90 are from Refs. 1 and 2, respectively.

Atomic Data and Nuclear Data Tables 26, 1, 1981

Recommended upper limits

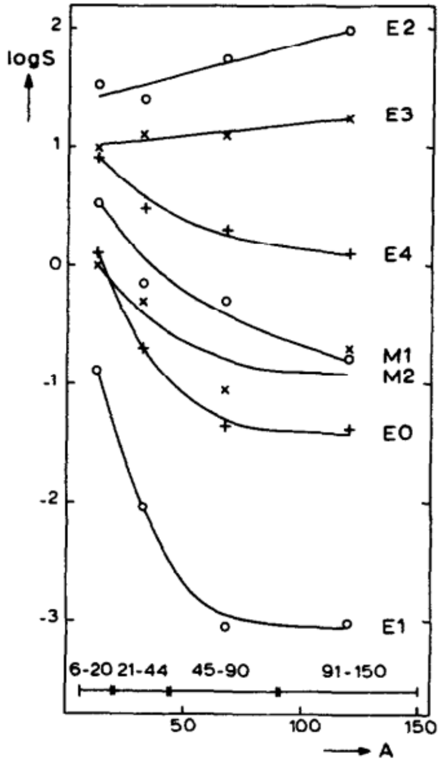


Fig. 7. Average strengths of strongest transitions as a function of A; for $E1$, $E2$, and $M1$ averages are taken of the strongest 10%, for $E3$ and $M2$ of the strongest 50%, and for $E0$ and $E4$ of all transitions. Strengths are expressed in W.u., except for $E0$ (Wi.u.). Data for A = 6–44 and A = 45–90 are from Refs. 1 and 2, respectively.

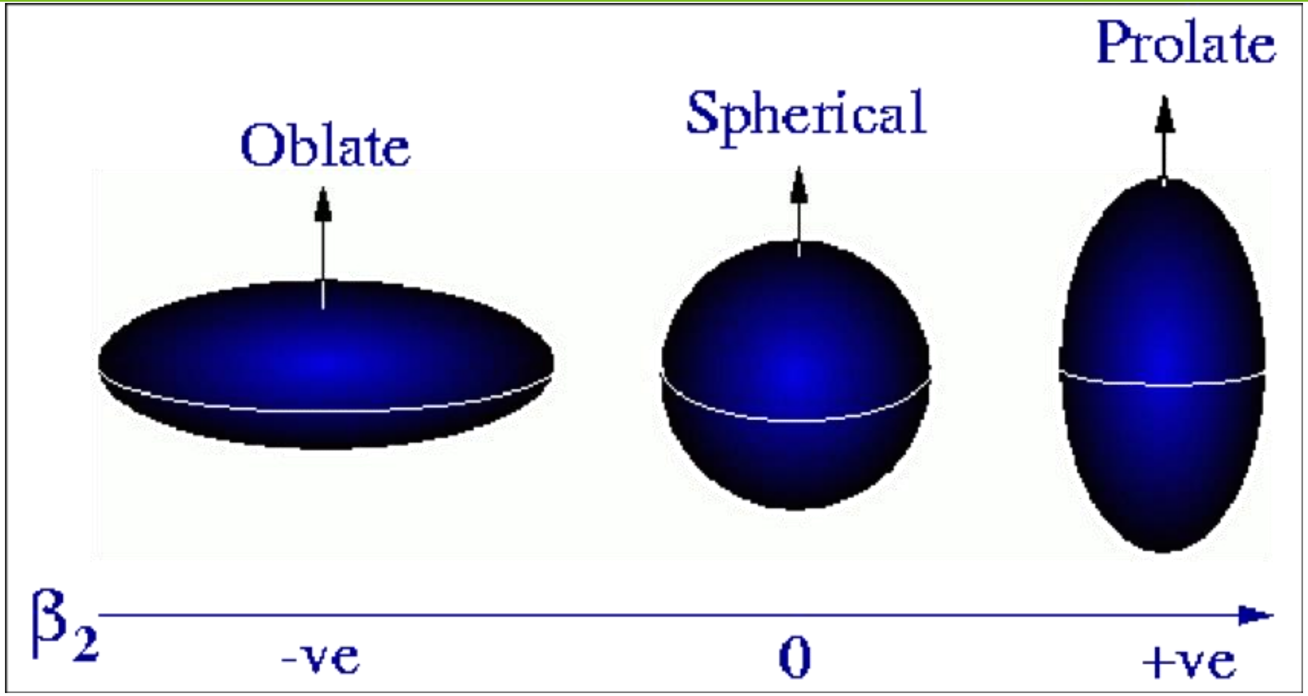
Table 2. $t_{1/2}$(W.u.)/$t_{1/2}$(exp) (Recommended Upper Limits)

Multipolarity	5≤A≤44[a]	45≤A≤90	91≤A≤150	A≥151
E1 (isovector)	0.5[b]	0.01	0.01	0.01
E2 (isoscalar)	100	300	300	1000
E3	50	100	100	100
E4	50	100	30	
M1 (isovector)	10[c]	3	3	2
M2 (isovector)	5	1	1	1
M3 (isovector)	10	10	10	10
M4		30	30	10

- By surveying a large number of transitions, RUL were proposed
- It is an orientation to assign multipolarity to transitions from measured B(XL)
- 40+ years old, could be outdated
- Currently being updated

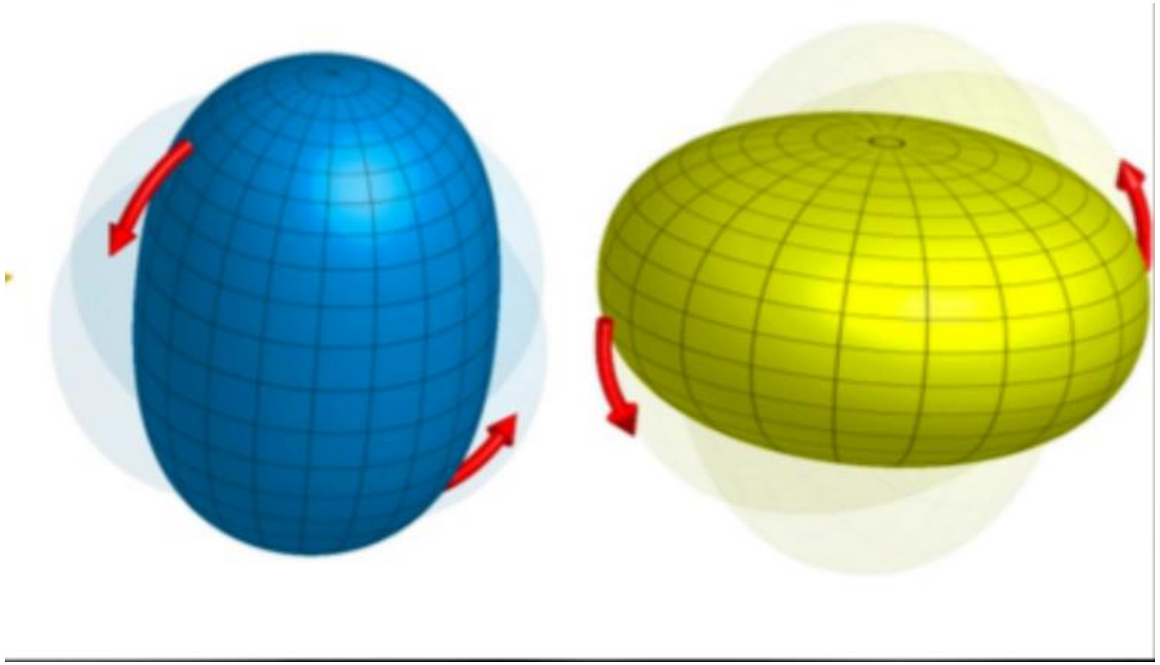
Atomic Data and Nuclear Data Tables 26, 1, 1981

Deformation

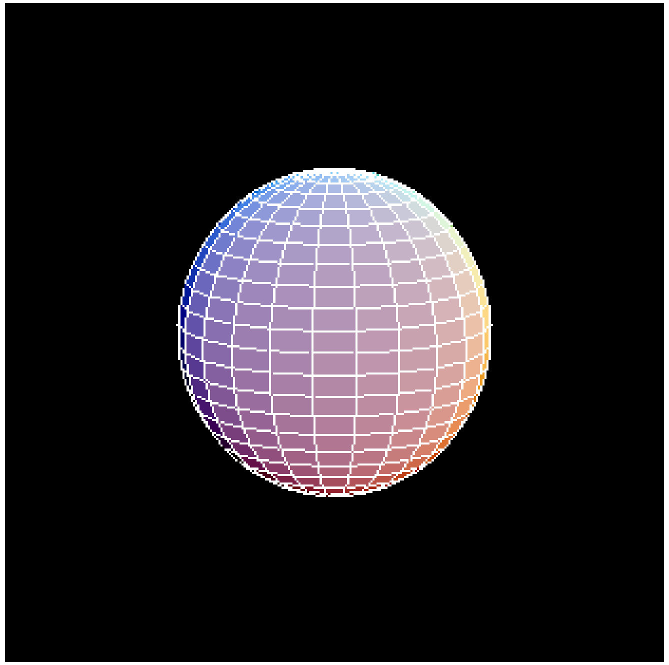


Collective motion

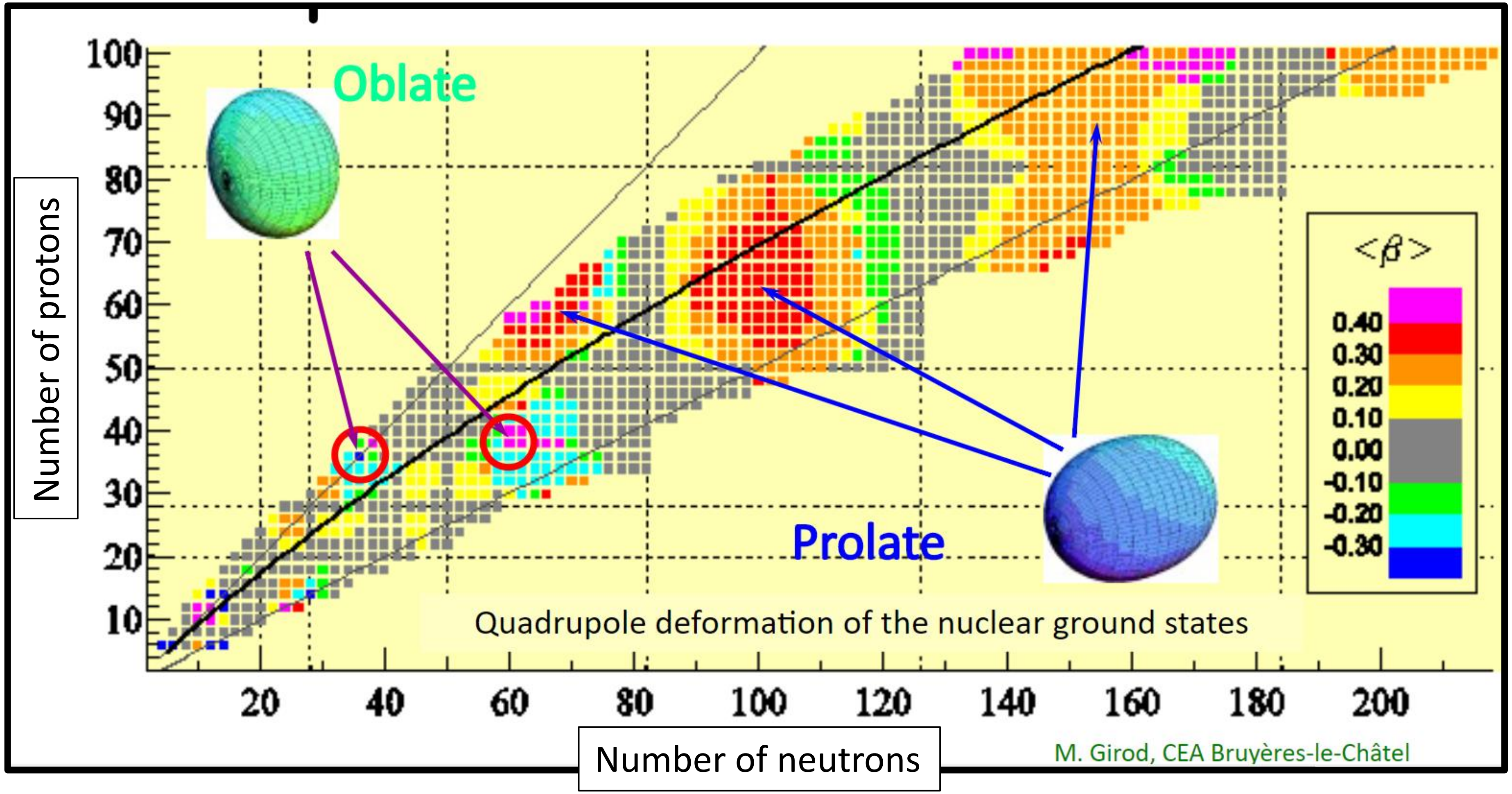
Rotation



Vibration



Deformation is a common phenomenon



Experimental nuclear fast timing

Experimental techniques

- Nuclear lifetimes span over 35 orders of magnitude!!!
- From below femtoseconds (10^{-15} s) to gigayears (~10^{20} s, or the age of the Universe)
- Each time range is studied with a different experimental technique

*Under very special circumstances

Technique	Lower limit	Upper limit
Chemical separation	Hours	∞^*
Electronic timing	10 ps (10^{-12})	∞^*
Doppler	10 fs (10^{-15})	10 ps (10^{-12})
Lineshape	1 fs (10^{-15})	100 fs (10^{-15})
Coulex	0^*	∞^*

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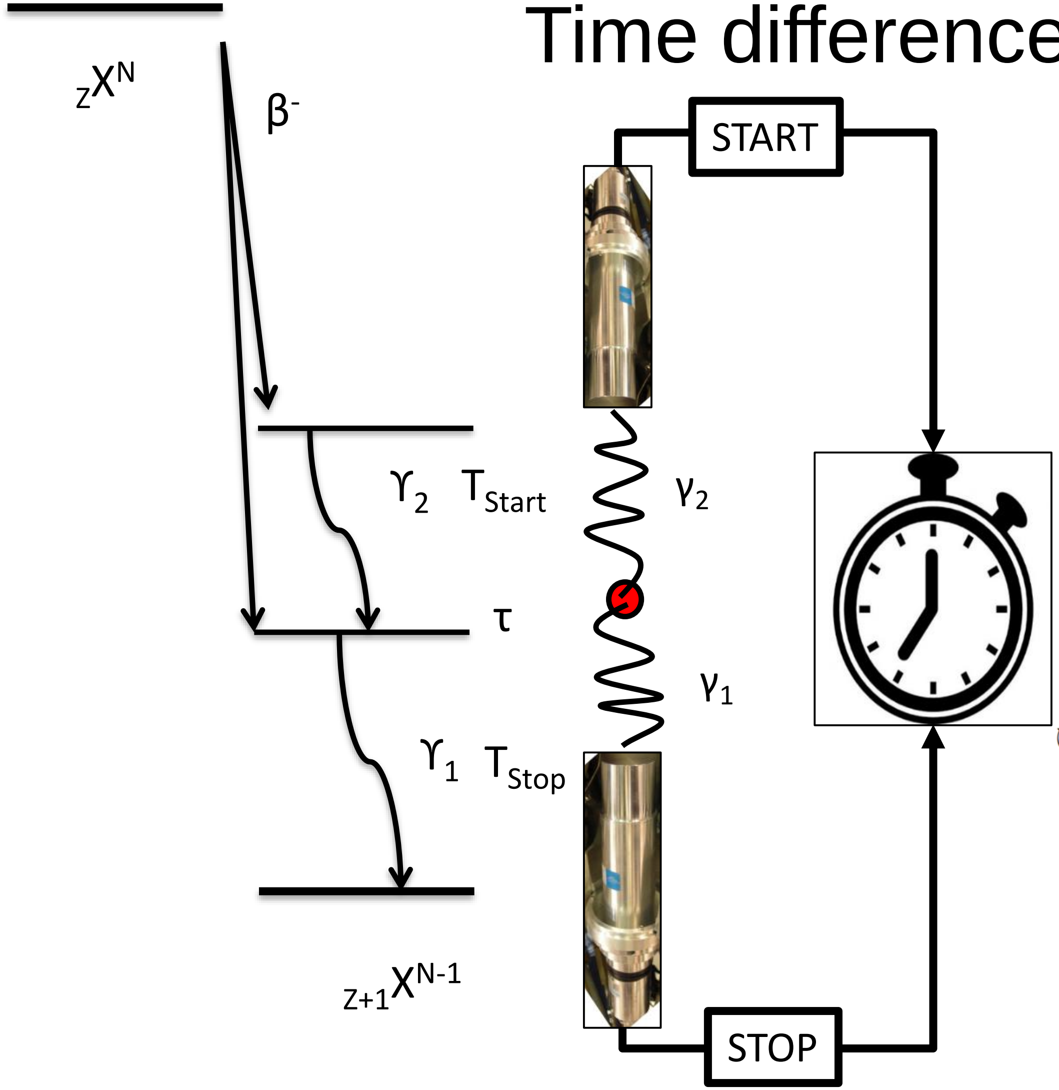
How short is a picosecond?

