

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



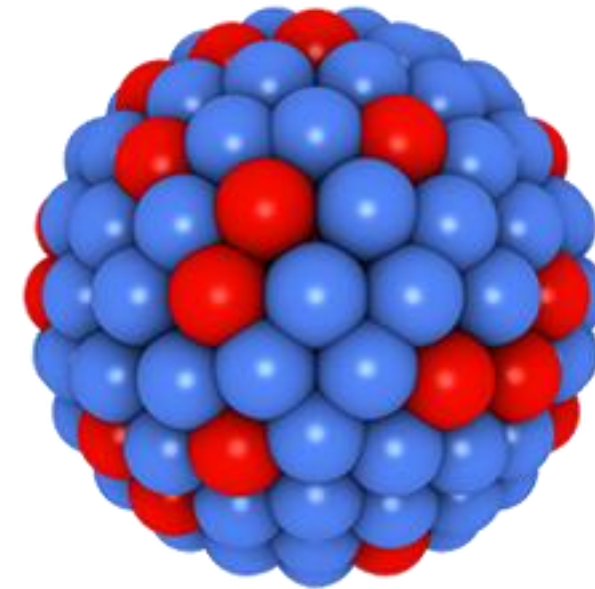
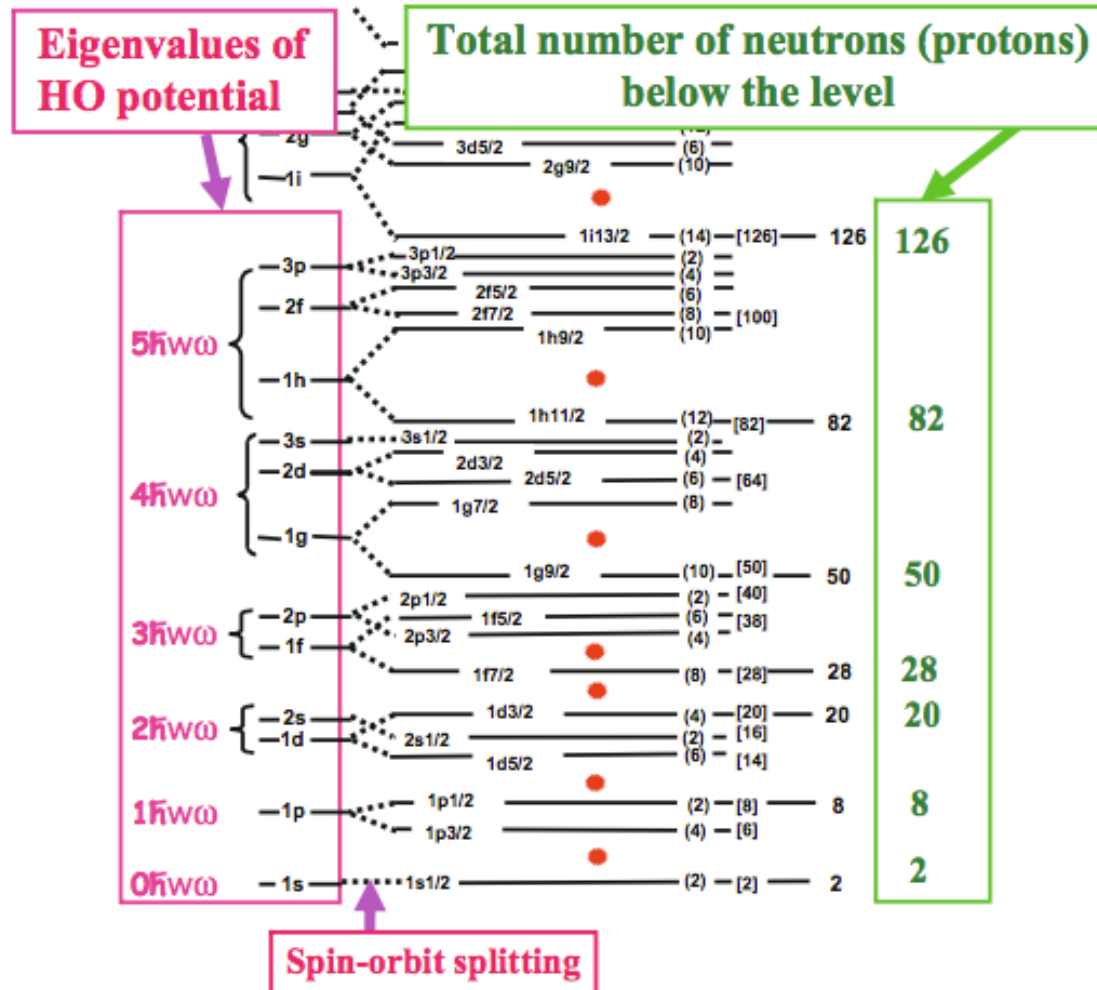
Máster Física Nuclear  
2024-2025

Bruno Olaizola  
CSIC-IEM

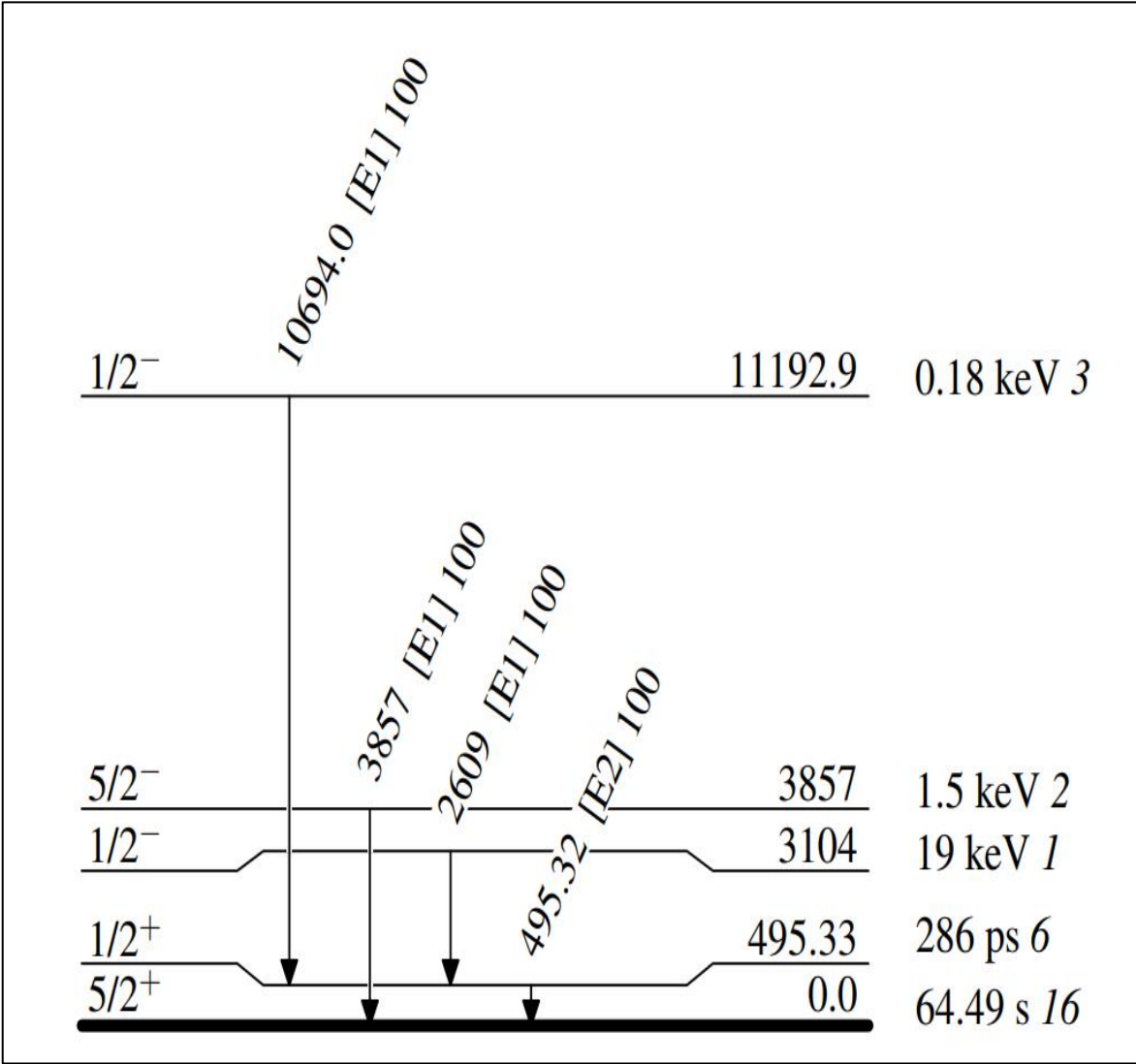
- Nuclear lifetimes and transition strengths
  - Collective motion
  - Weisskopf estimate
- Electronic fast timing
  - Experimental setup
  - Analysis method
- Practical applications:
  - Time-of-Flight Positron Emission Tomography (ToF-PET)
  - Perturbed Angular Correlations (PACs)
  - Proton therapy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