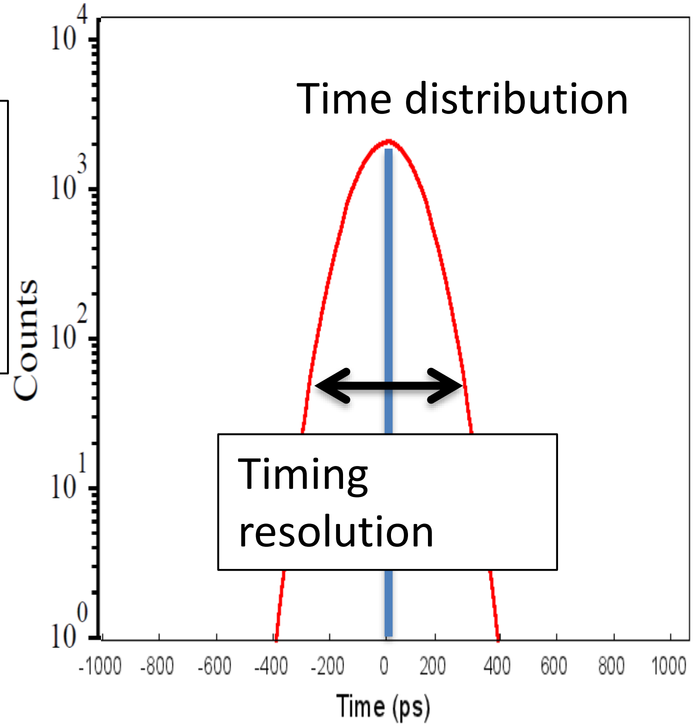
- 1 picosecond = 10^{-12} seconds
- That's 0.000000000001 seconds
- It takes photons (fastest particles in the universe) ~3.3 ps to travel 1 mm in vacuum
- When working in this time frame, the speed of light cannot be considered instantaneous anymore
- Indeed, *c* is one of the main limitations

Simplified nuclear electronic timing

Time difference

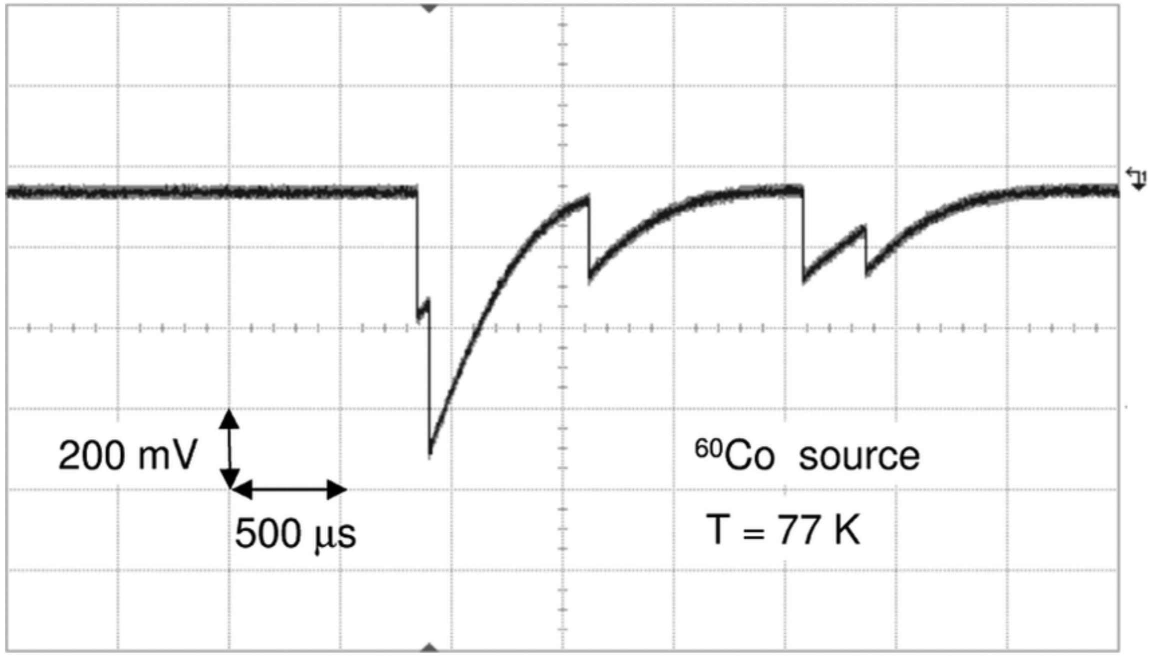


$\Delta T = T_{Stop} - T_{Start}$

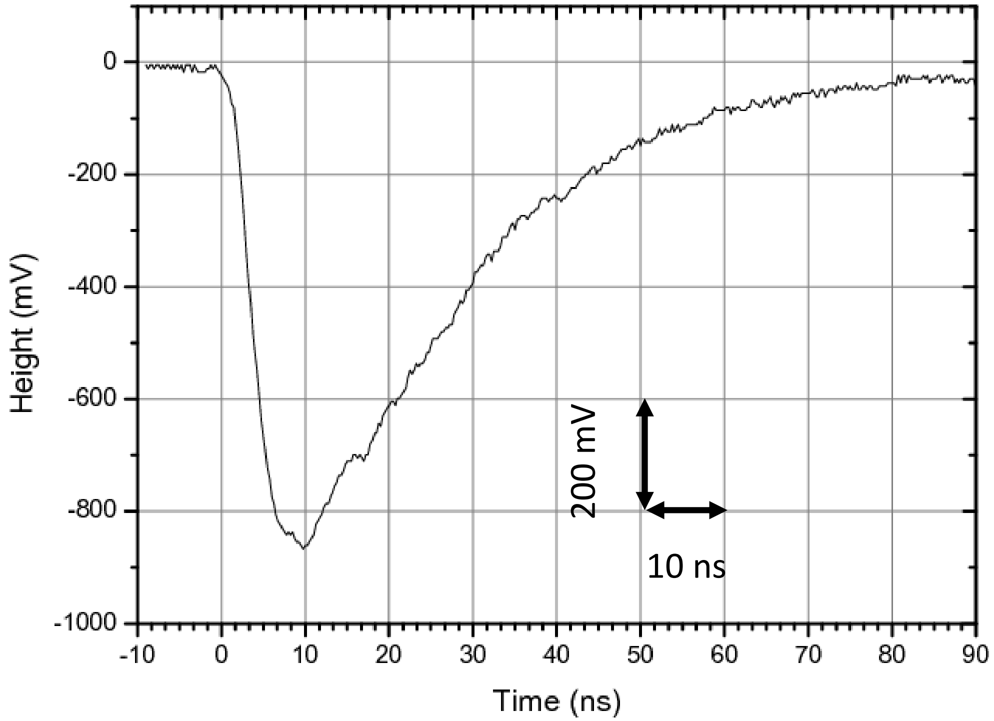


Signals

Semiconductor

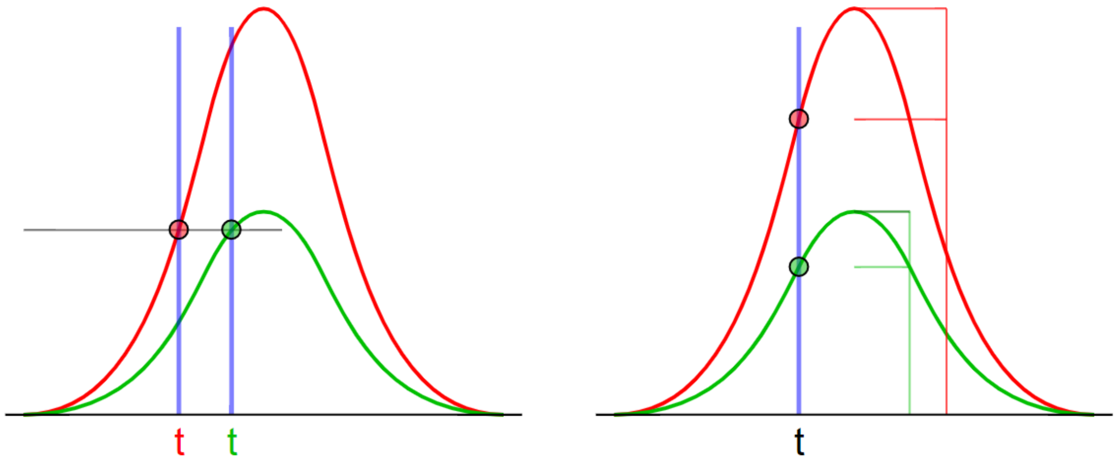


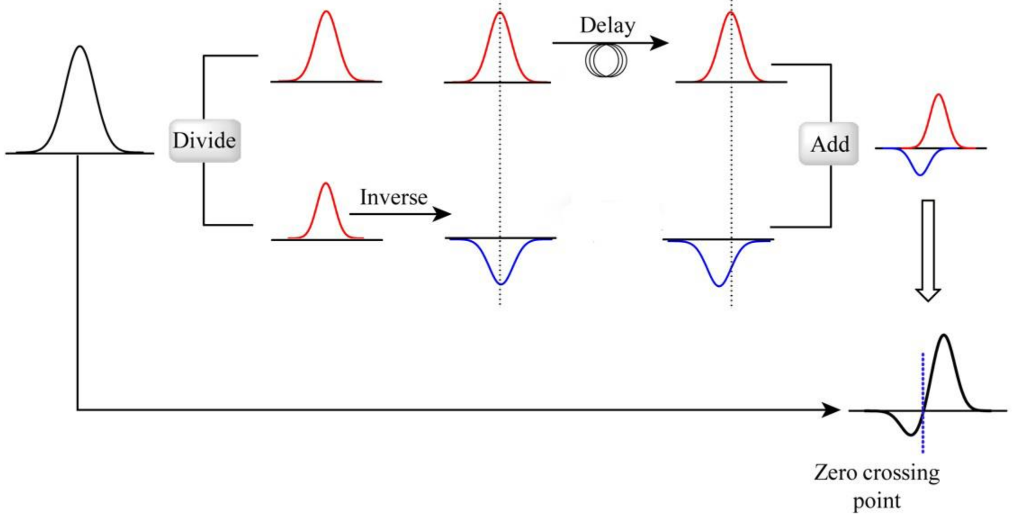
Scintillator



Technical Report for the Design, Construction and Commissioning of FATIMA, the FAst TIMing Array

Constant faction discriminator

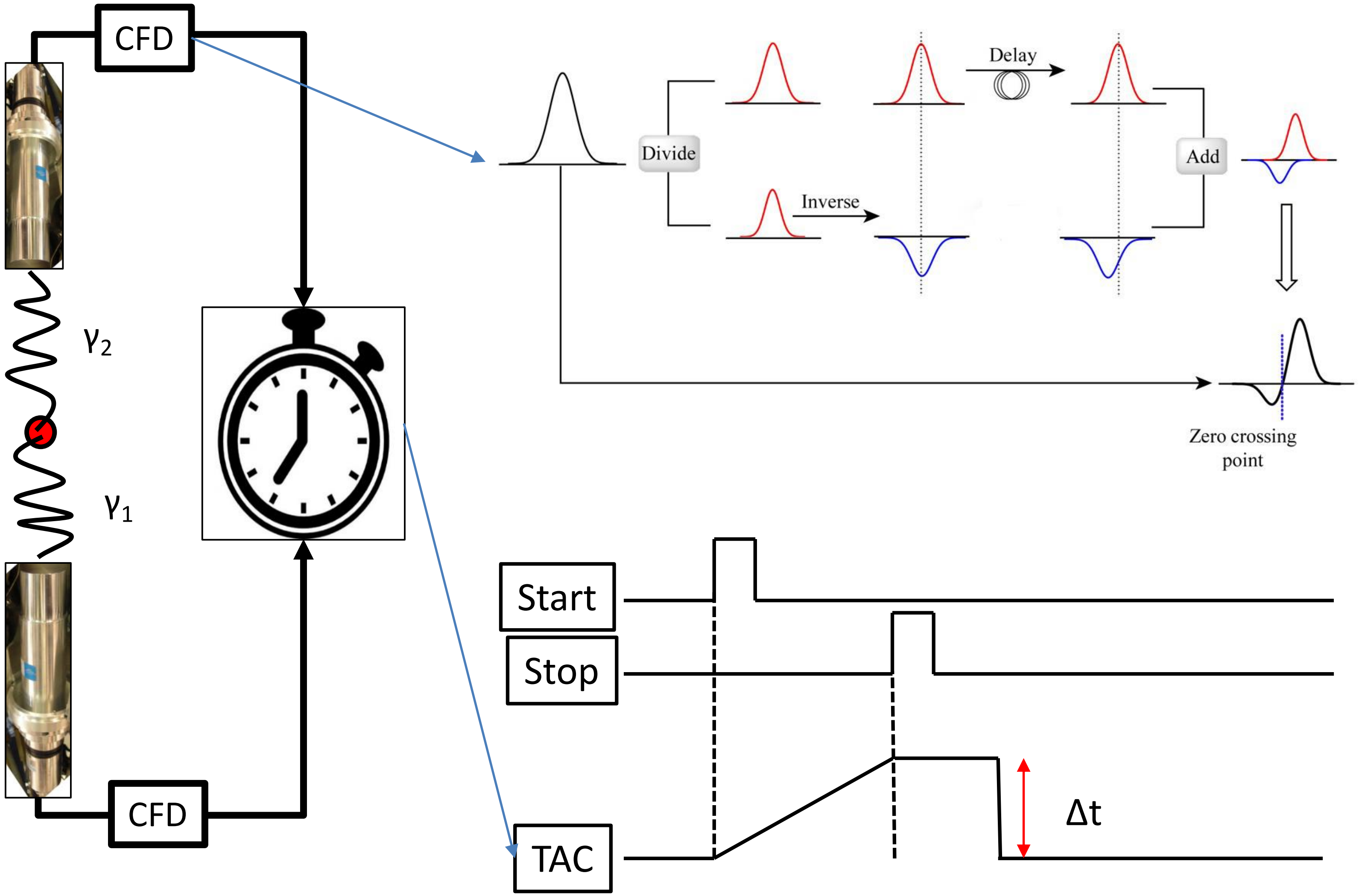




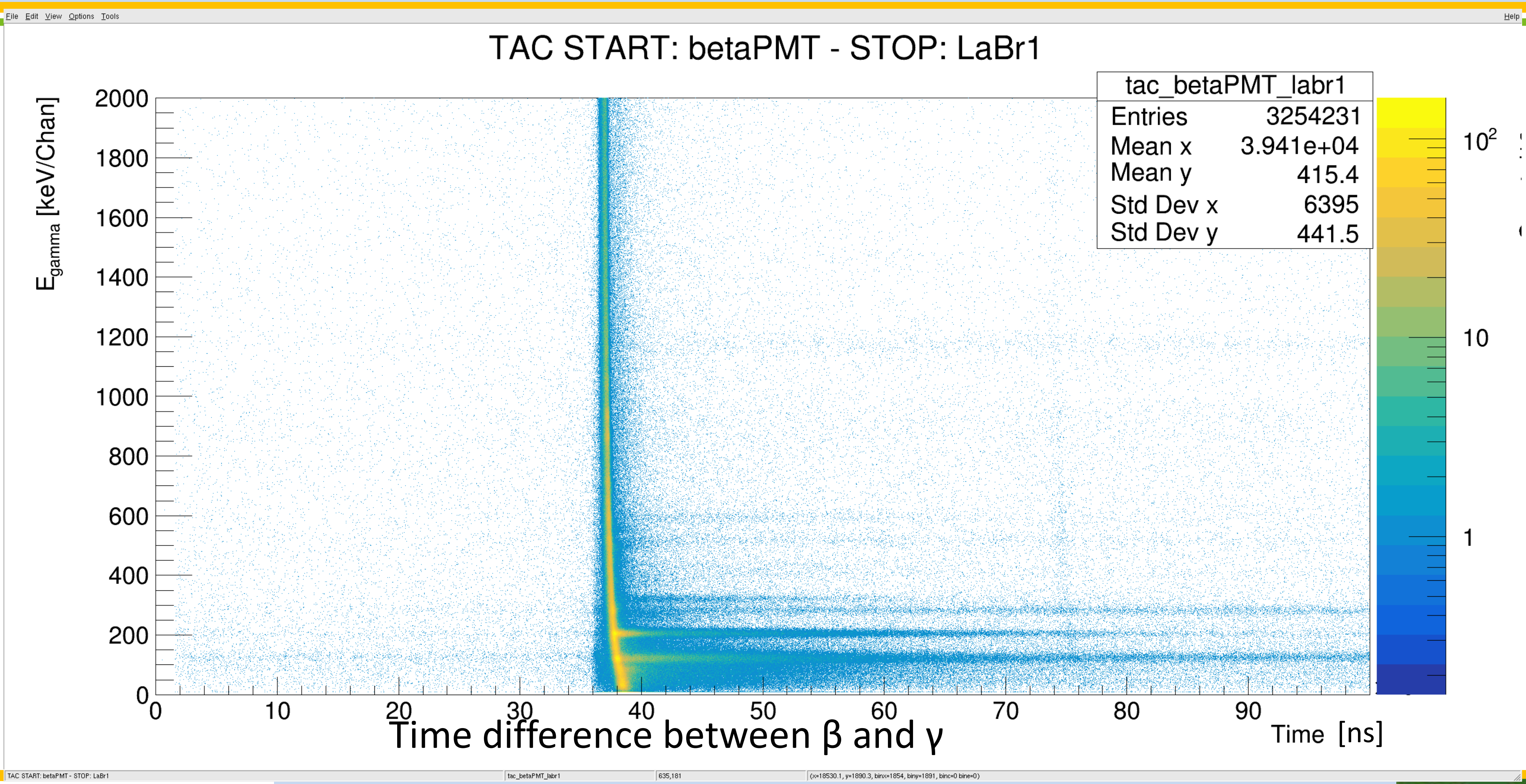
Wikicommons

Electronics

- **CFD:** constant fraction discriminator
- **TAC:** Time to amplitude converted

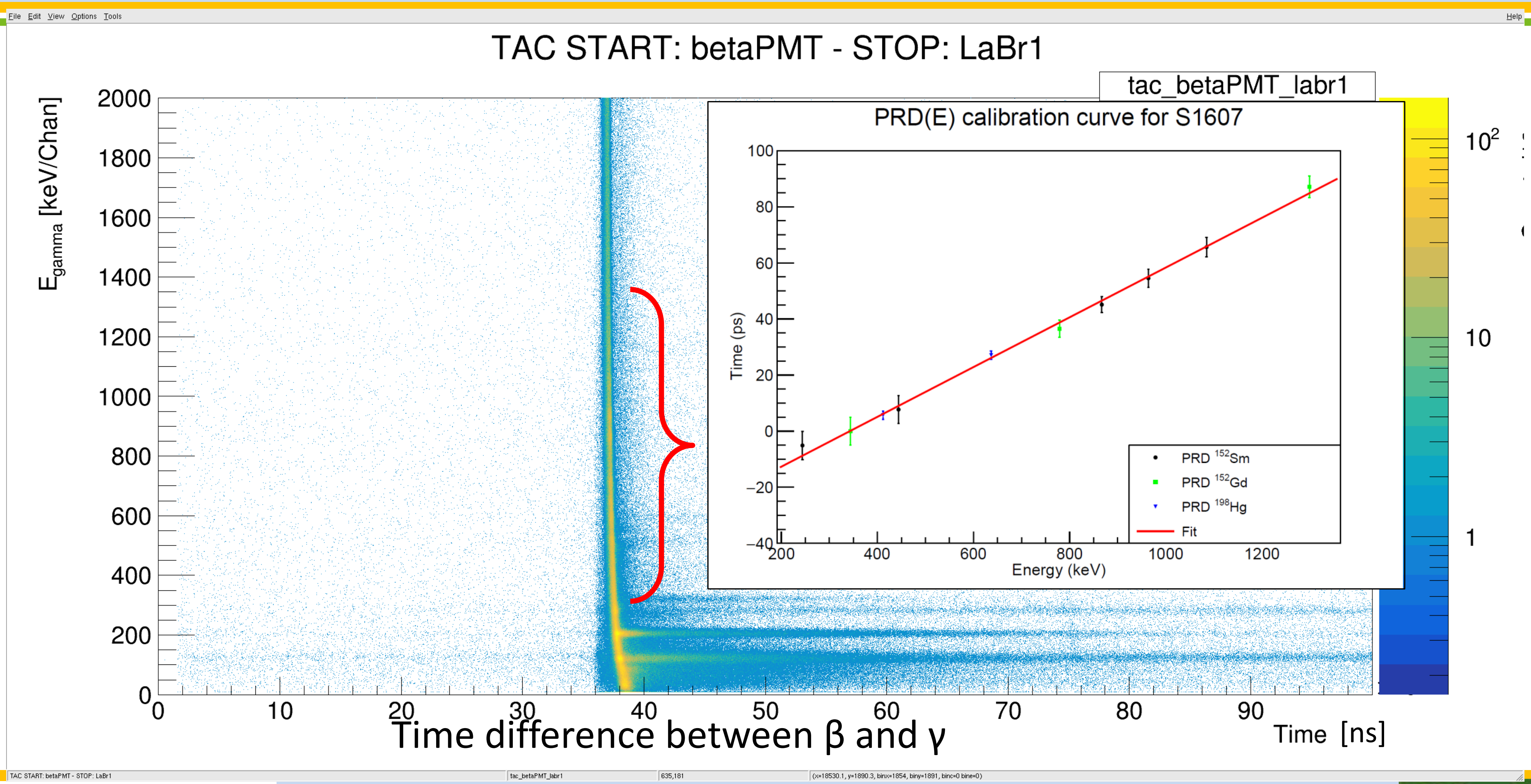


Time walk

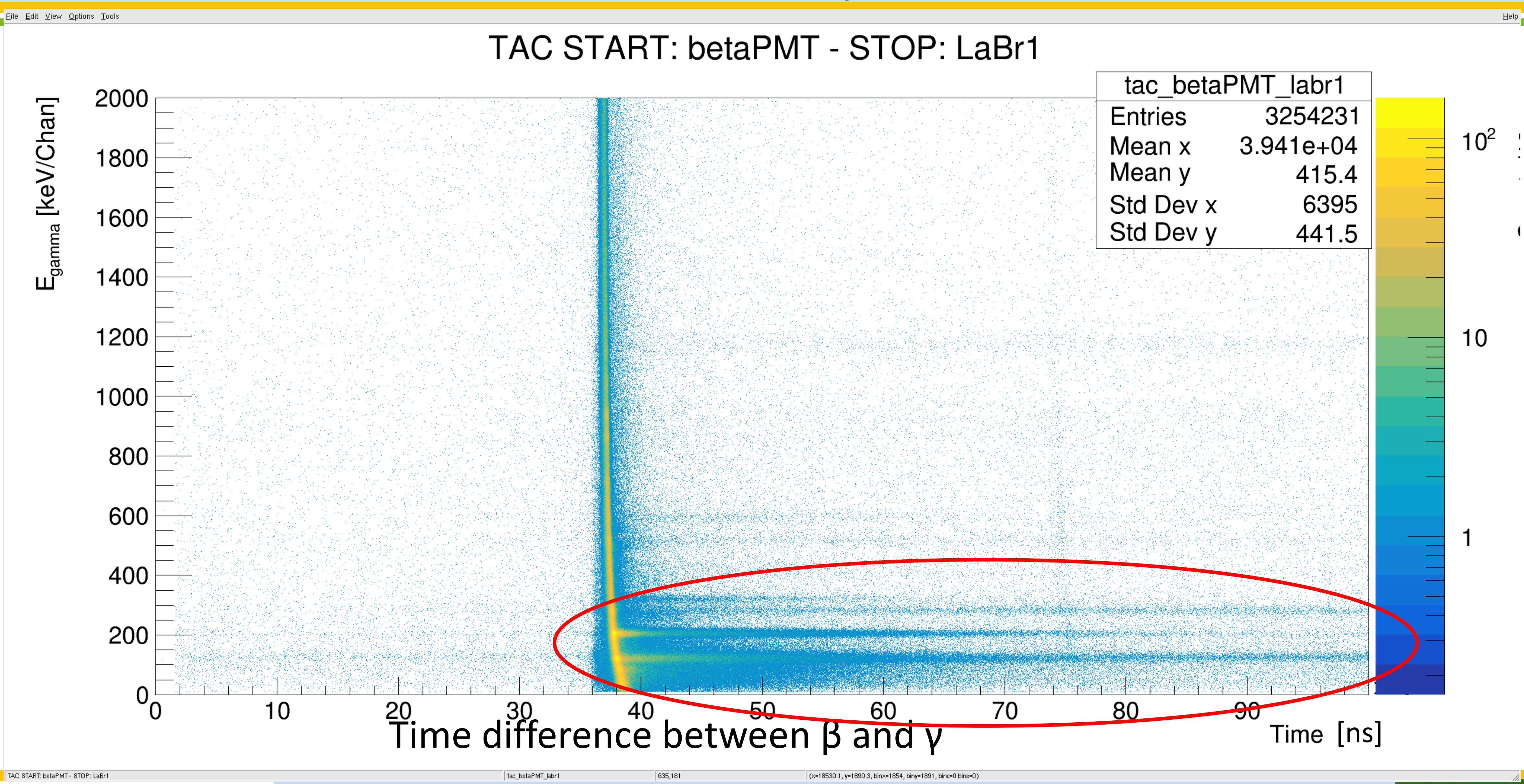


Time difference between β and γ

Time walk

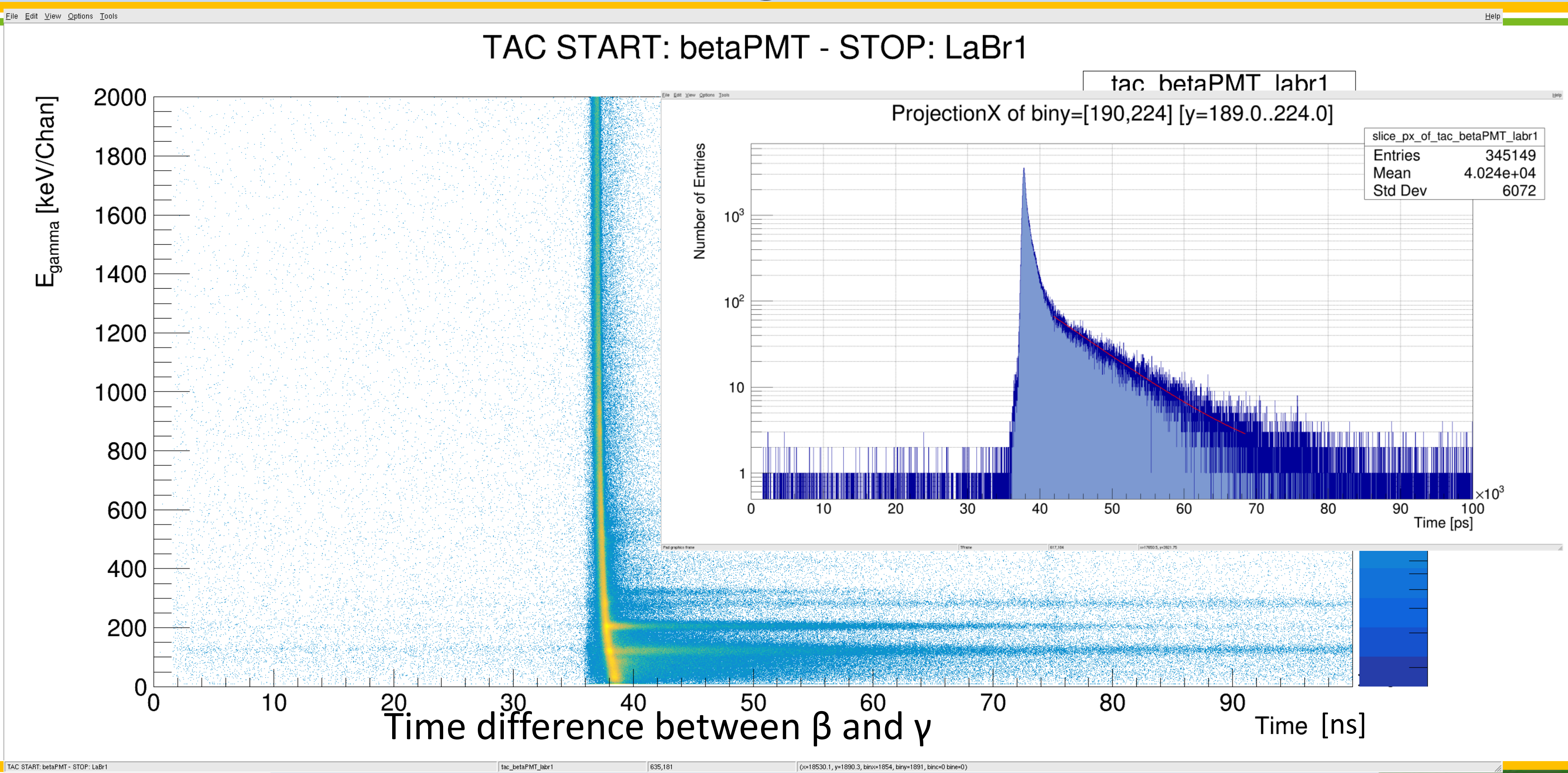

File Edit View Options Tools
Help
TAC START: betaPMT - STOP: LaBr1
tac_betaPMT_labr1
E_{gamma} [keV/Chan]
Time [ns]
PRD(E) calibration curve for S1607
Time (ps)
Energy (keV)
PRD ^{152}Sm
PRD ^{152}Gd
PRD ^{198}Hg
Fit
Time difference between β and γ

Bonus question



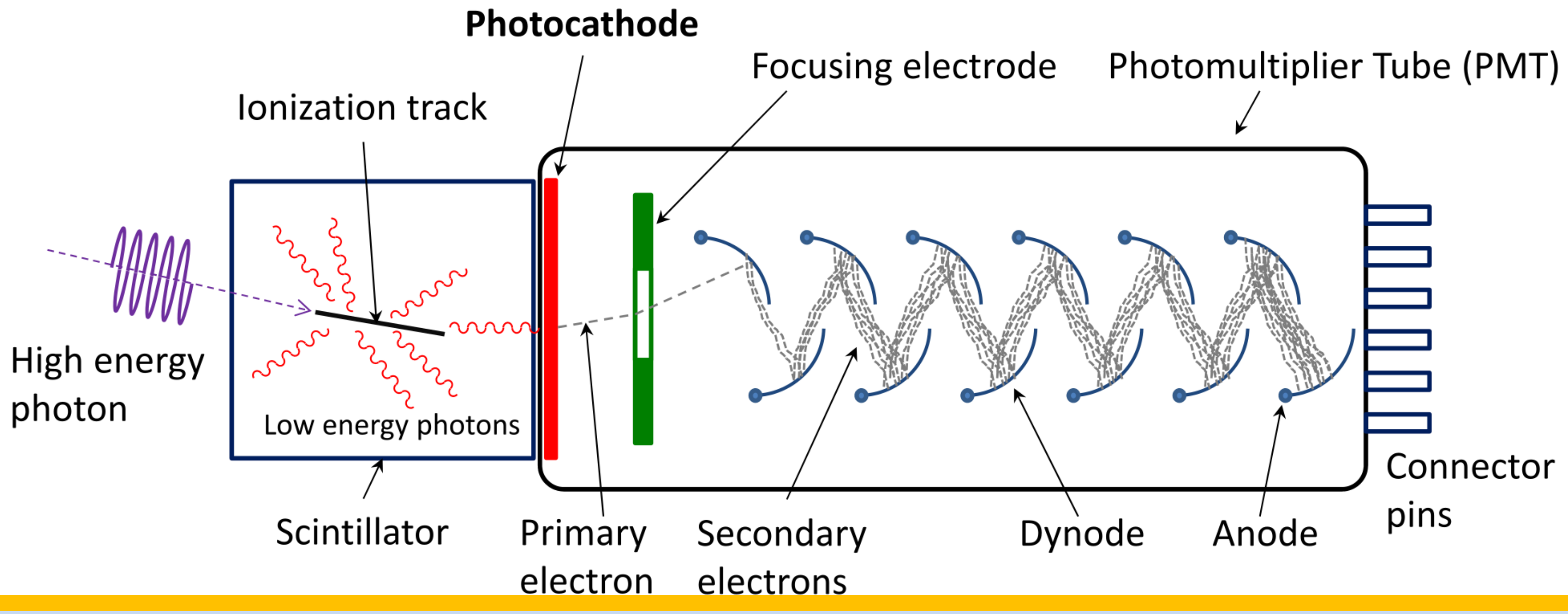
Time difference between β and γ

Long lifetime



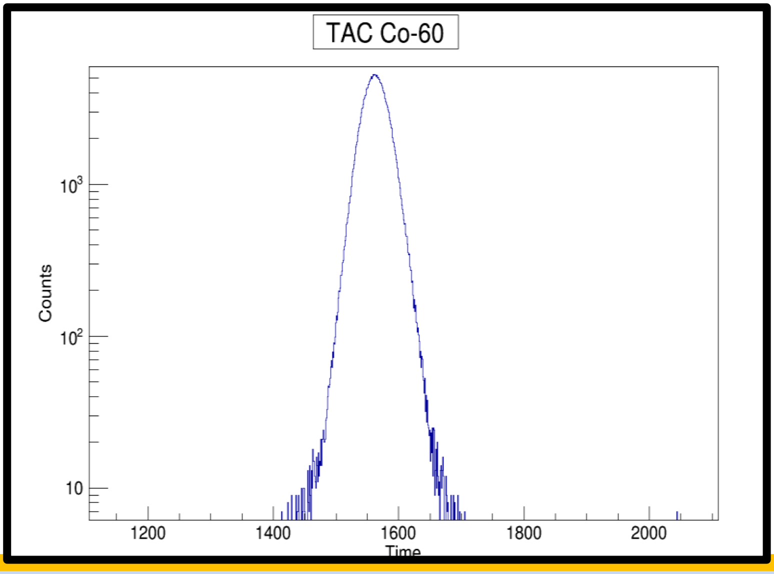
Measuring gamma-rays

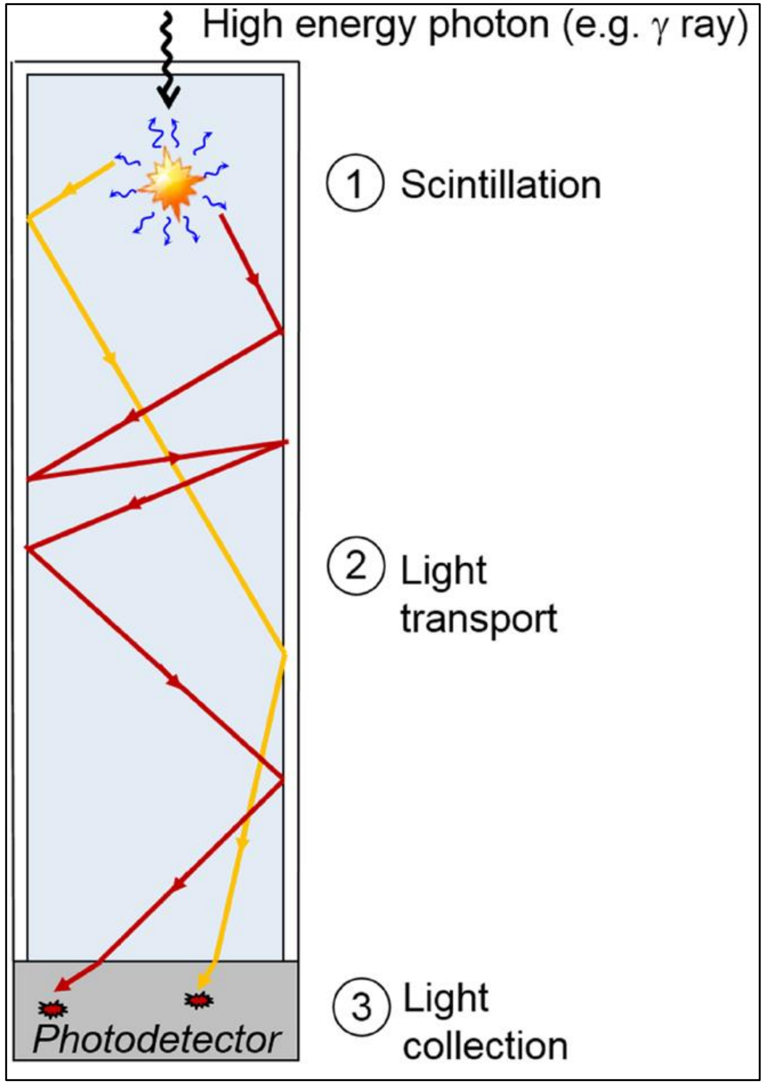
- Scintillators (like $LaBr_3(Ce)$) are the fastest detectors nowadays
- The incident photon excites the crystal molecules
- They quickly de-excite emitting UV
- The electrons from the photoelectric effect give us the signal
- Photoelectric effect is more likely with UV rays



Timing resolution

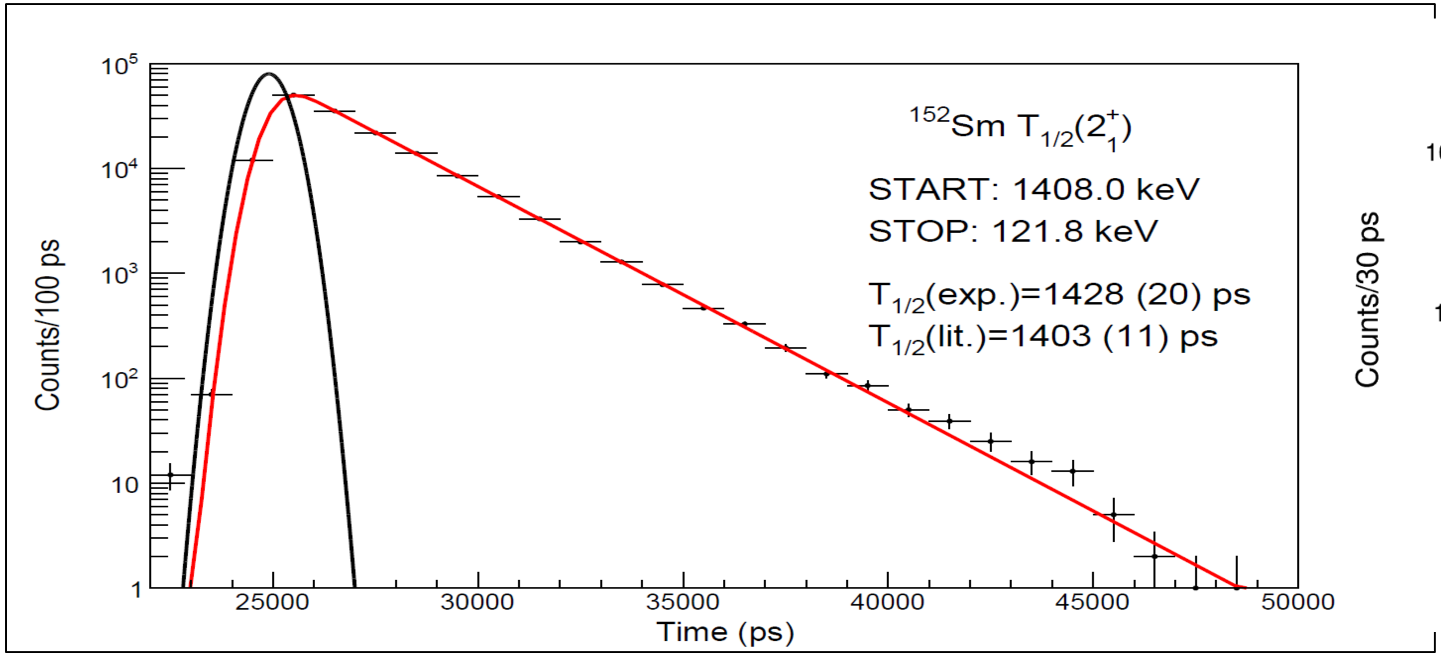
- Timing resolution is the time-width of a prompt (t~0 ps) signal
- Scintillators crystals are a few cm long and wide
- Light takes time bouncing inside the crystal
- Timing resolution mainly depends on crystal size