# Nuclear lifetimes and transition strengths

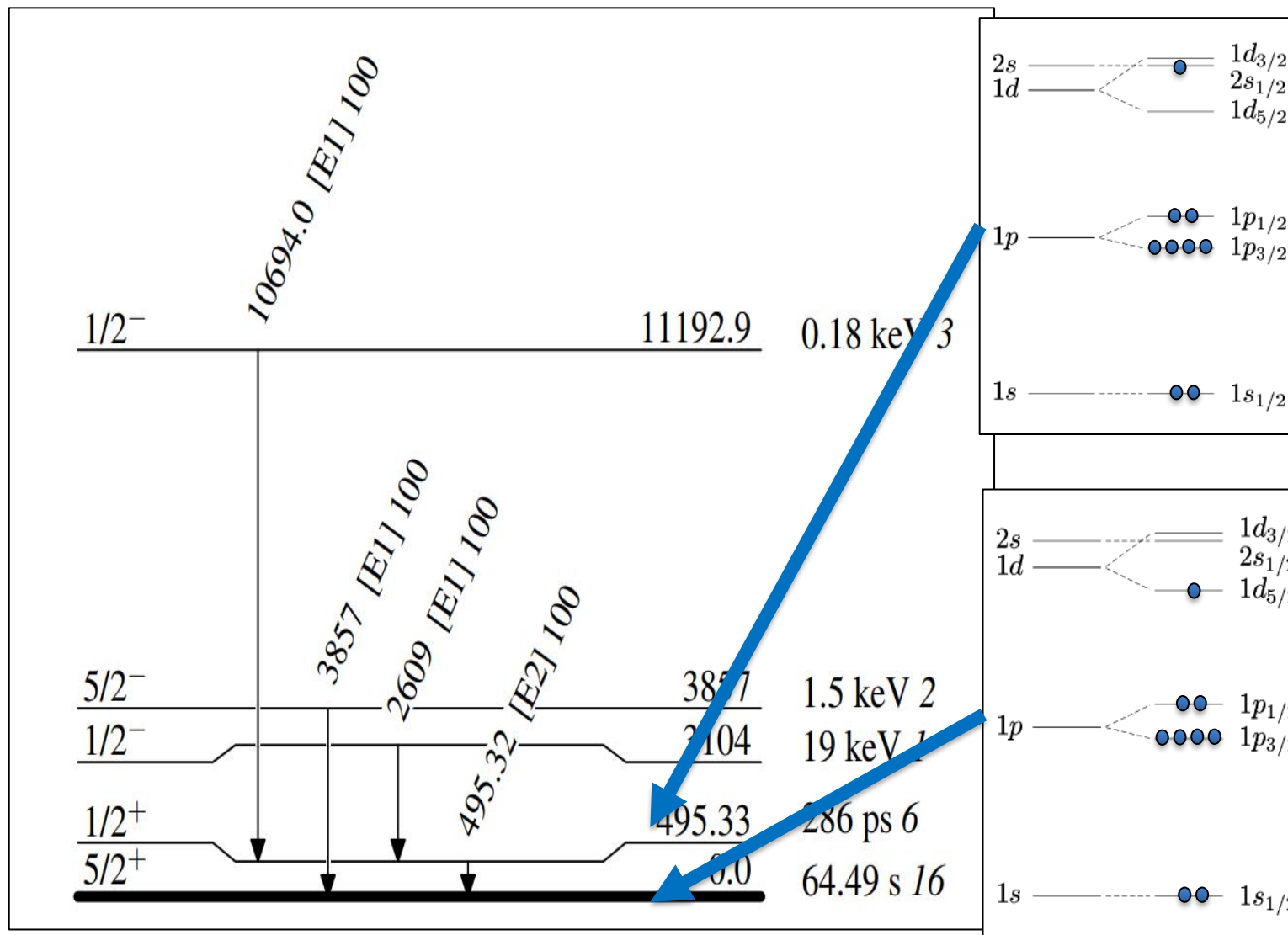
# Nuclear shell model



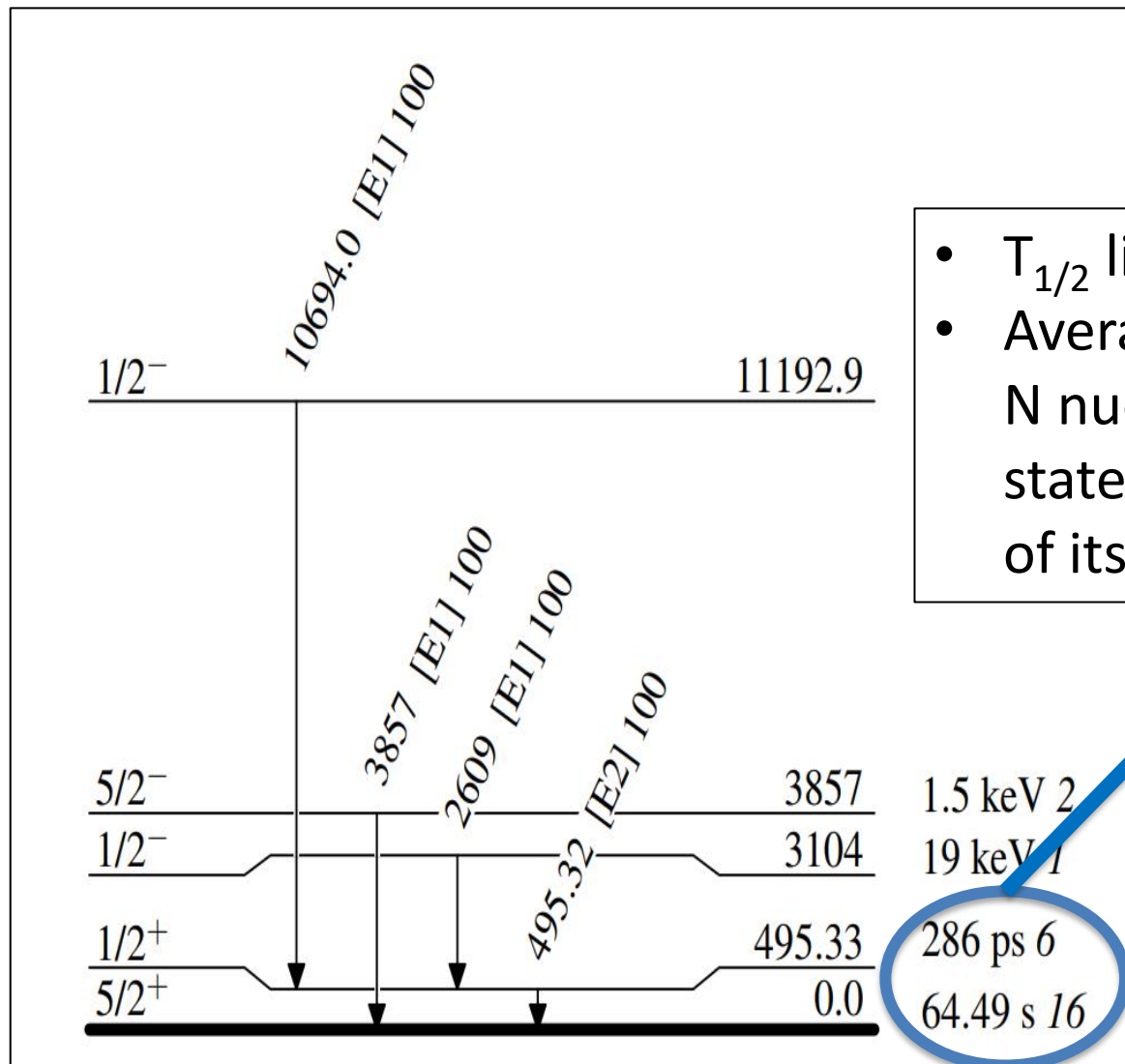
# Nuclear level scheme



# Nuclear level scheme



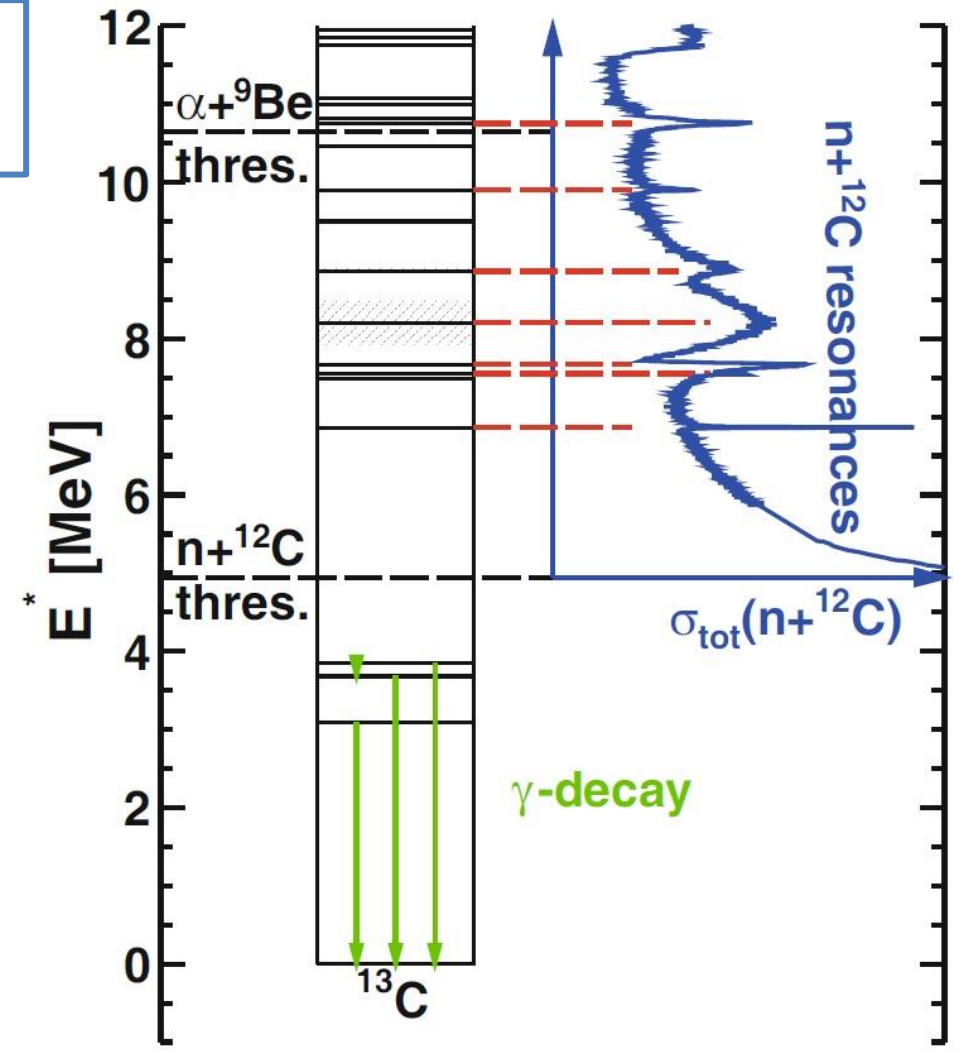
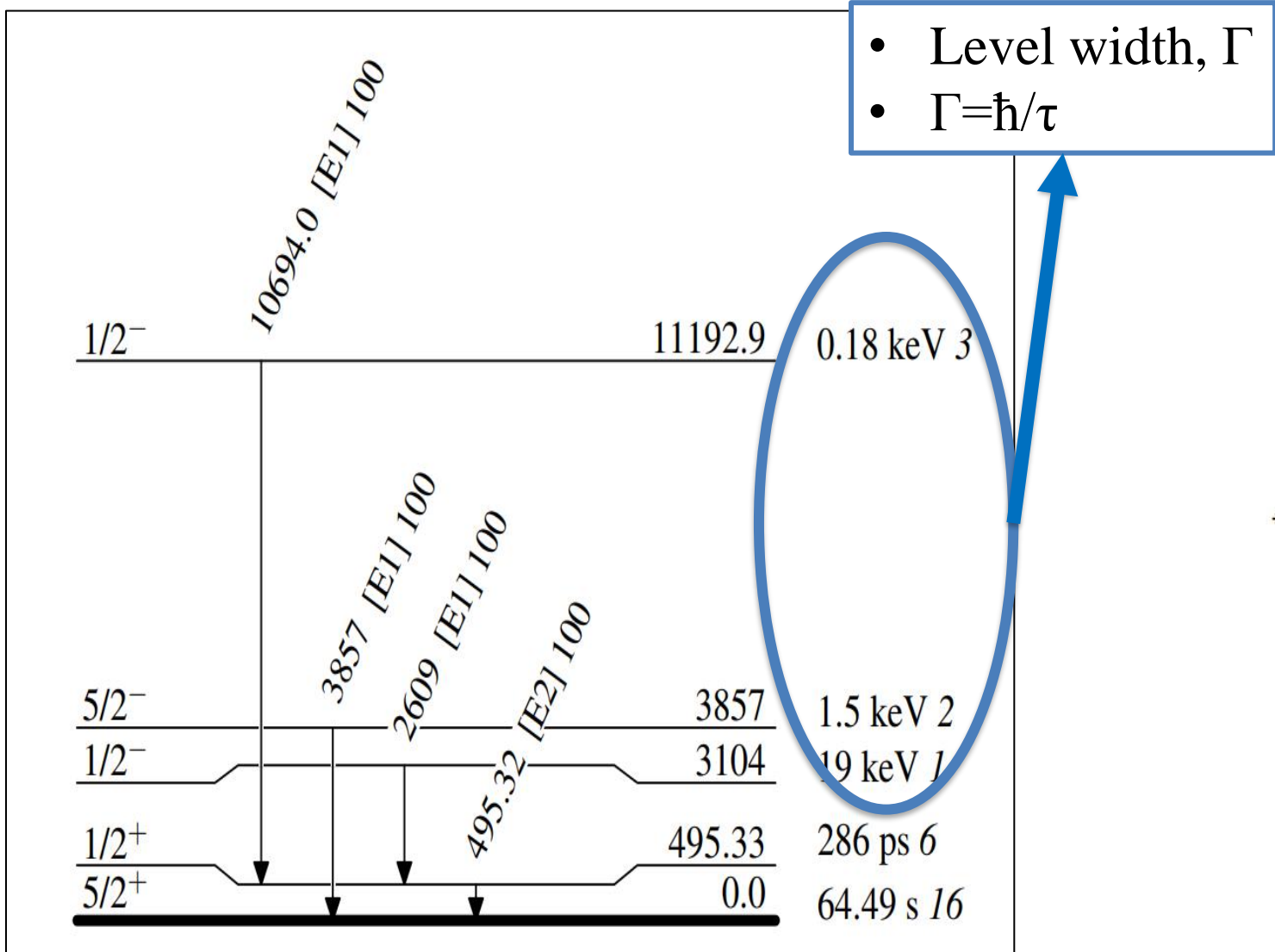
# Nuclear level scheme