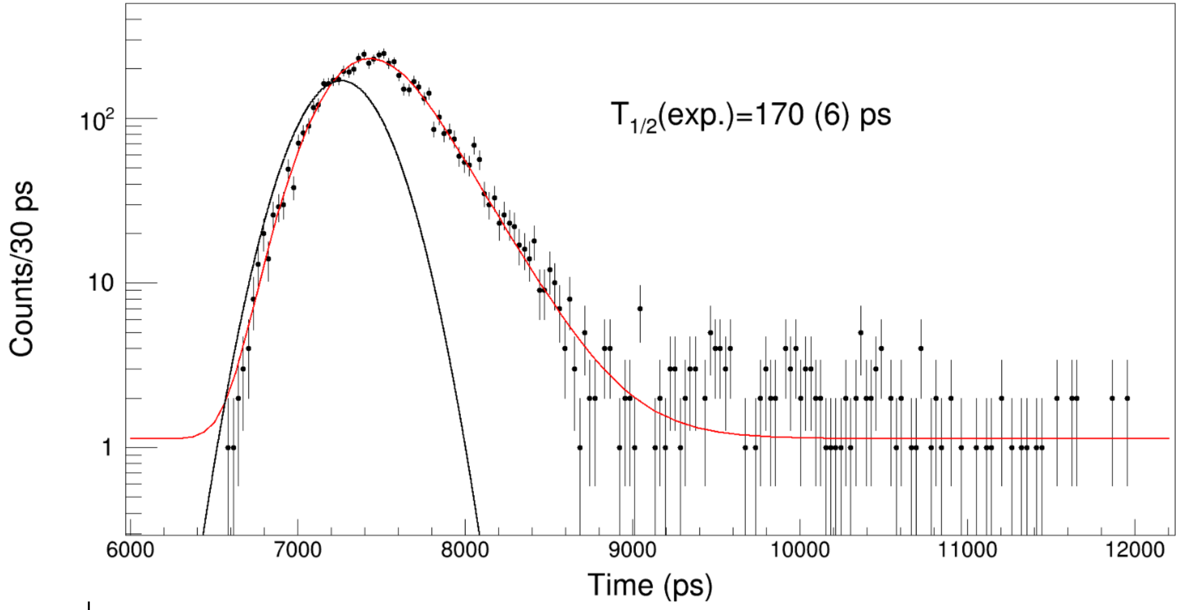




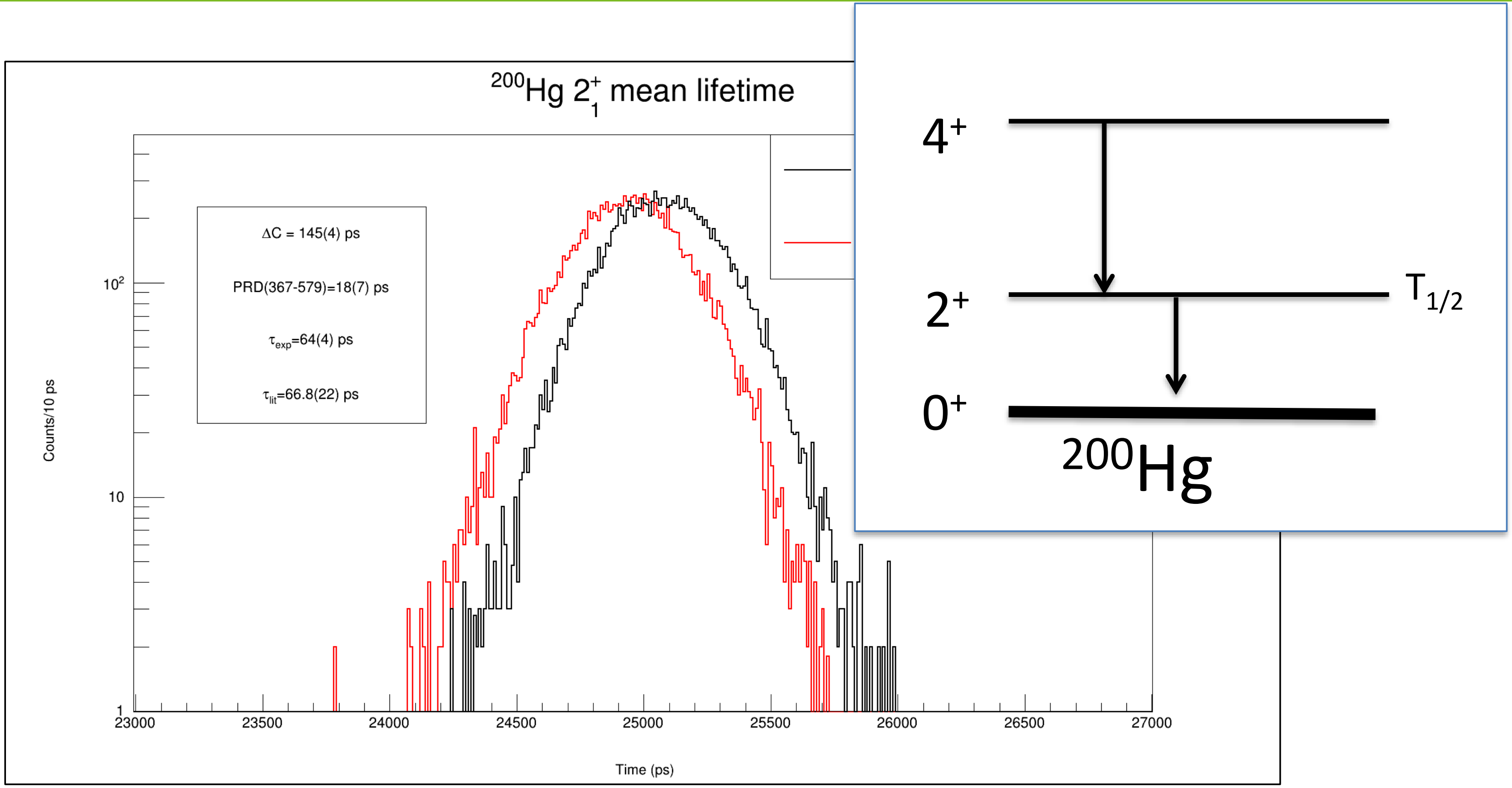
Convolution method

- Prompt part may be approximated to a Gaussian shape, down to 3 orders of magnitude
- Slope method may be used for $T_{1/2}$ ~ timing resolution
- Fit of the timing distribution to a prompt response plus an exponential decay
- $F(t_j) = \gamma \int_A^{+\infty} e^{-\delta(t_j - t)} e^{-\lambda(t-A)} dt$





Centroid shift method



2^+ state in ^{200}Hg

$T_{1/2}$(literature)=46.4(4) ps

$T_{1/2}$(experiment)=44(3) ps

Generalized centroid difference method

The centroid of the time distribution will be given by:

$C = \tau_0 + \tau_{START} + \tau_{STOP} + \tau_{level}$

τ_0=a constant delay of the setup, cannot be determined (in general)

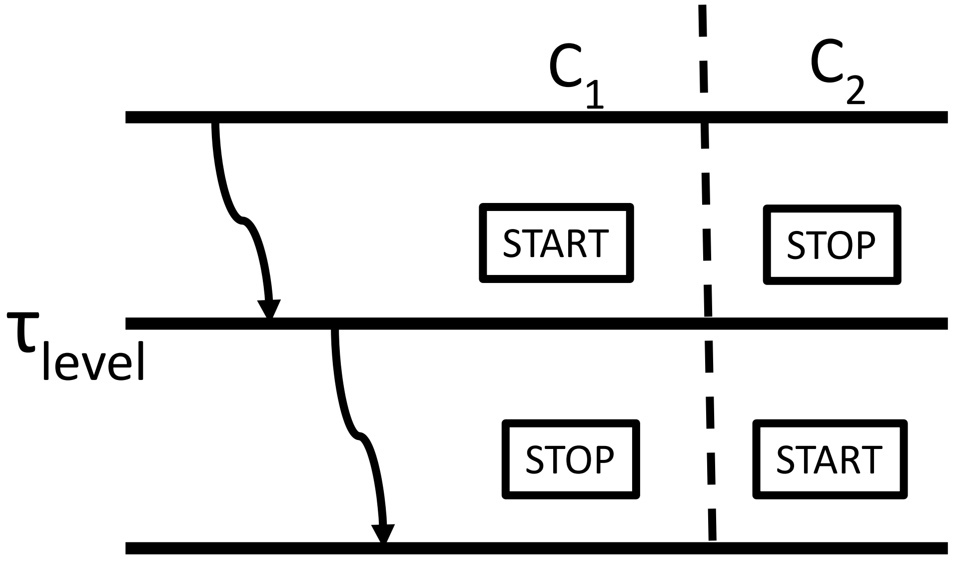
τ_{START}= time walk of the START detector (depends on the measured energy)

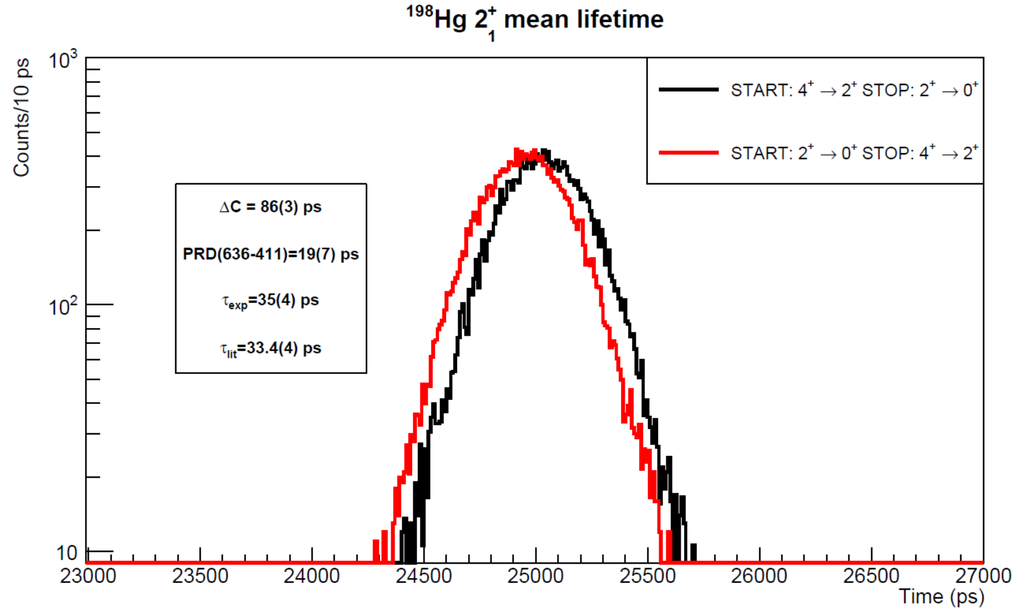
τ_{STOP}= time walk of the STOP detector (depends on the measured energy)

τ_{level}= Lifetime we want to measure

Thus, if we calibrate the walk of our detectors, we can do a centroid shift to cancel τ_0

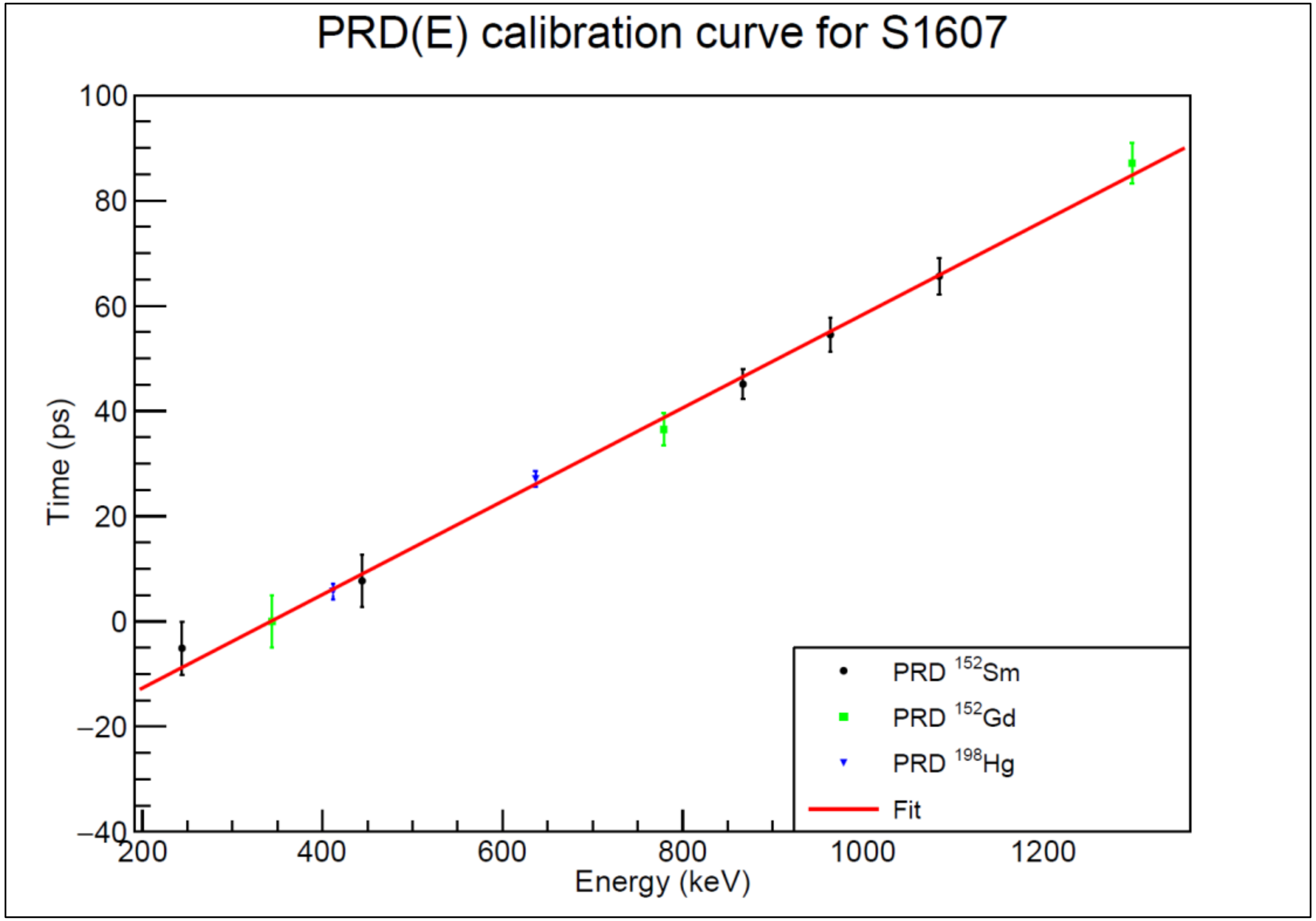
$$C_1 = \tau_{level} + \tau_0 + \tau_{WALK}(\gamma_1)$$
$$C_2 = -\tau_{level} + \tau_0 + \tau_{WALK}(\gamma_2)$$
$$\Delta C = C_1 - C_2 = (\tau_{WALK}(\gamma_1) - \tau_{WALK}(\gamma_2)) + 2\tau_{level}$$
$$\Delta C = 2\tau_{level} + PRD(\Delta E)$$



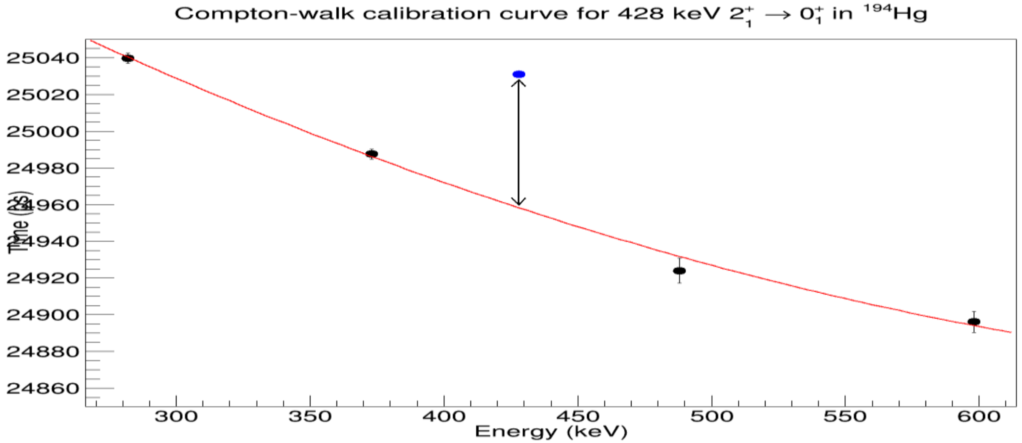


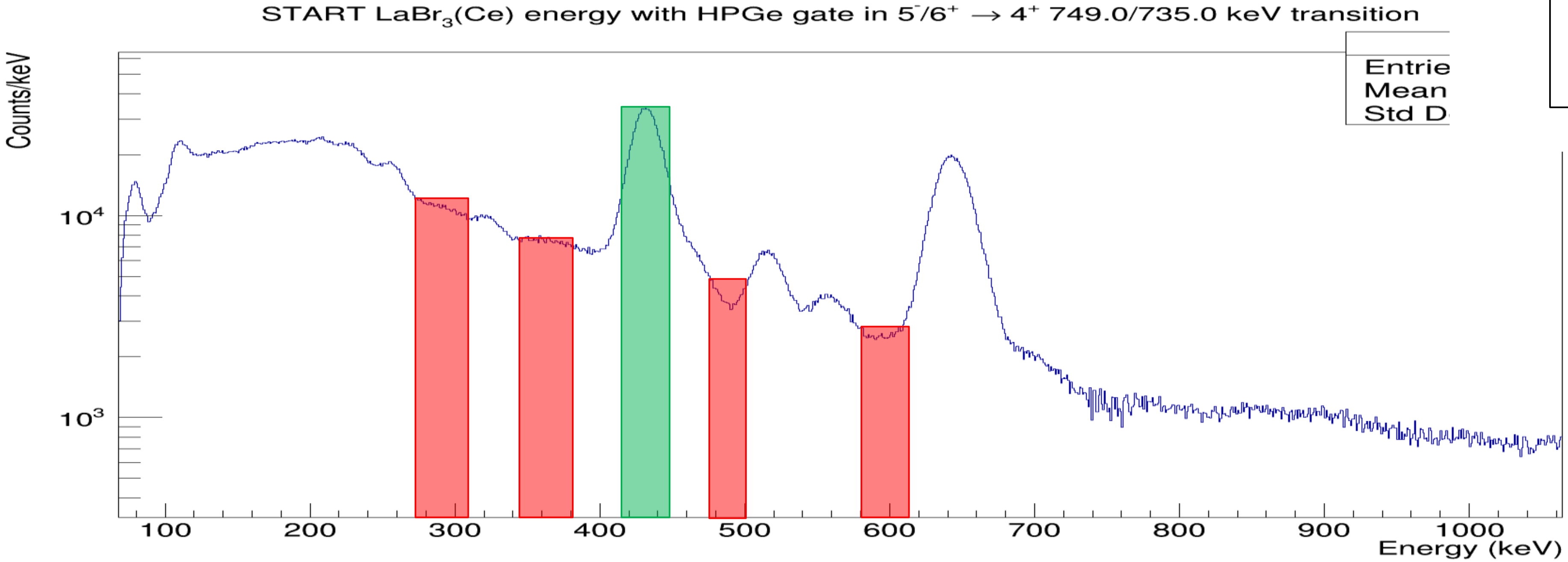
Relative PRD calibration curve

- Timing response calibration
- ^{152}Eu commercial source
- Lifetimes are precisely measured
- Values are corrected by the literature lifetimes
- Uncertainty ~5 ps for the overwhelming Compton background