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

286 ps 6  
64.49 s 16

# Nuclear level scheme



R.J. Charity, Eur. Phys. J. Plus (2016) 131: 63



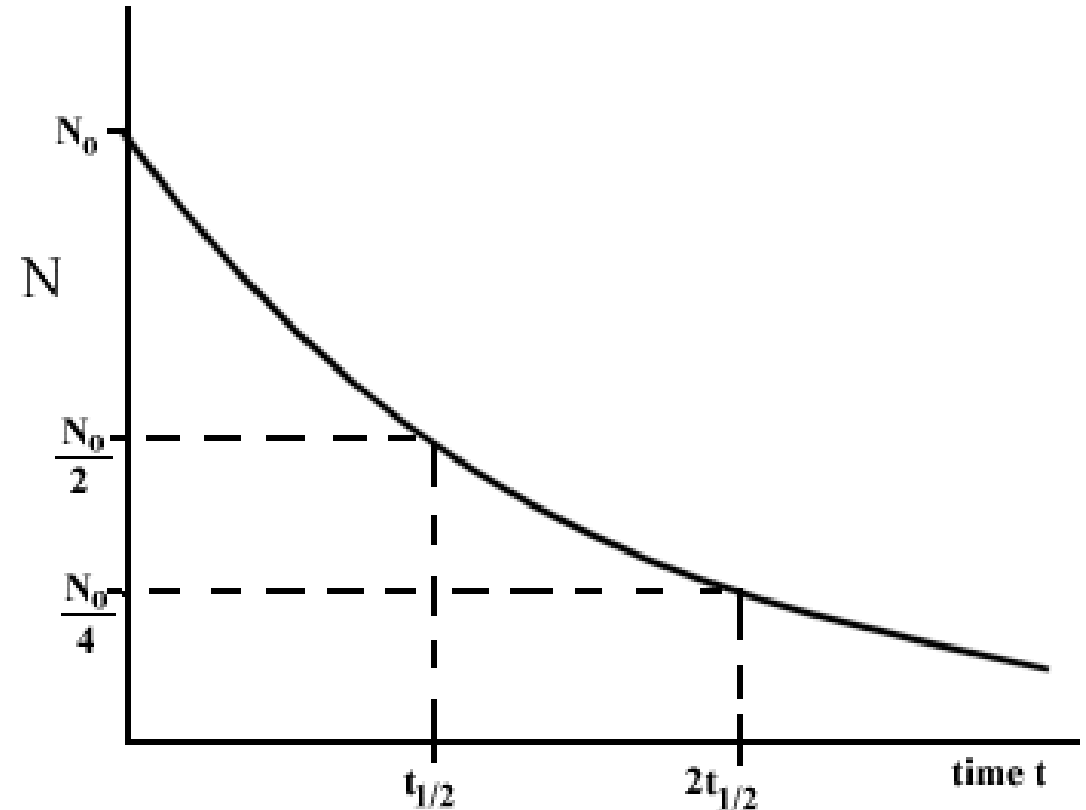
# Nuclear half-life

- The activity of the sample changes as:  $A(t) = A_0 e^{-\lambda t}$
- From  $\lambda =$  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

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

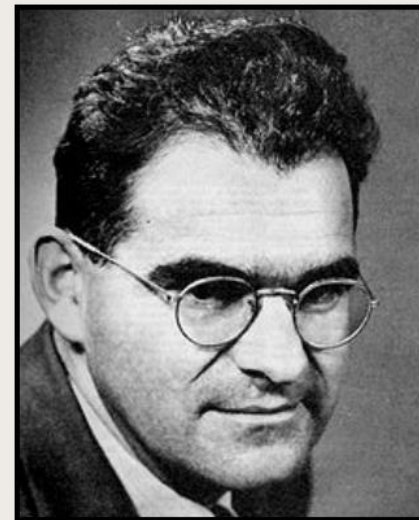
Which relates the transition probability (and therefore the lifetime) to the reduced transition matrix element,  $B(\lambda L)$

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

# Weisskopf estimates

So what?

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



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

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

# Weisskopf estimates

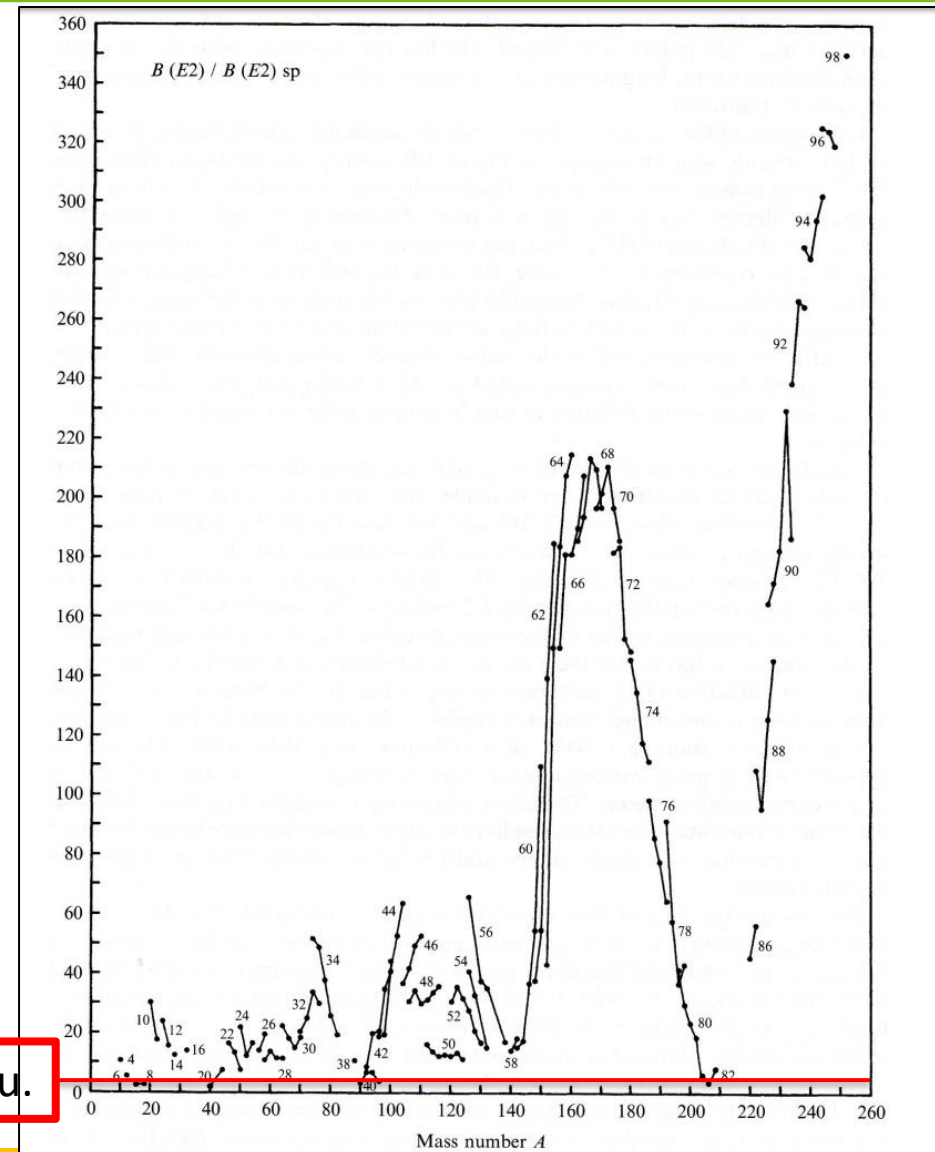
**Table 1. Formulae for single-particle transition half-lives, corrected for internal conversion.**

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

- We measure B(XL) and compare to Weisskopf estimates
- Gives us an idea if the transition can be described as single particle
- Depend on  $T_{1/2}$ , branching ratio, E and A (actually, radius  $R \propto A^{1/3}$ )
- All observables that can be measured
- In most cases,  $T_{1/2}$  is the hardest
- Strong dependence with E
  - The lower the energy  $\rightarrow$  the much longer the lifetime. Example for E2:
    - $E_1 = 100 \text{ keV} \rightarrow \tau_1 = 500 \text{ ps}$
    - $E_2 = 200 \text{ keV} \rightarrow \tau_2 = 15 \text{ ps}$

# Single particles are not enough

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



$B(E2) = 1$  W.u.