Compton correction



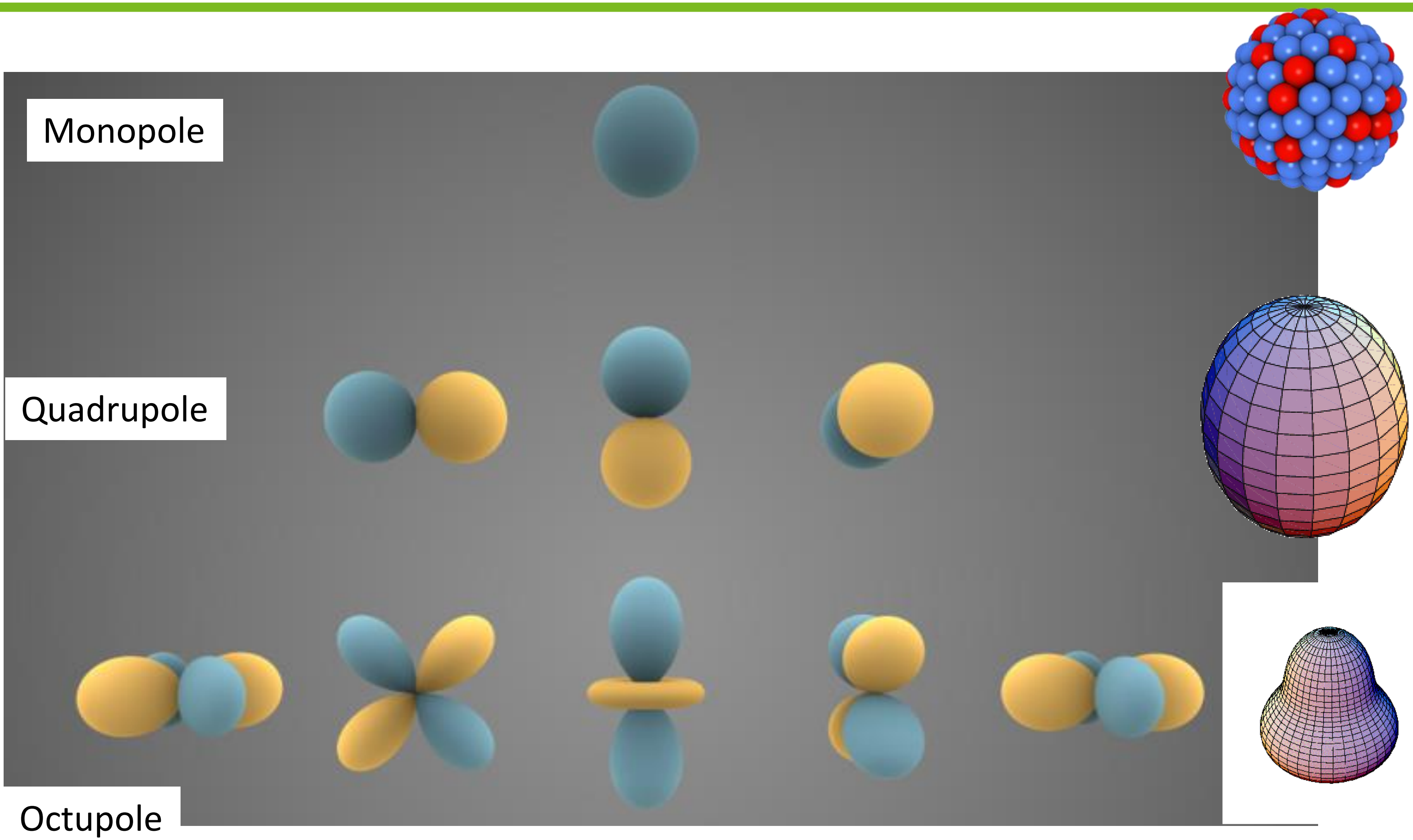


$$A_T C_T = A_P C_P + A_C C_c$$

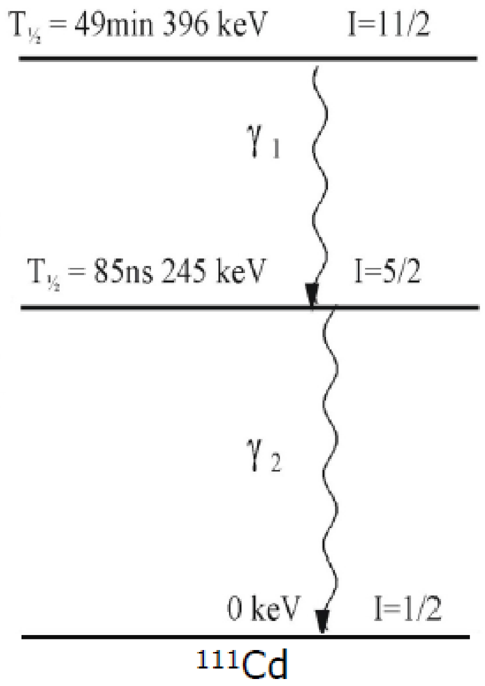
$$C_P = \frac{A_T C_T - A_C C_c}{A_T - A_C}$$

Practical applications:
Perturbed Angular Correlations

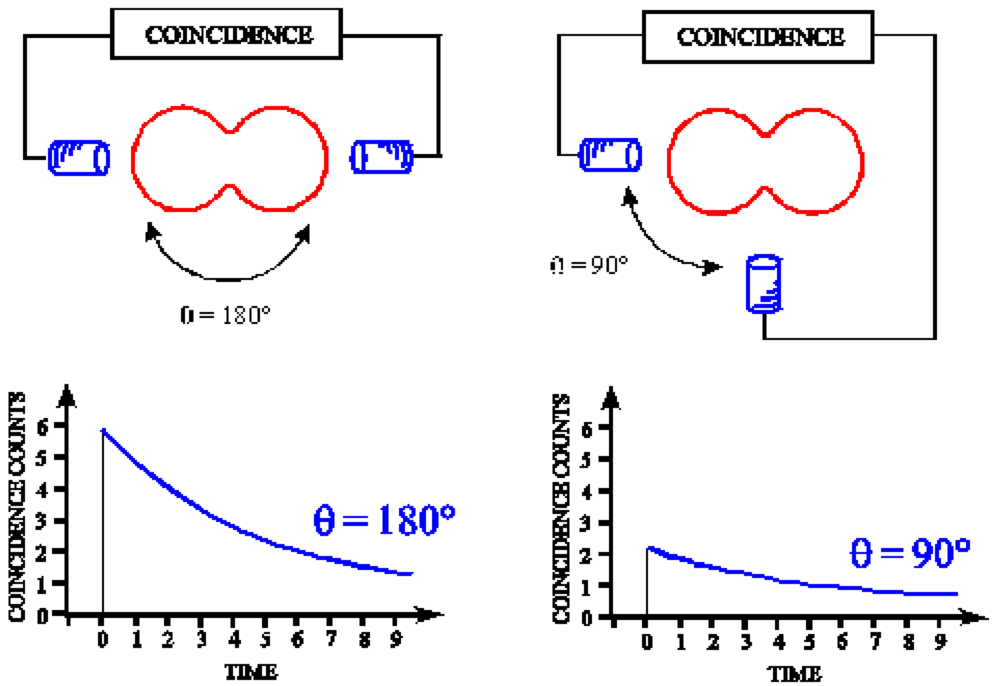
Multipole radiation



PAC Spectroscopy



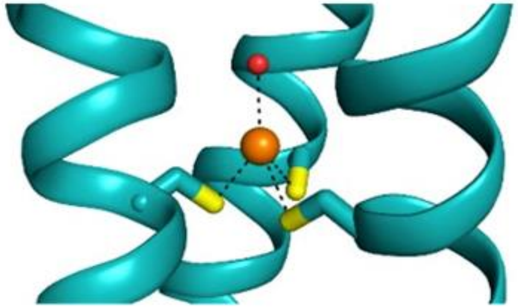
γ - γ angular correlation

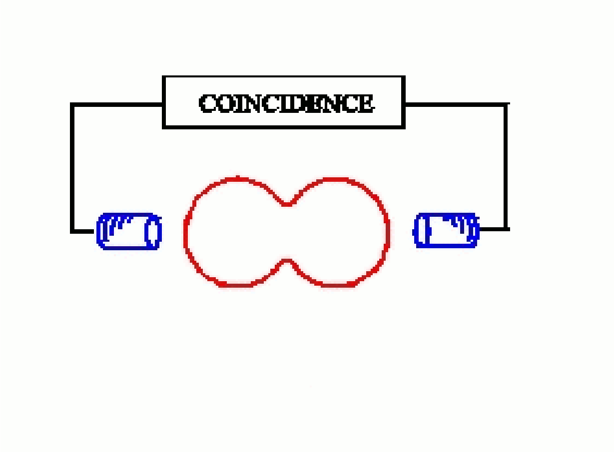


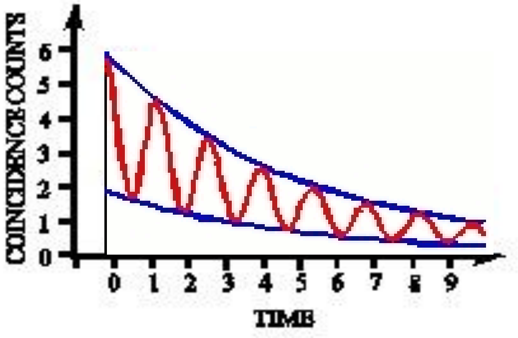
(www.uni-leipzig.de)

Perturbed angular corellation

Now if the electric/magnetic field is created by other atoms in a molecule then the Perturbed γ - γ angular correlation is a very sensitive probe!



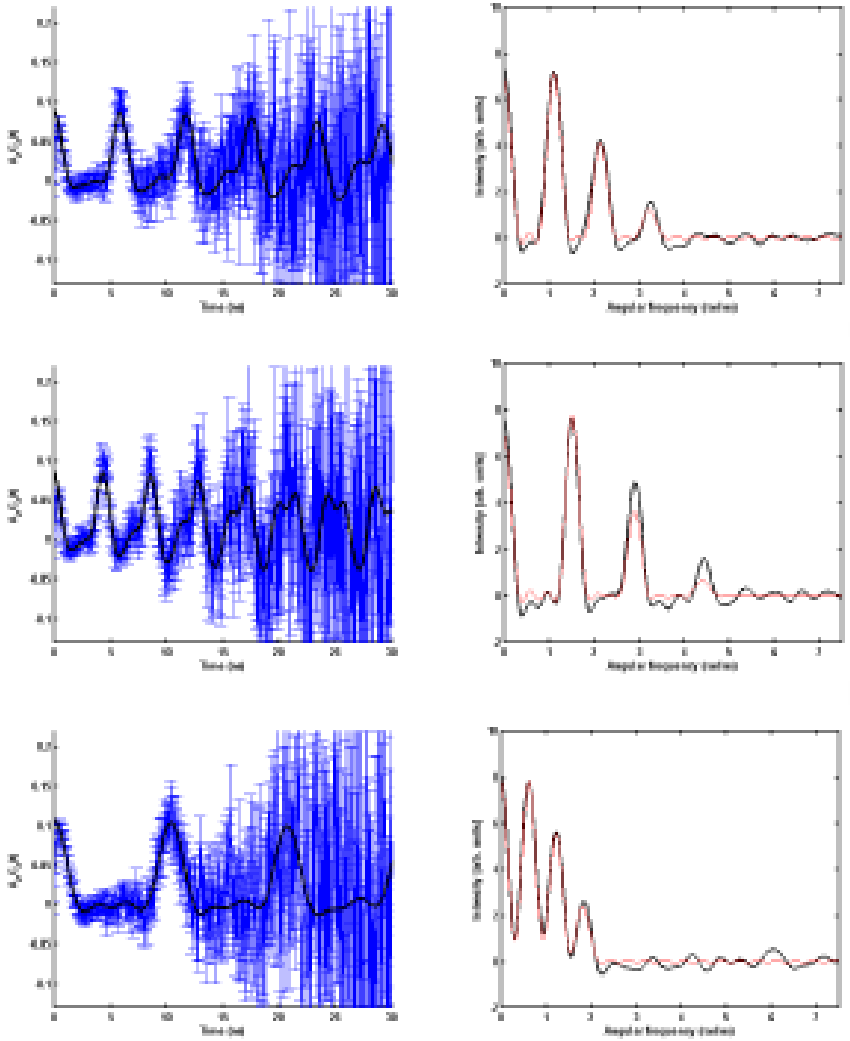




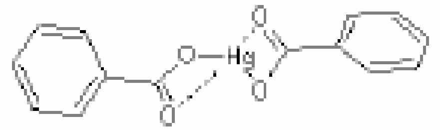
(www.uni-leipzig.de)

- This technique requires detectors with good energy resolution and excellent timing resolution.
- $LaBr_3$ scintillators are the ideal choice.

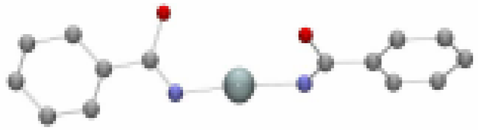
PAC Spectroscopy reveals coordination chemistry