# Recommended upper limits

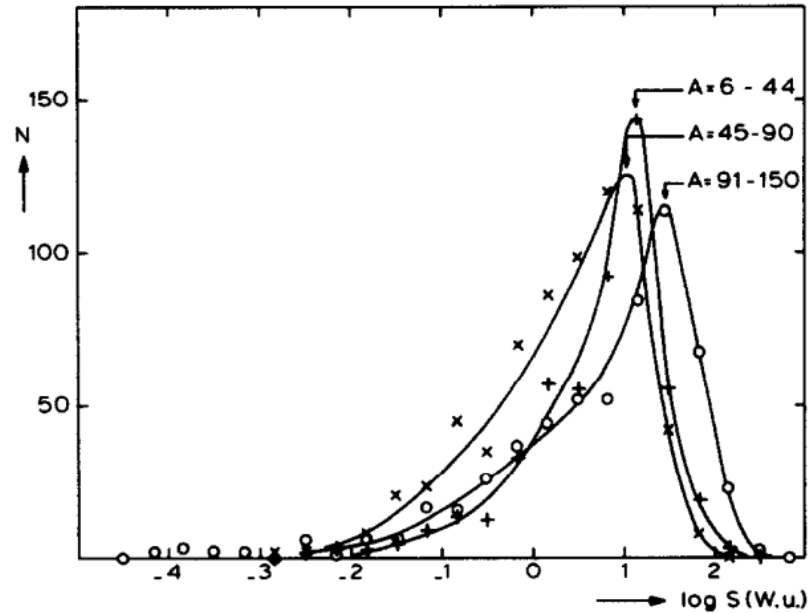


Fig. 5. Comparison of  $E2$  strength distributions for different  $A$ -regions. Data for  $A = 6-44$  and  $A = 45-90$  are from Refs. 1 and 2, respectively.

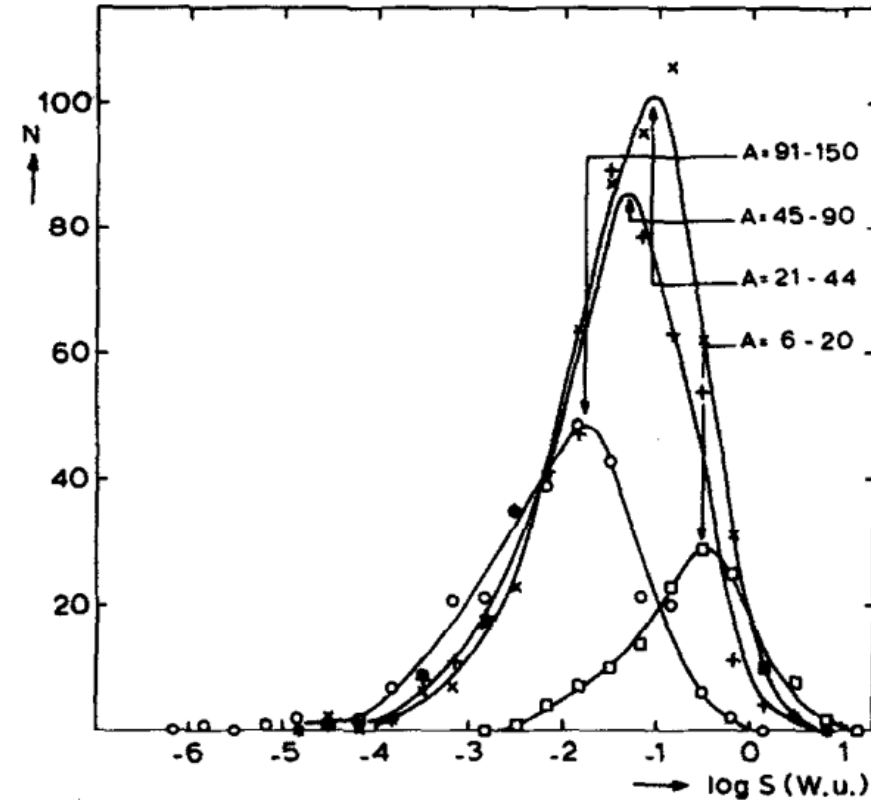


Fig. 6. Comparison of  $M1$  strength distributions for different  $A$ -regions. Data for  $A = 6-44$  and  $A = 45-90$  are from Refs. 1 and 2, respectively.

# Recommended upper limits

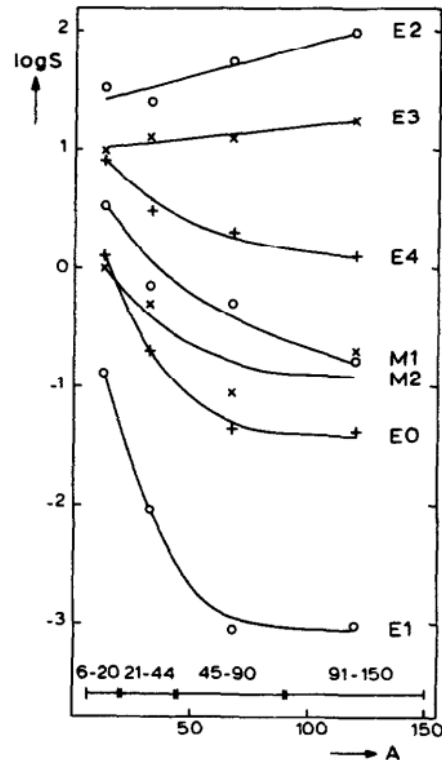


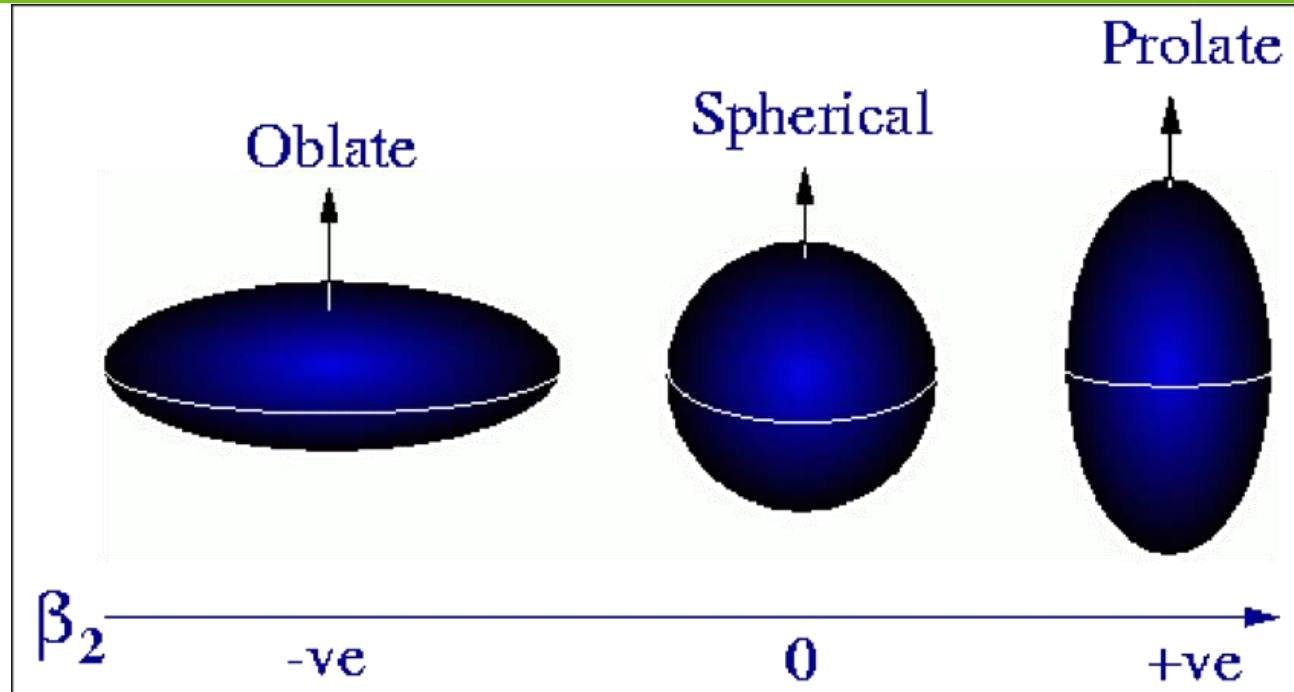
Fig. 7. Average strengths of strongest transitions as a function of  $A$ ; for  $E1$ ,  $E2$ , and  $M1$  averages are taken of the strongest 10%, for  $E3$  and  $M2$  of the strongest 50%, and for  $E0$  and  $E4$  of all transitions. Strengths are expressed in W.u., except for  $E0$  (Wi.u.). Data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

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

Multipolarity	$5 \leq A \leq 44^a$	$45 \leq A \leq 90$	$91 \leq A \leq 150$	$A \geq 151$
$E1$ (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  $B(\text{XL})$
- 40+ years old, could be outdated
- NNDC is currently working on a new compilation

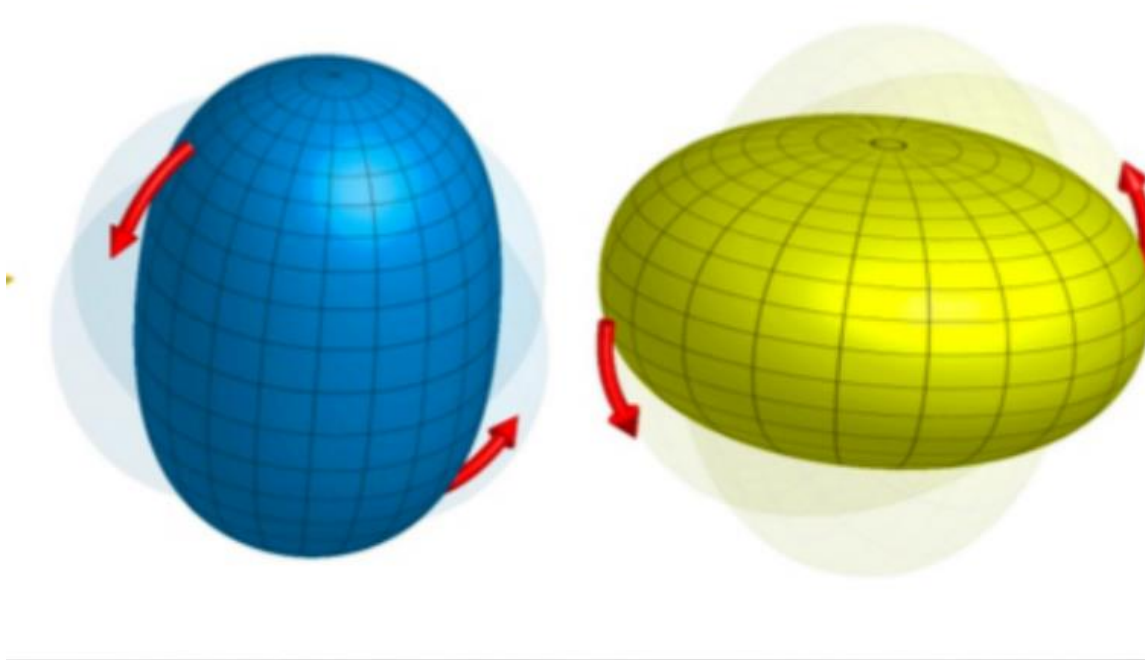
# Deformation



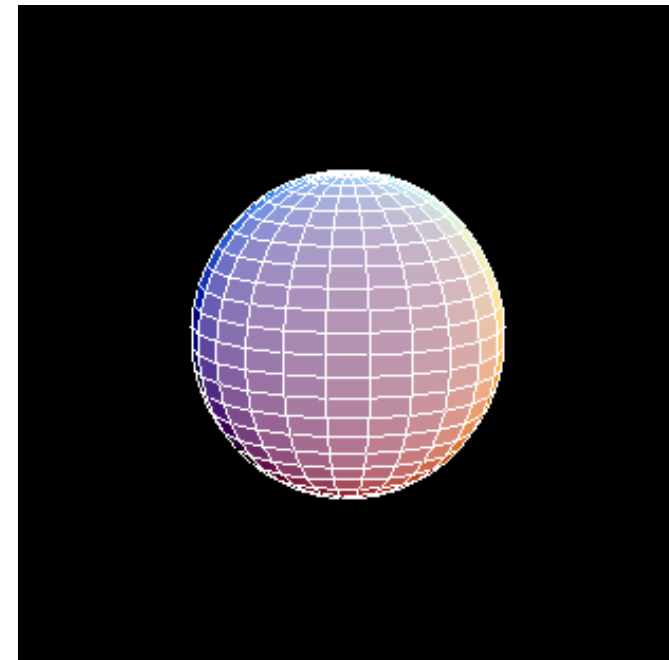


# Collective motion

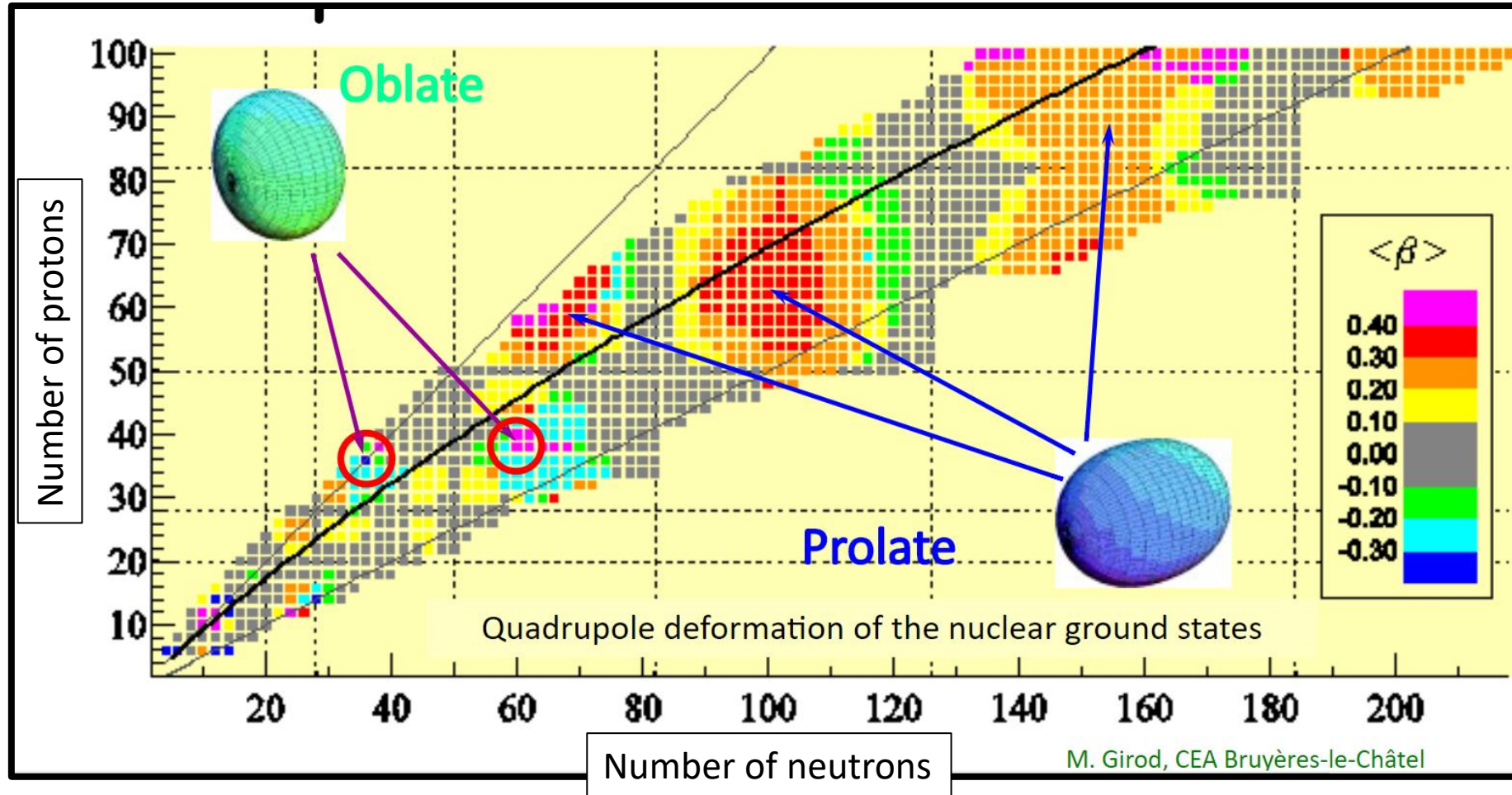
## Rotation



## 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 ( $\sim 10^{20}$  s, or the age of the Universe)
- Each time range is studied with a different experimental technique

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

\*Under very special circumstances

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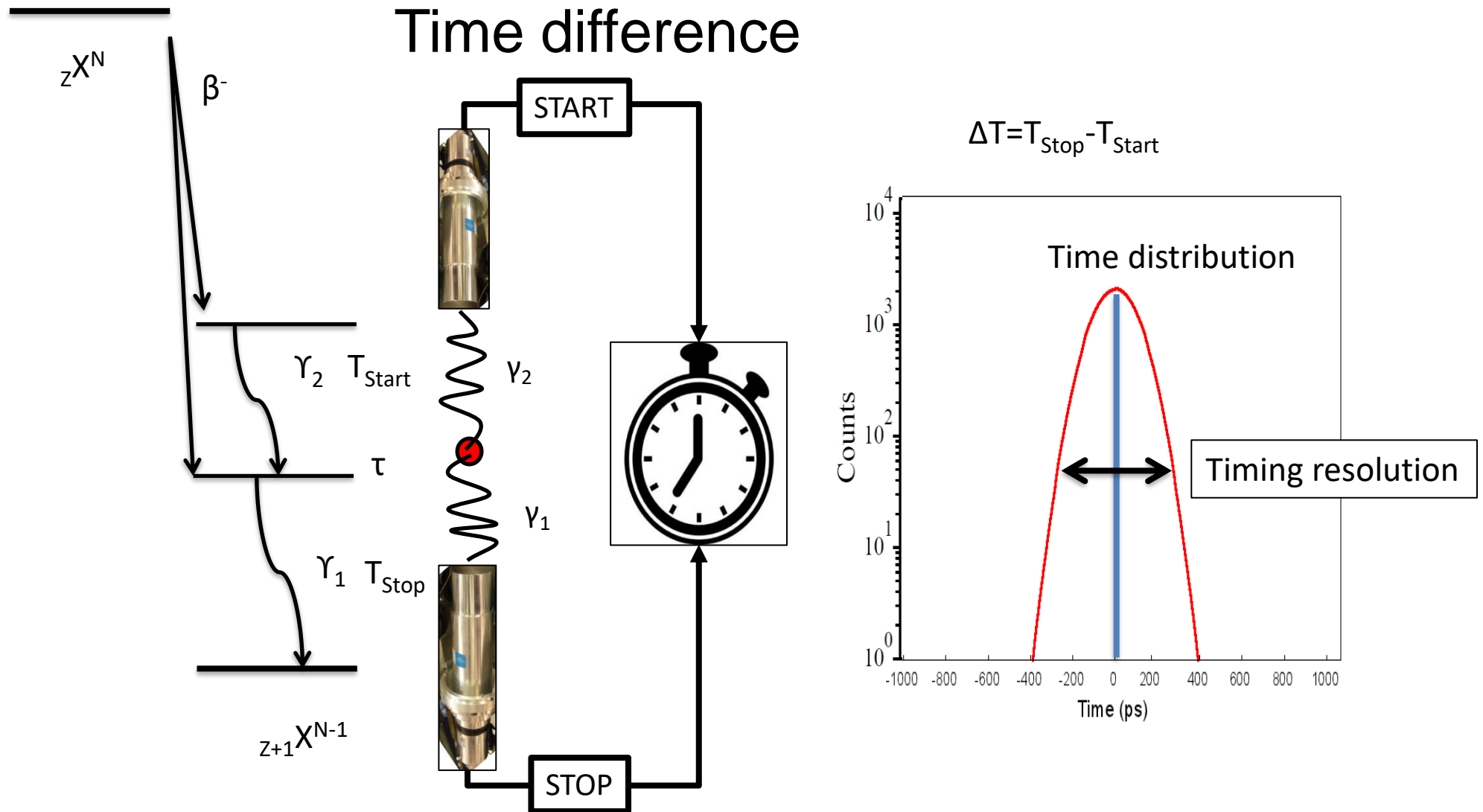
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Coulex	$0^*$	$\infty^*$

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# How short is a picosecond?