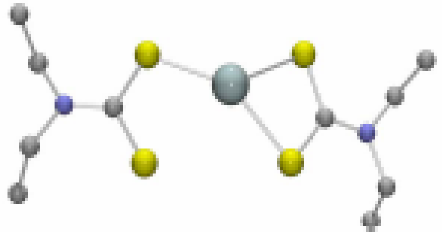
$Hg(benzoato)_2$



$Hg(benzamido)_2$

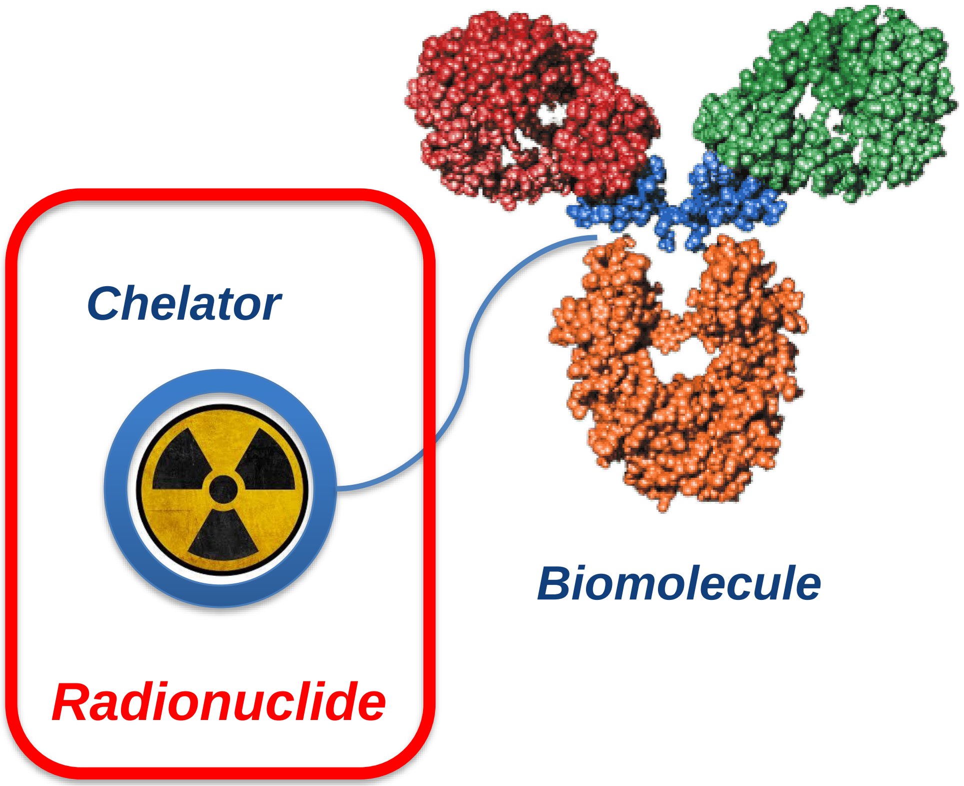


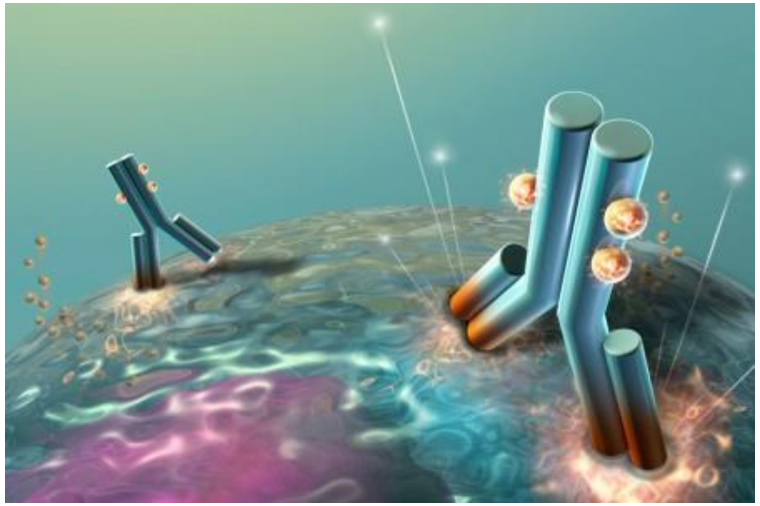
$Hg(Et_2NCS_2)_2$



Radiopharmaceuticals

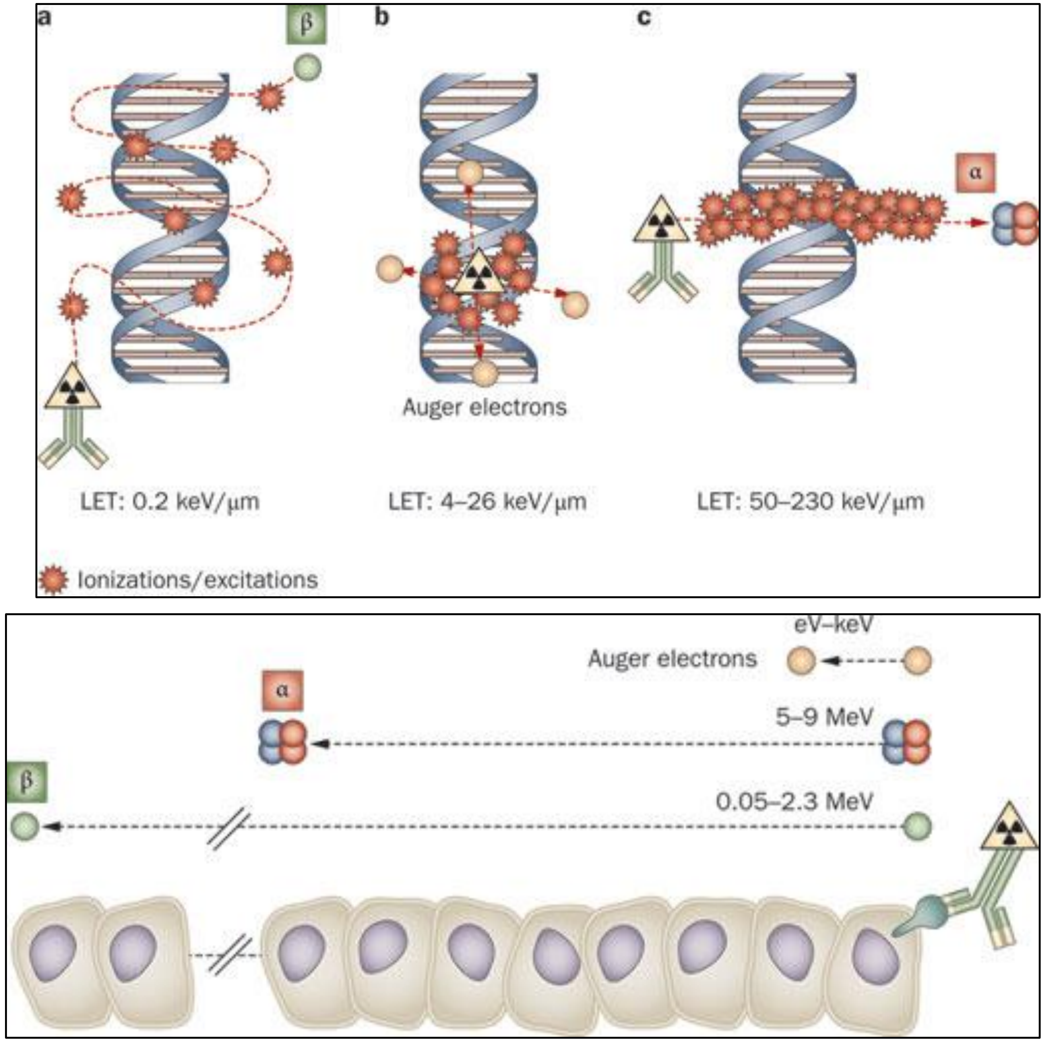
Desire is to place the radionuclide in a carrier molecule which will deliver it directly to the target cancer cells. Can dream of "Designer molecules"





Specific target

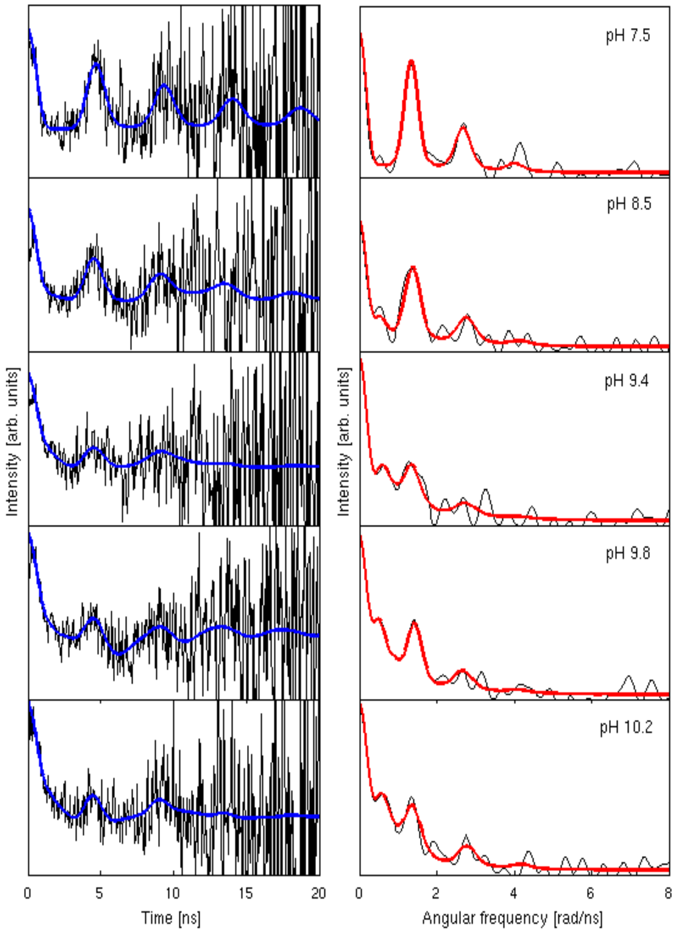
Targeted Radionuclide Therapy (TRT)

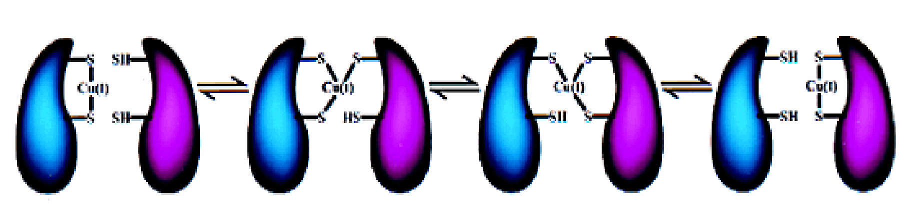


- *Pouget J.-P. et al. (2011) Clinical radioimmunotherapy—the role of radiobiology Nat. Rev. Clin. Oncol.*

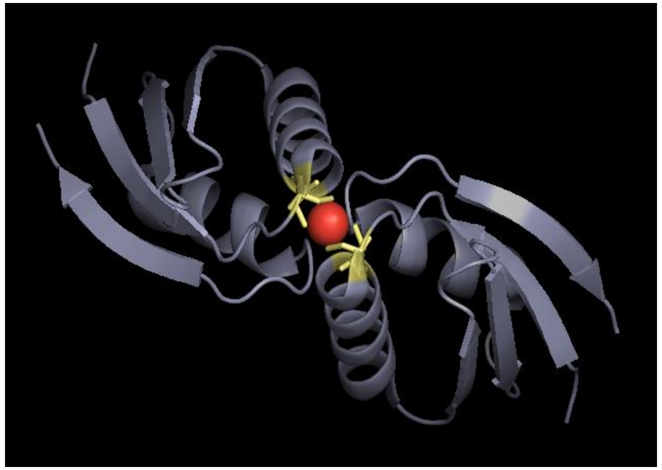
PAC Spectroscopy characterizes protein-protein interactions

The metal ion binding site changes with pH level



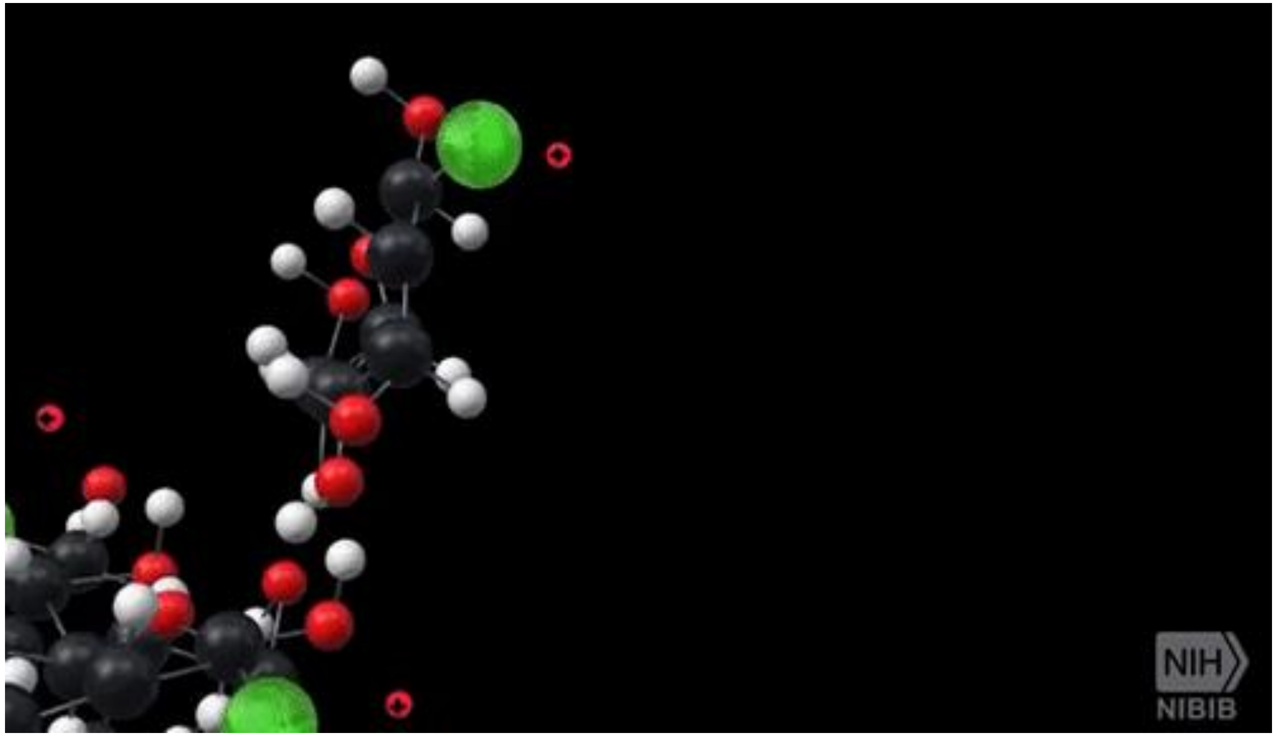


Wernimont et al. Nature Structural Biology 7, 766 - 771 (2000)



Practical applications:
PET-TOF

Positron-electron annihilation



- Positron (e^+) is the anti-particle of the electron
- If they touch, they annihilate $E=mc^2$
- Mass of e^-/e^+ is 511 keV/c^2
- Momentum conservation, **two** 511-keV photons are emitted is **opposite directions**

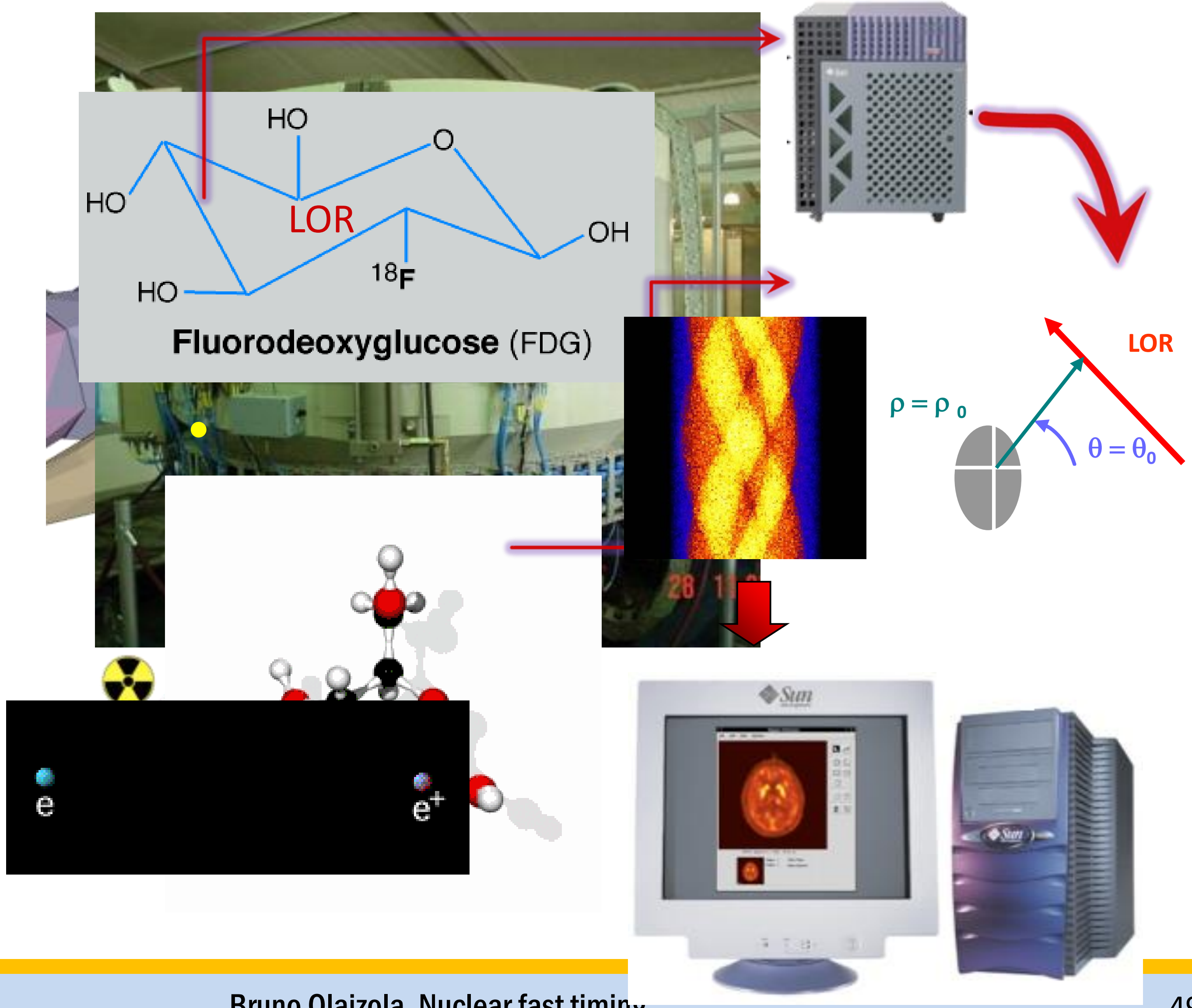
Positron Emission Tomography (PET)

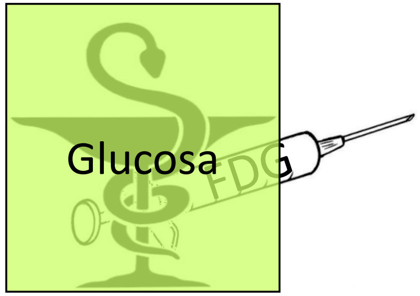
Positron Emission Tomography (PET)