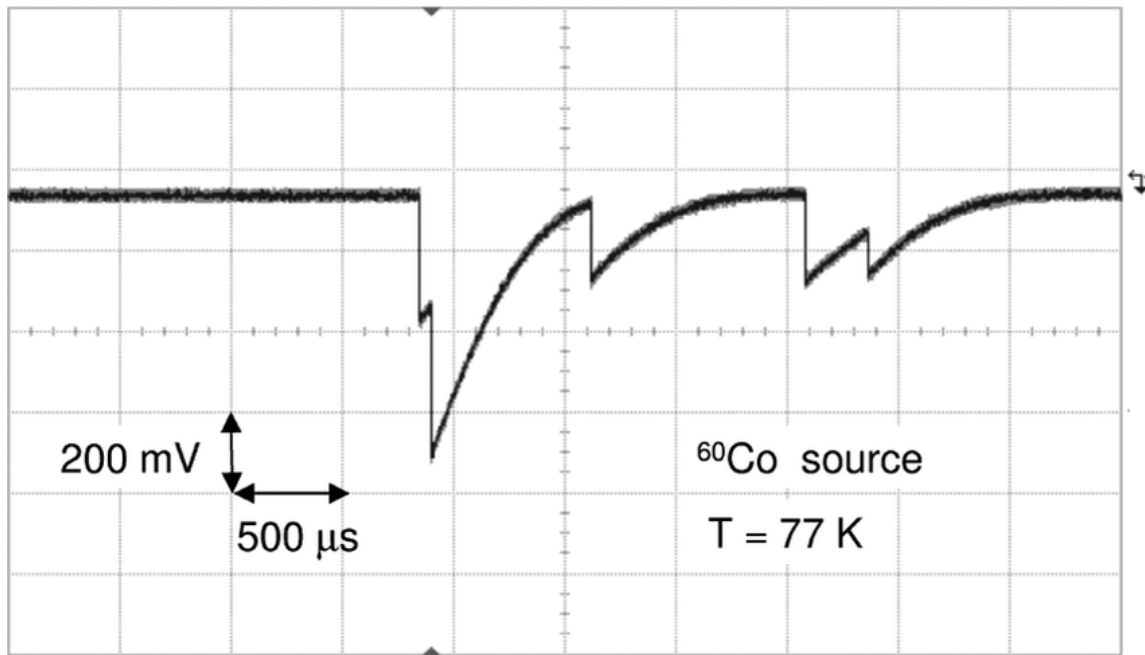
- 1 picosecond =  $10^{-12}$  seconds
- That's 0.000000000001 seconds
- It takes photons (fastest particles in the universe)  $\sim 3.3$  ps to travel 1 mm in vacuum
- When working in this time frame, the speed of light cannot be considered instantaneous anymore
- Indeed,  $c$  is one of the main limitations

# Simplified nuclear electronic timing

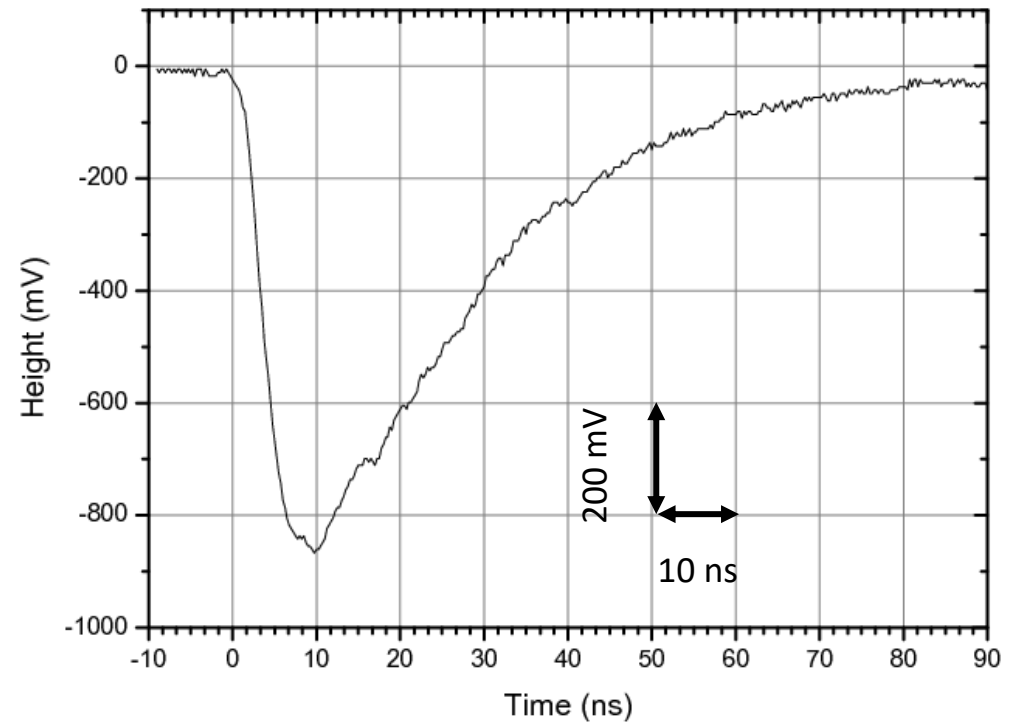


# Signals

## Semiconductor



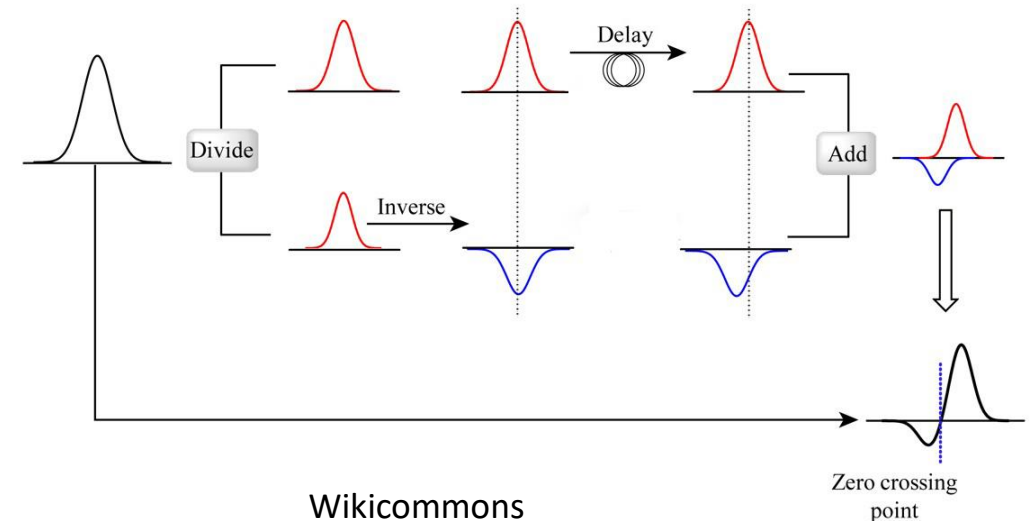
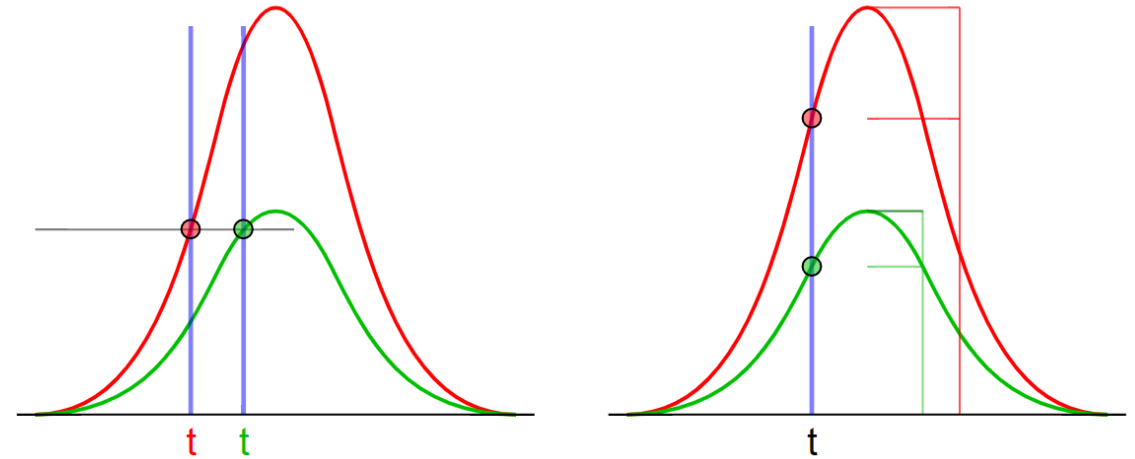
## Scintillator





# Constant fraction discriminator

- How to determine when the signal arrives
- Leading edge (threshold trigger)
  - Faster
  - Strong energy dependency (time walk)
- Constant fraction discriminator
  - Slightly slower
  - Greatly reduces energy dependency

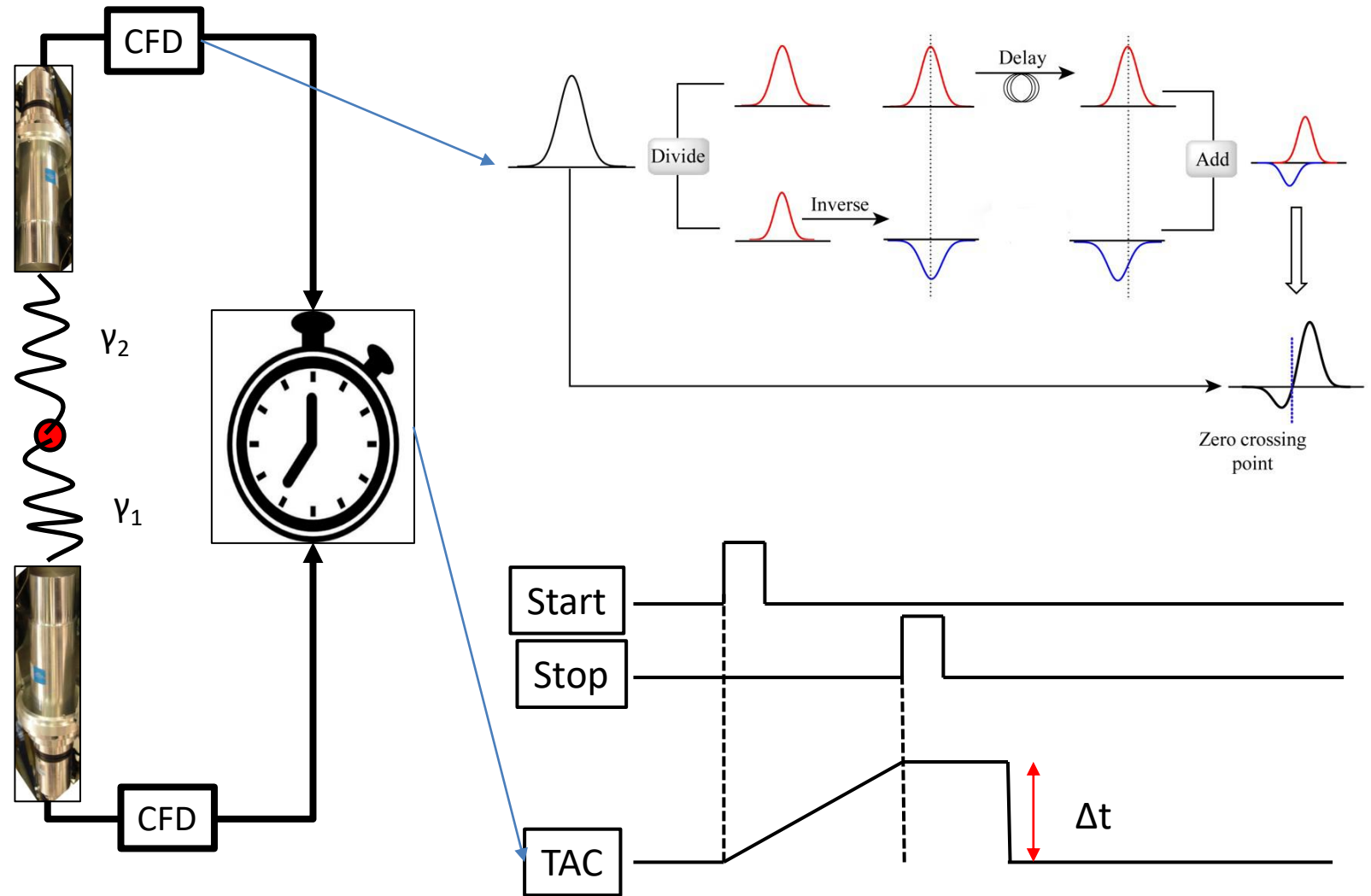


Wikicommons

Zero crossing point

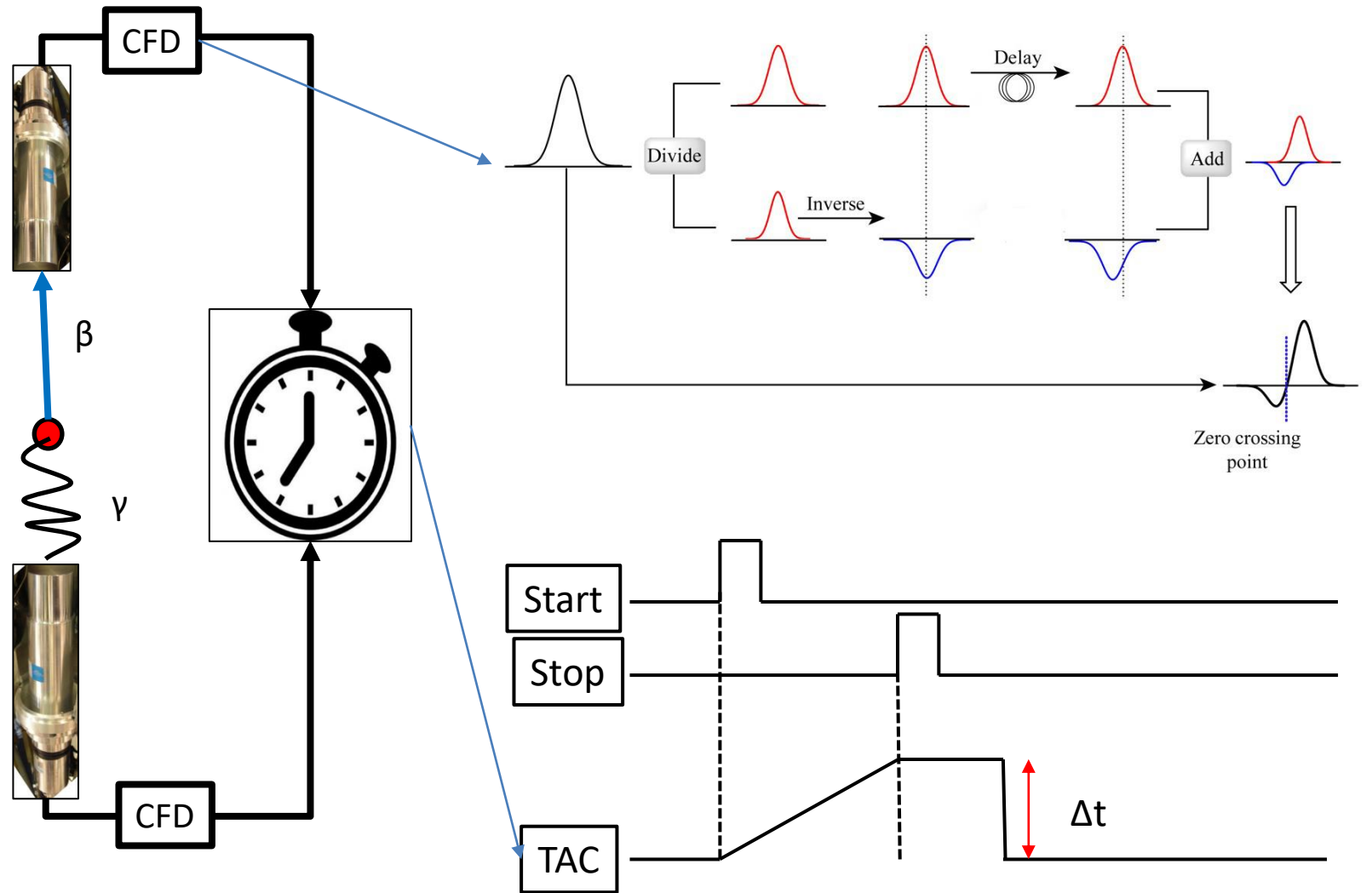
# Electronics

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



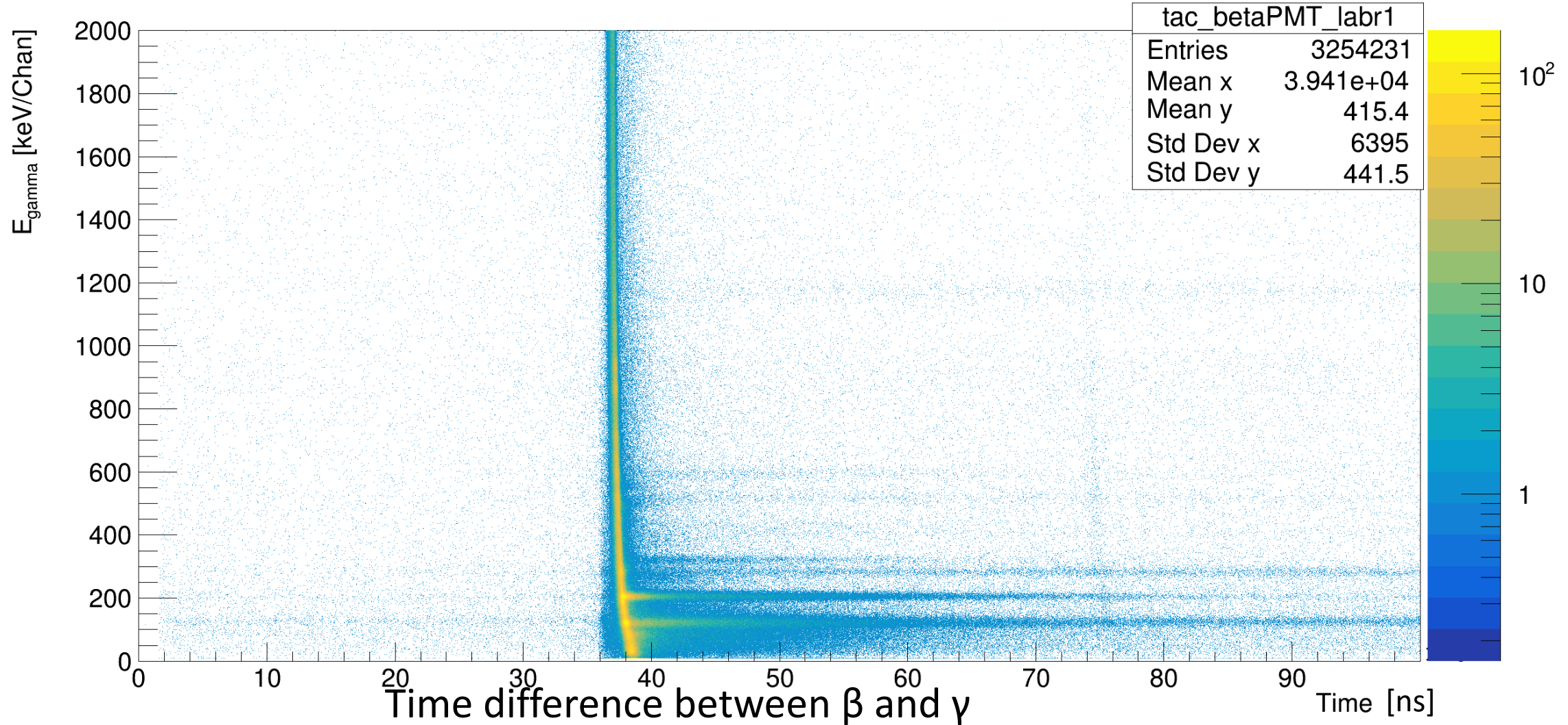
# Electronics

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



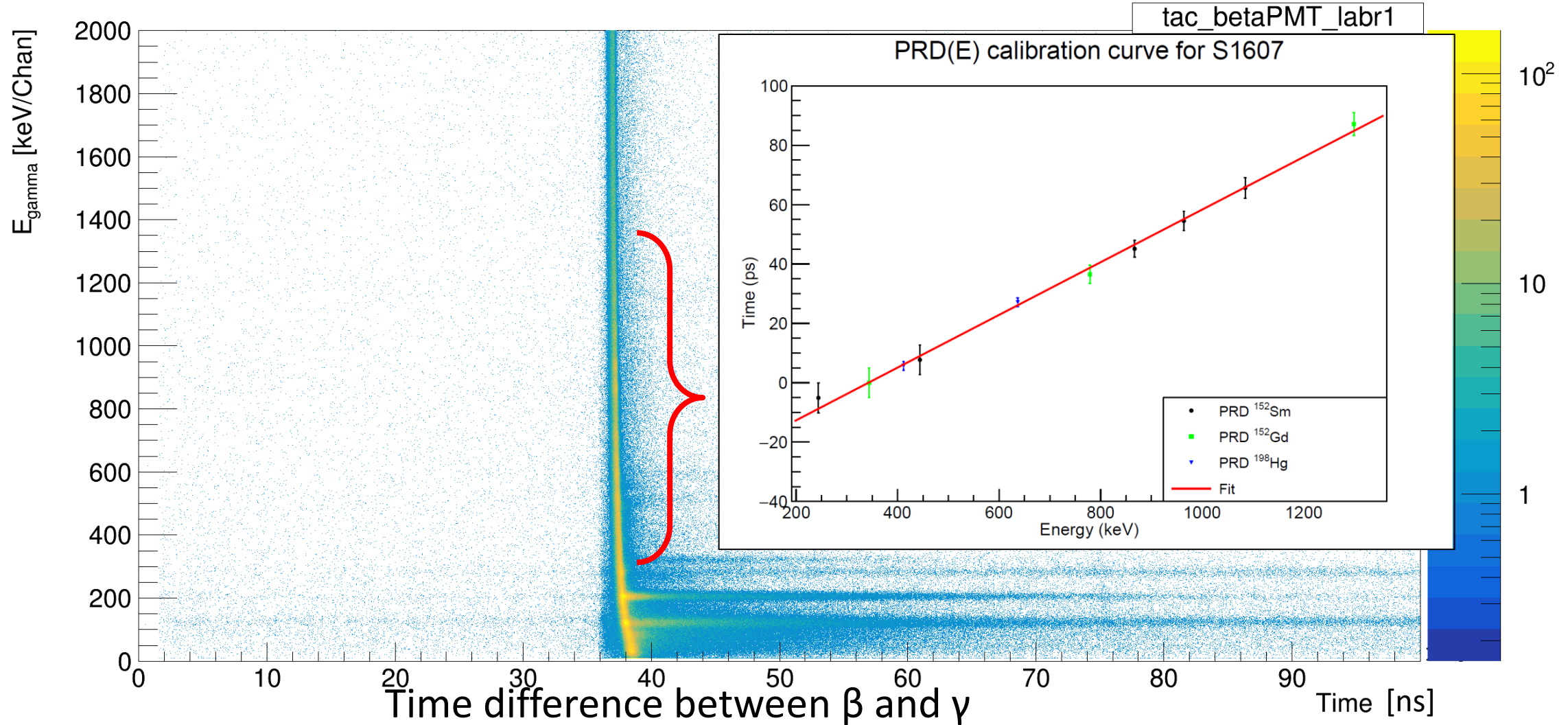