$^{18}O(p,n)^{18}F$

p^+

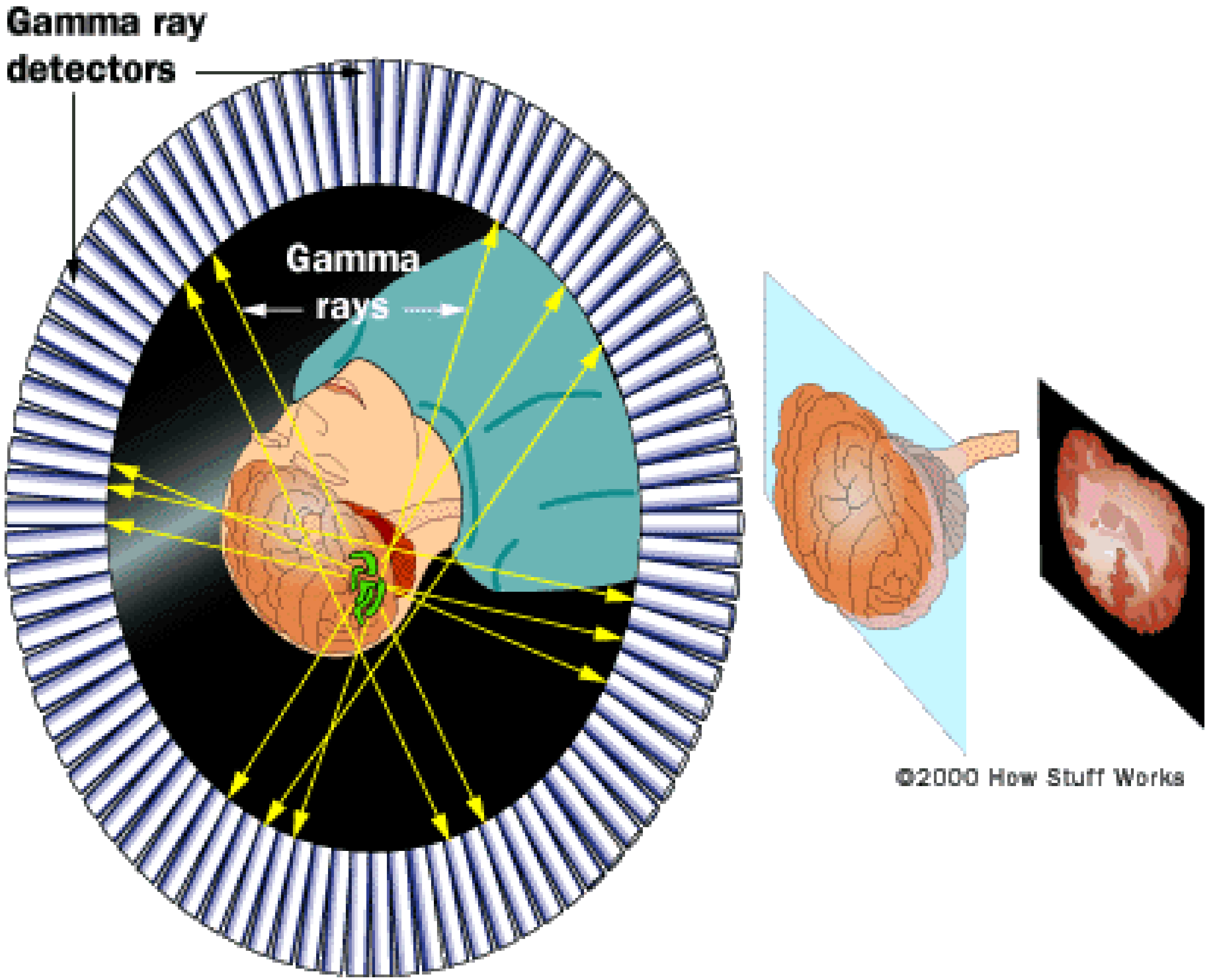
^{18}F n



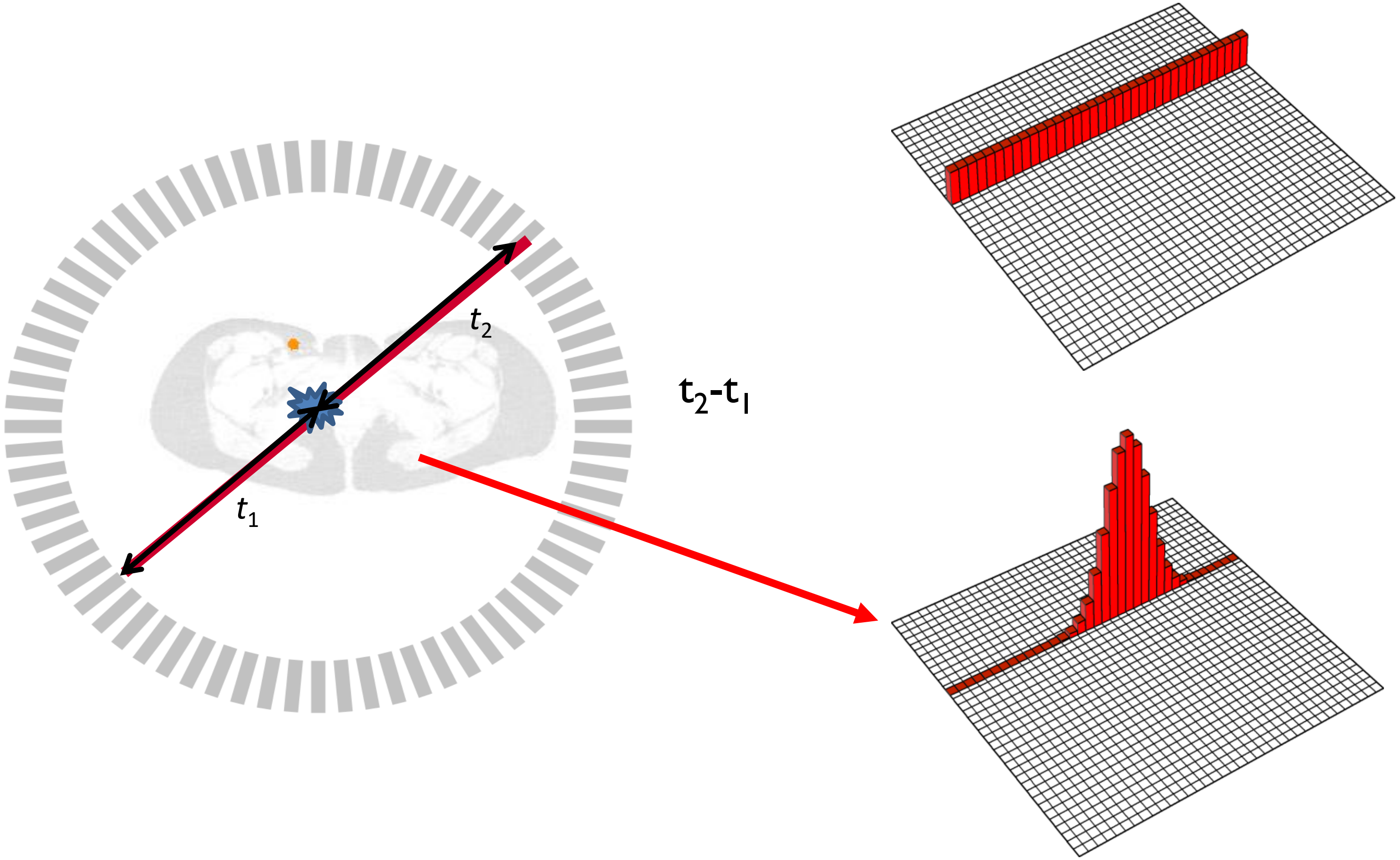


Courtesy of K. Abushab UCM-Spain

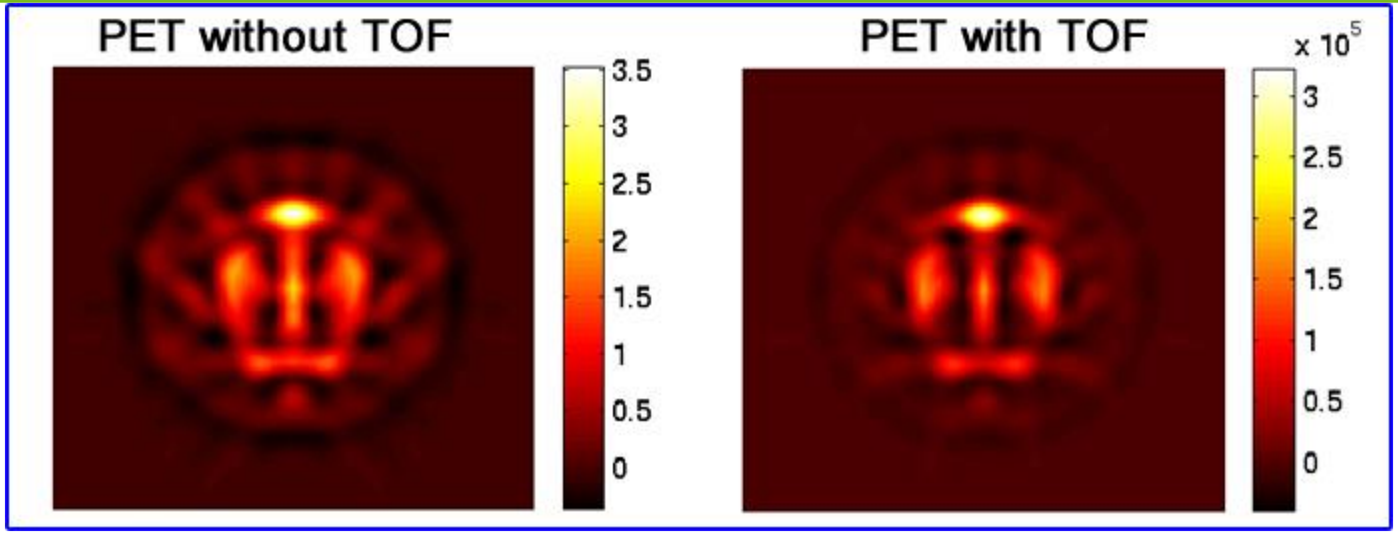
Positron Emission Tomography (PET)

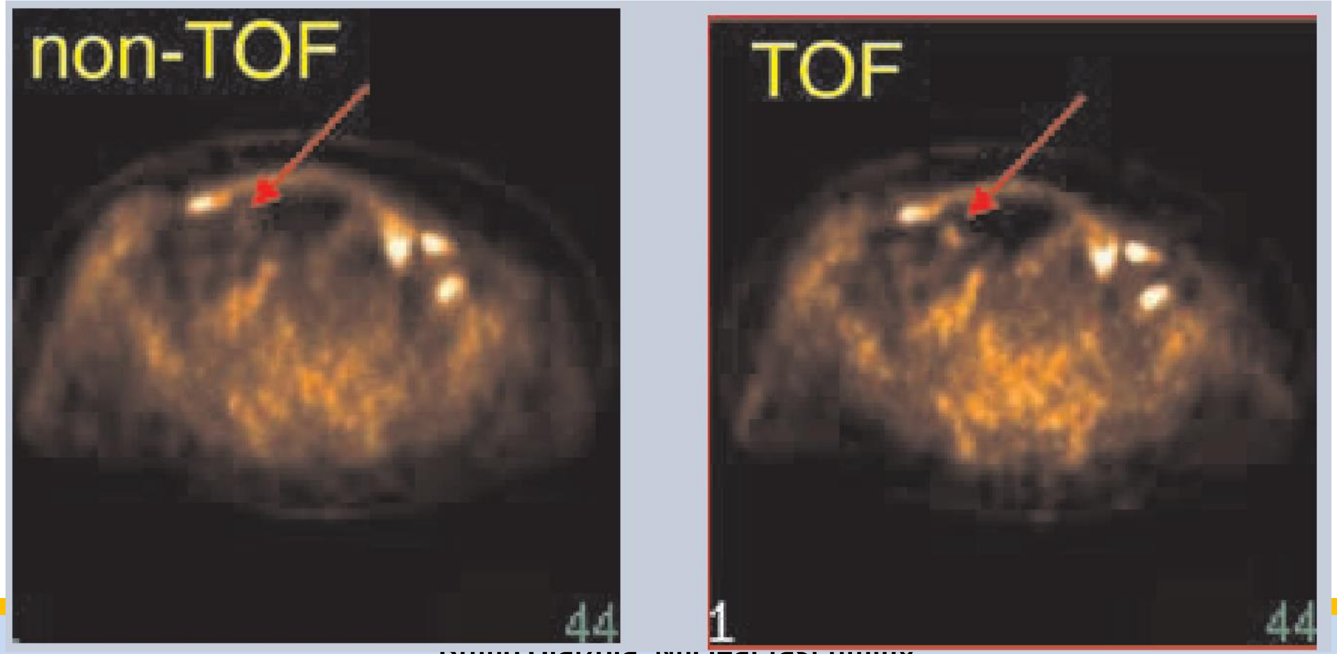


PET – Time of Flight (ToF)



Increased resolution

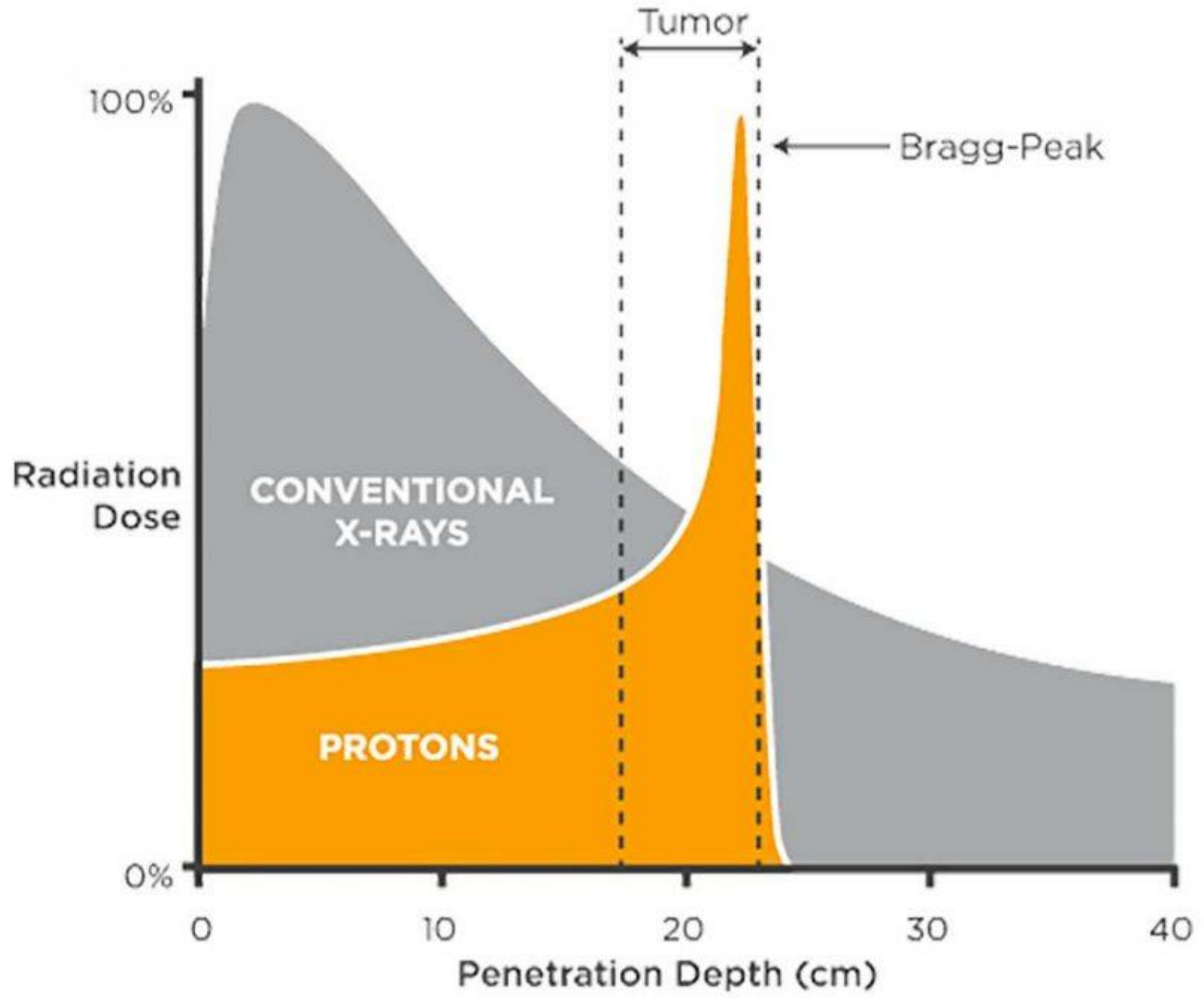




GFN-UCM

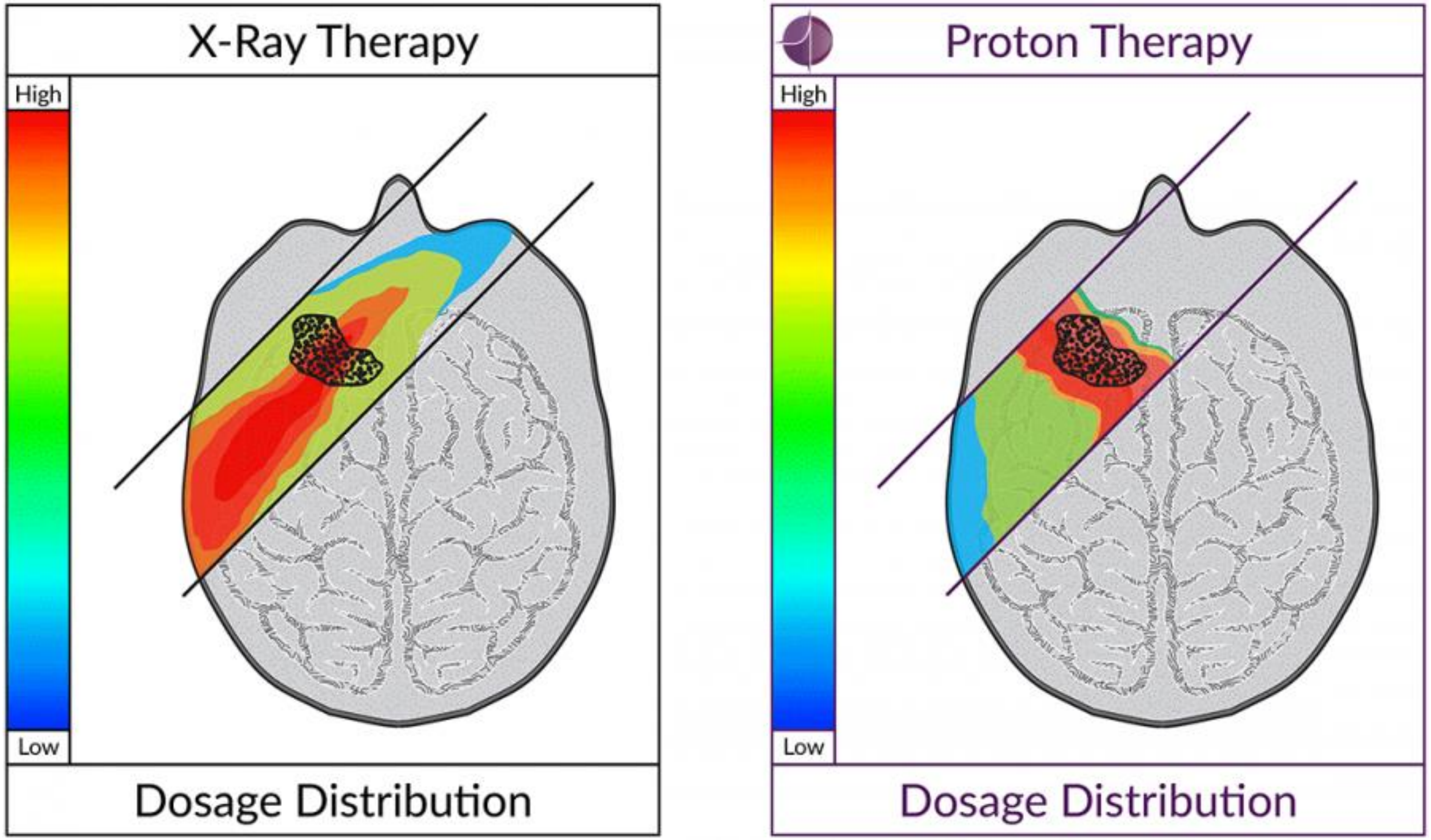
Practical applications:
Proton therapy range verification

Bragg curve



Tumor
100%
Bragg-Peak
Radiation
Dose
CONVENTIONAL
X-RAYS
PROTONS
0%
0
10
20
30
40
Penetration Depth (cm)

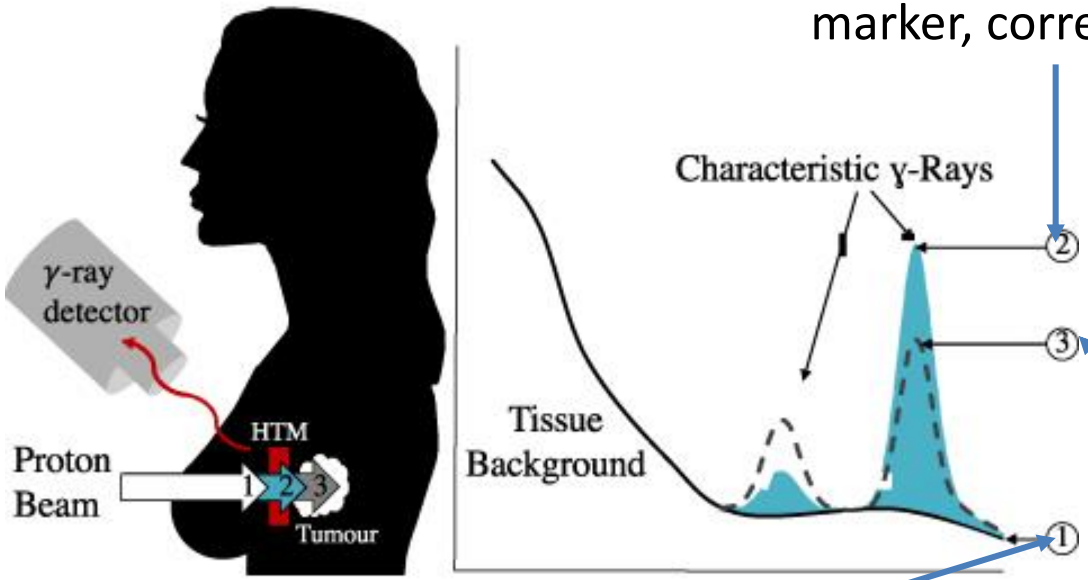
Protontherapy



Range verification

- We insert a metal foil (Mo) in front of the tumor
- Nuclear reaction with p⁺ emits characteristic gamma rays
- Ratio between peaks depends on p⁺ energy

2- Good peak ratio→ Bragg peak next metal marker, correct energy

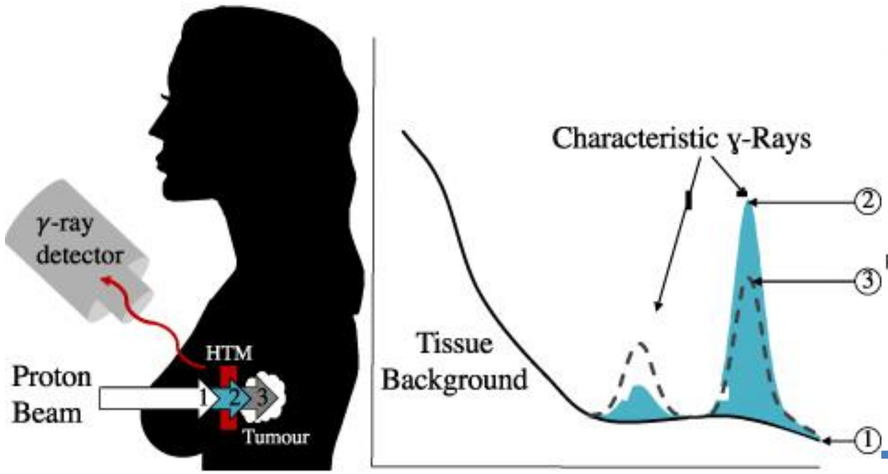


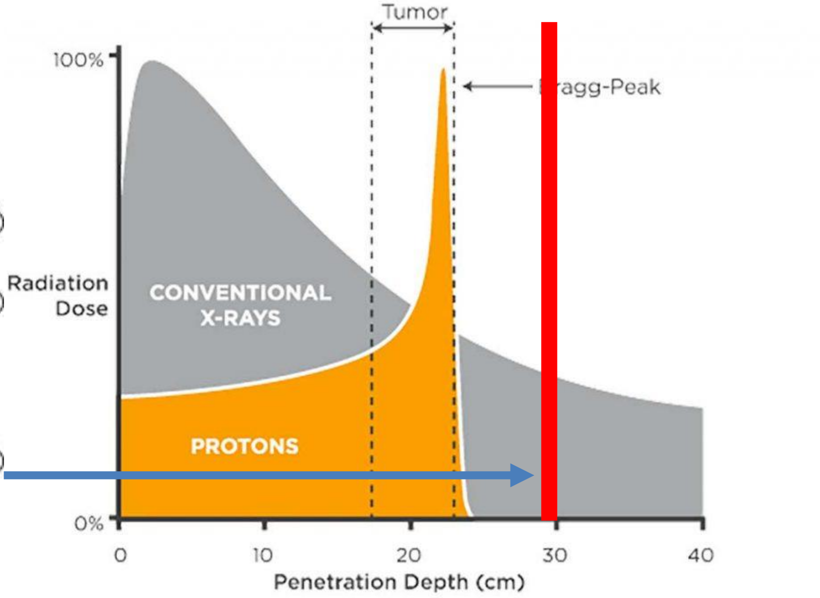
3- Wrong peak ratio→ Bragg peak beyond metal marker, too high energy

1- No peaks → proton energy too low

C Burbadge *et al* 2021 *Phys. Med. Biol.* **66** 025005

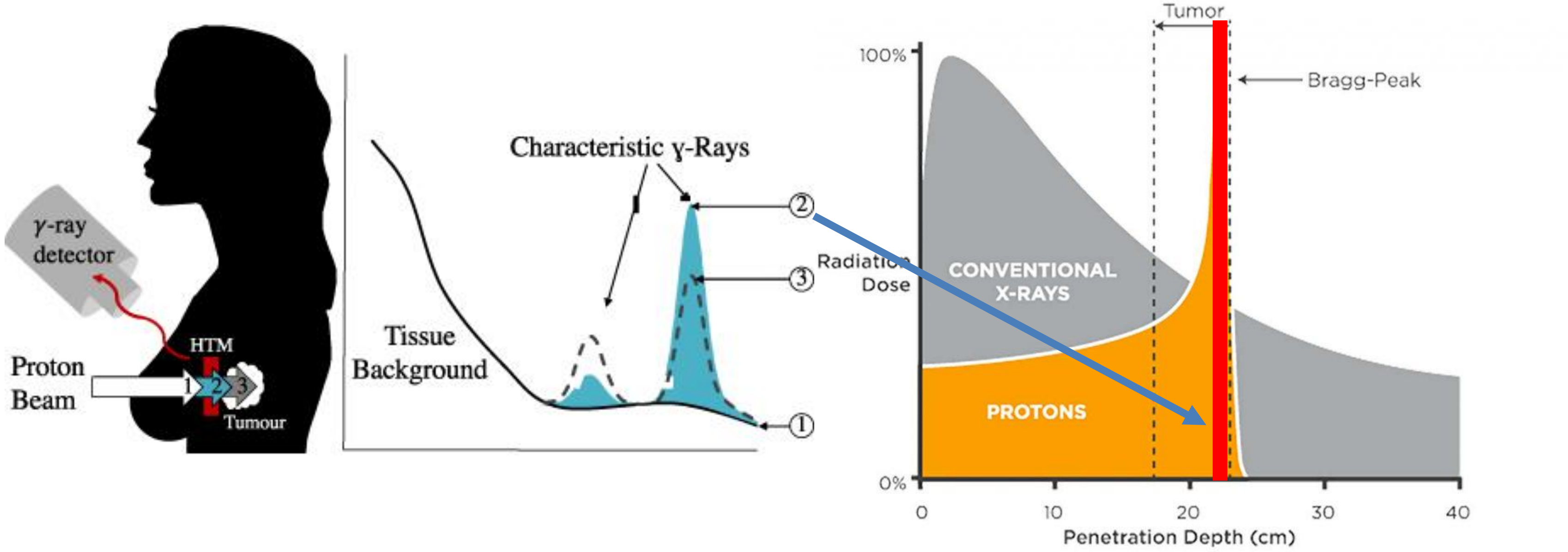
Range verification





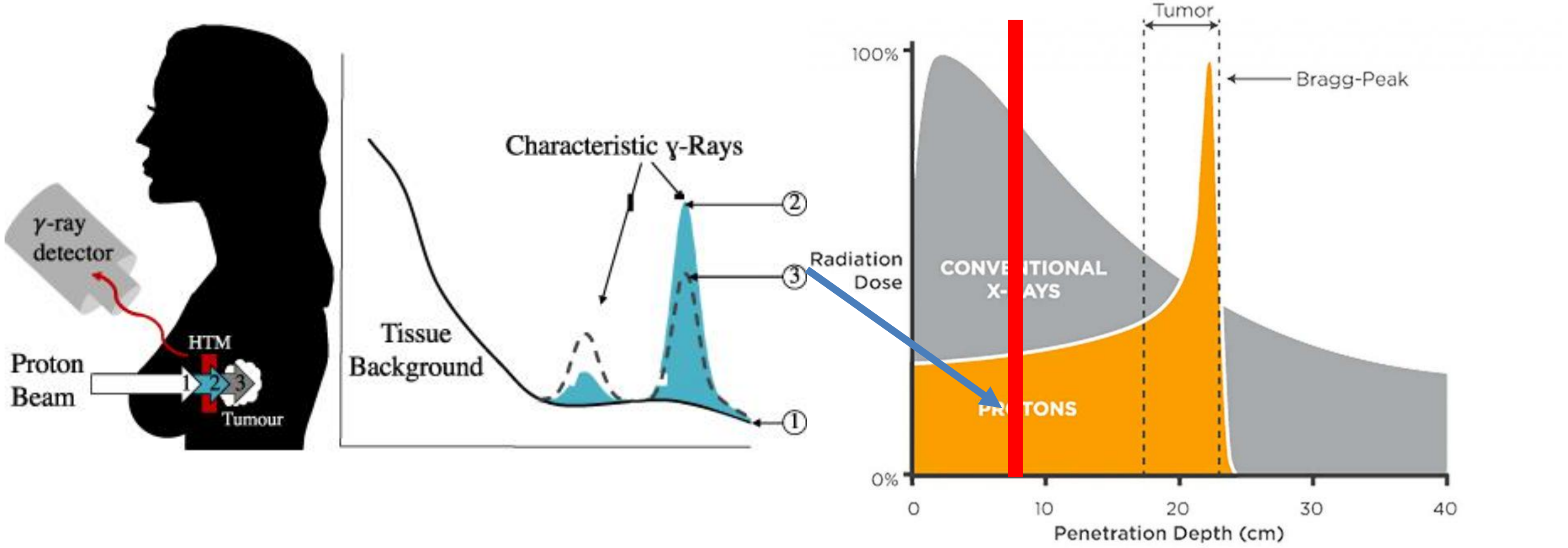
- Protons stop before metal foil
- No nuclear reaction
- Only tissue background

Range verification



- Bragg peak at the metal foil
- Maximum proton energy induces nuclear reaction
- Characteristic gamma ray appears

Range verification

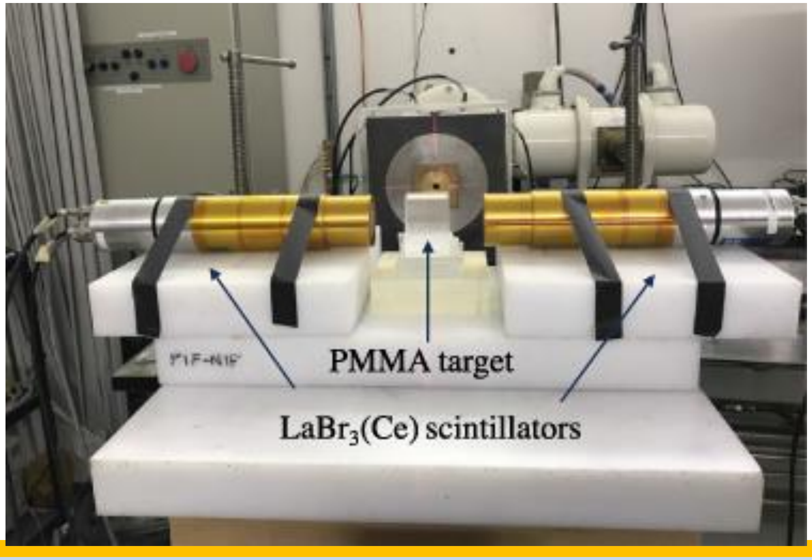


- Bragg peak beyond the metal foil
- Only partial proton energy induces nuclear reactions
- Different gamma peak ratio

Range verification

- Interaction of p^+ with tissue will create large gamma-ray fields
- Rates on the detectors well over 50 kHz
- Requires extremely fast detectors, such as $LaBr_3$
- The technique allows for online range verification
- Sub-mm precision achieved

C Burbadge *et al* 2021 *Phys. Med. Biol.* **66** 025005



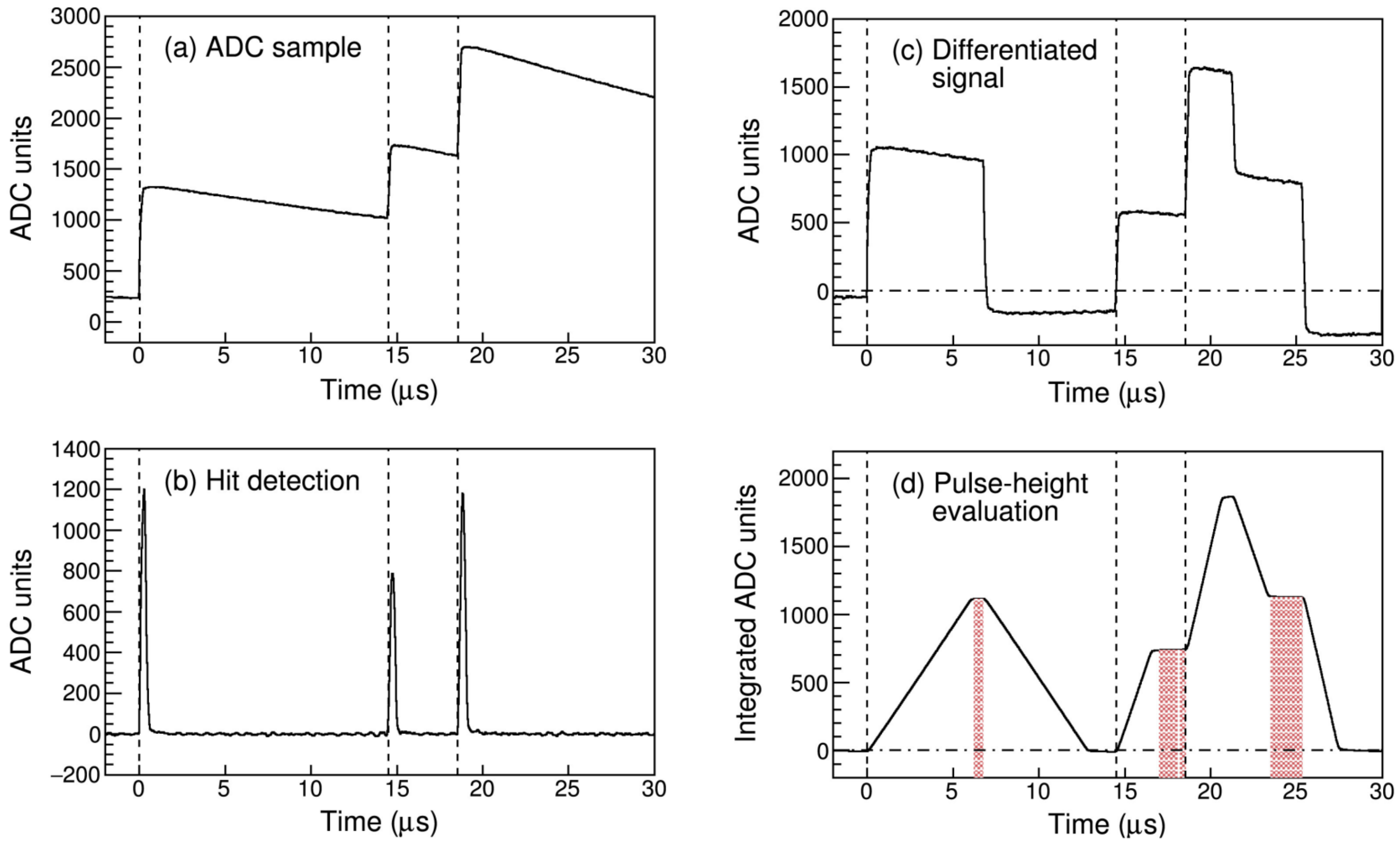
Summary

- Lifetime measurement is one of the most powerful probes we have to study nuclear structure
- Scintillators allow to measure timing down to ~10 ps (10×10^{-12} s)
- The fast-timing method has practical applications:
  - Protein structure and interaction
  - Medical imaging (PET-ToF)
  - Proton therapy range verification

Any questions?

You can always contact me at
bruno.olaizola@csic.es

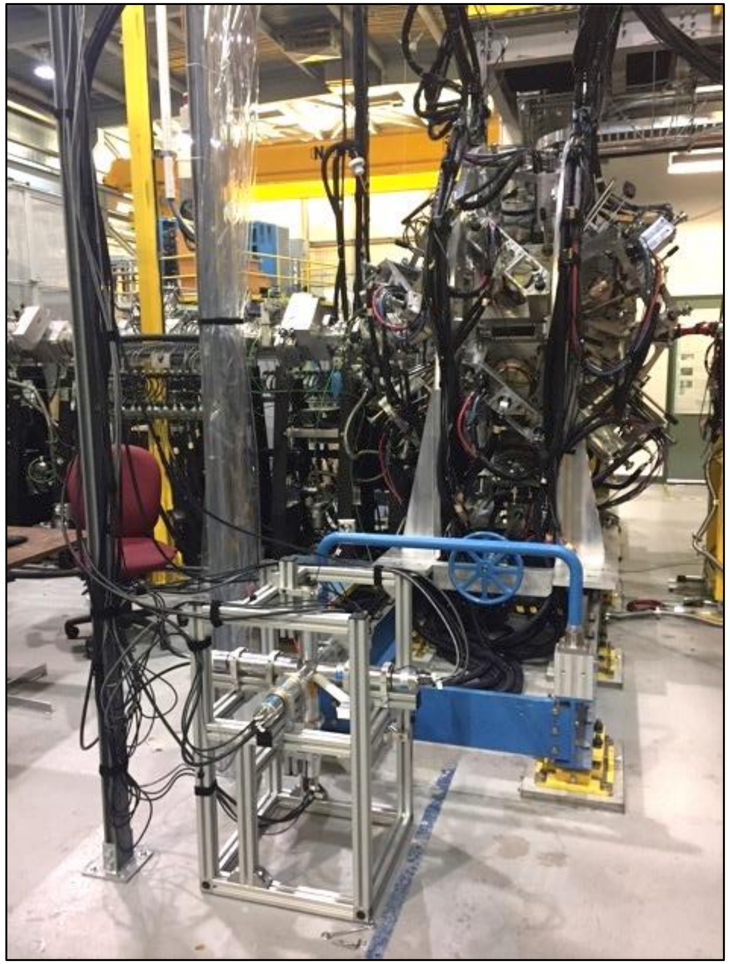
Pile up



(a) ADC sample
(b) Hit detection
(c) Differentiated signal
(d) Pulse-height evaluation
ADC units
Integrated ADC units
Time (μs)

PAELLA at TRIUMF

Perturbed **A**ngular corr**EL**ations **L**abr **A**rray



$$B\left({}^{E}_{M}\lambda, L_i \rightarrow L_f\right) = \sum_{\mu M_f} \left|\left\langle I_f M_f \middle| M\left({}^{E}_{M}\lambda, \mu\right) \middle| I_i M_i \right\rangle\right|^2$$