# Time walk

TAC START: betaPMT - STOP: LaBr1



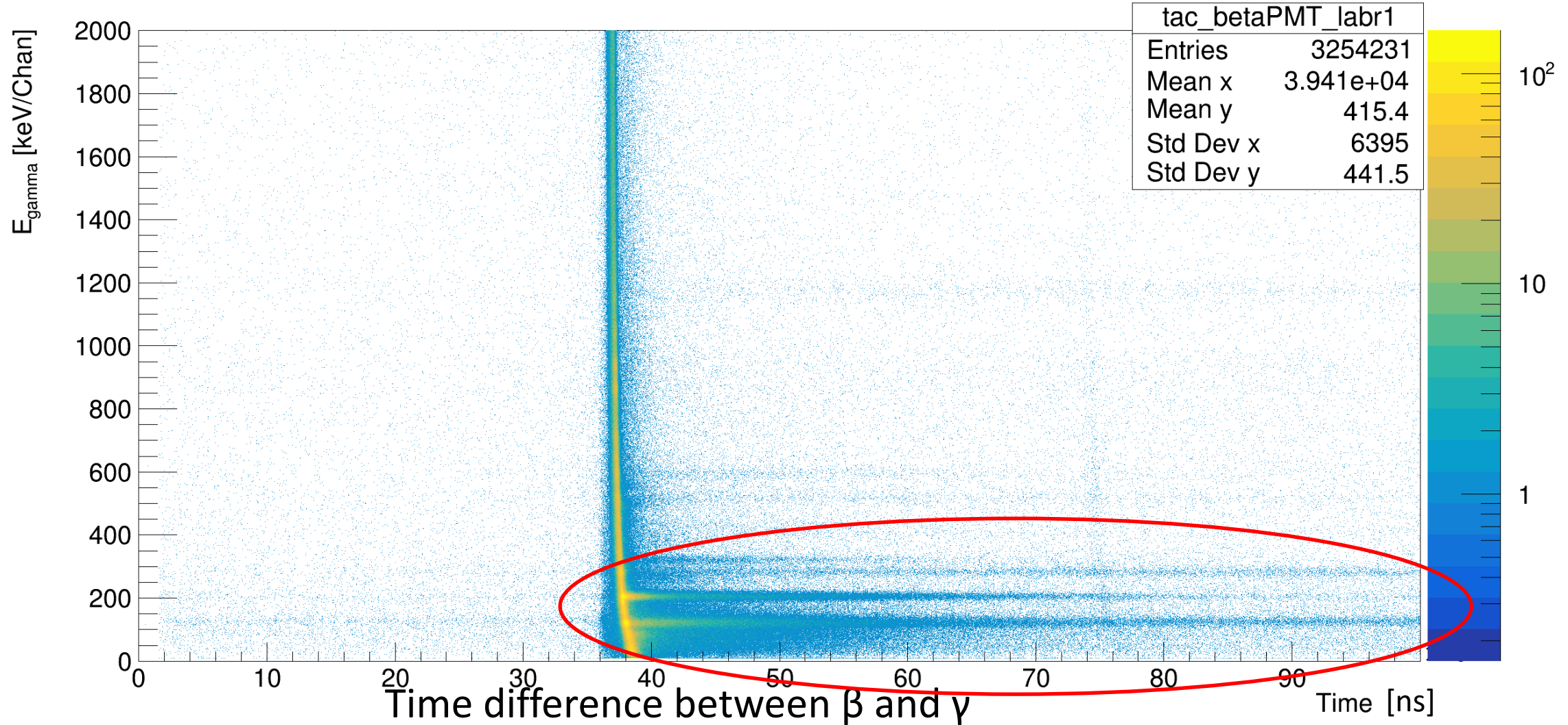
# Time walk

TAC START: betaPMT - STOP: LaBr1



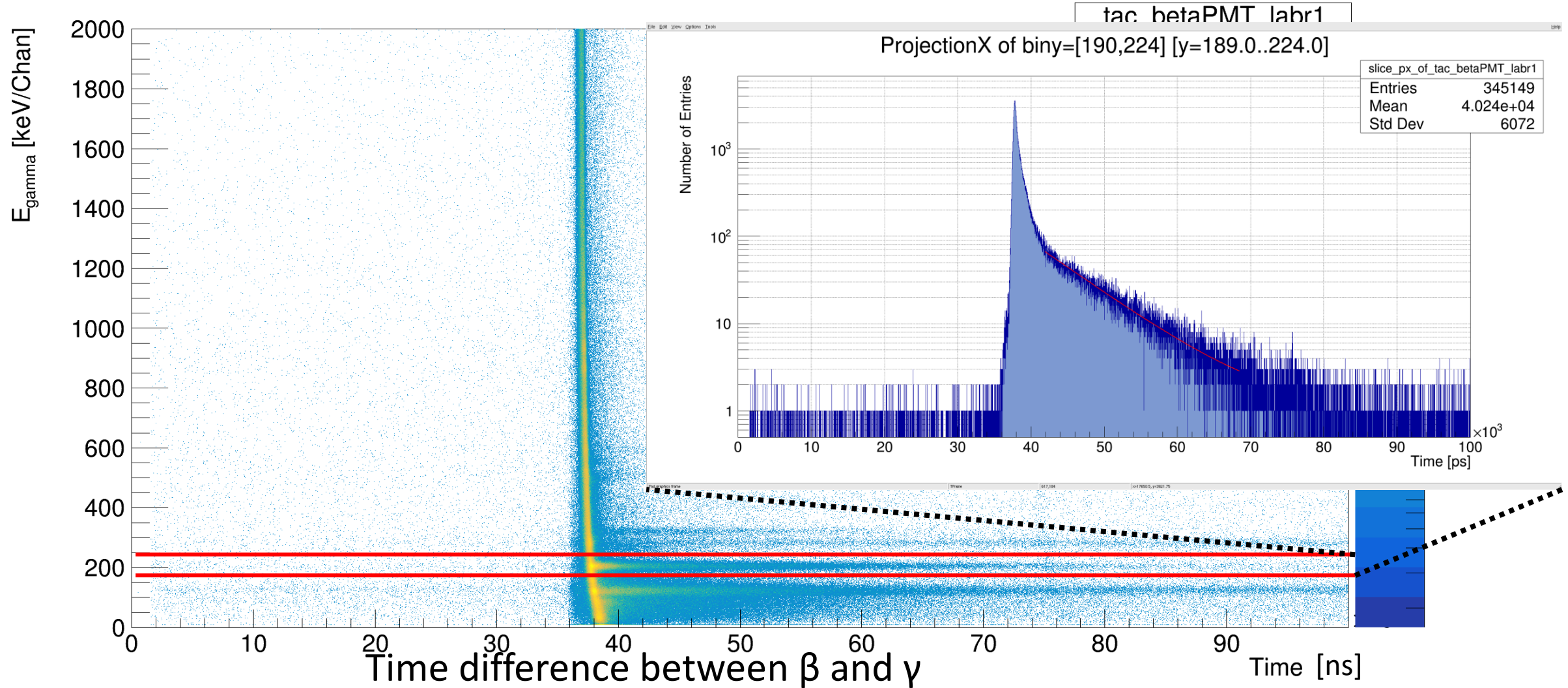
# Bonus question

TAC START: betaPMT - STOP: LaBr1



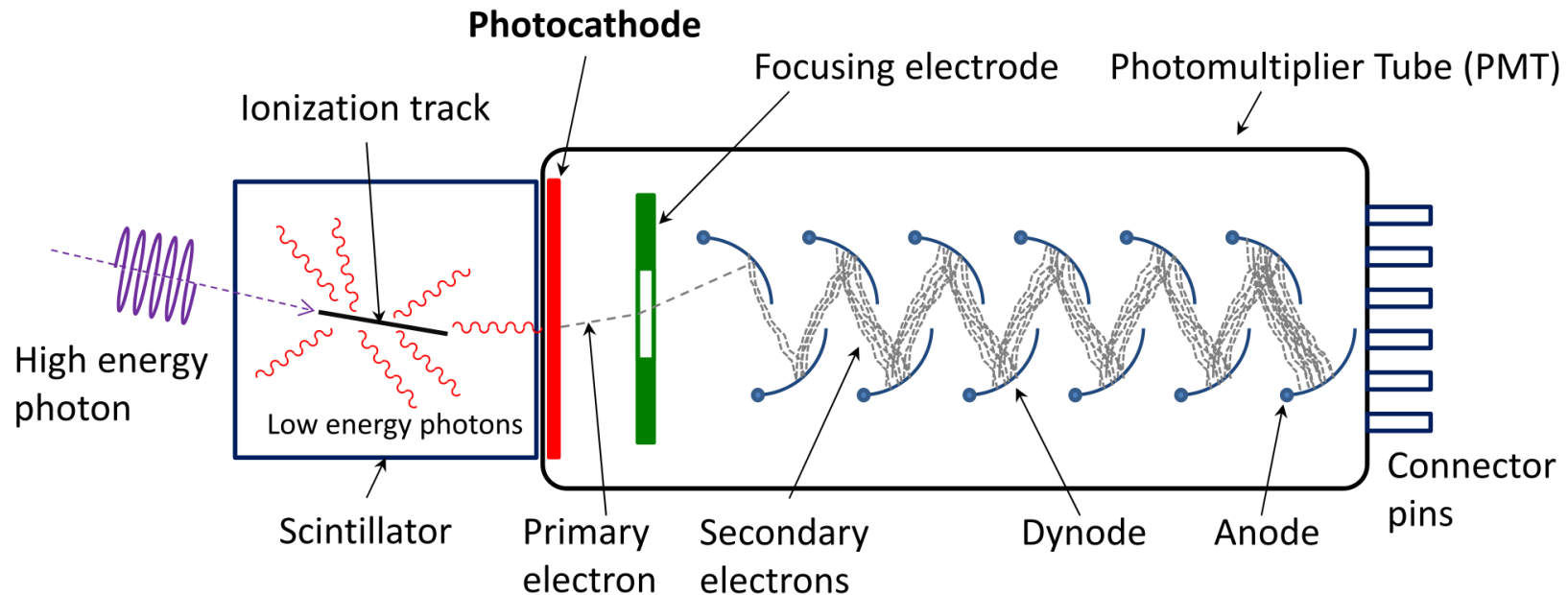
# Long lifetime

TAC START: betaPMT - STOP: LaBr1



# Measuring gamma-rays

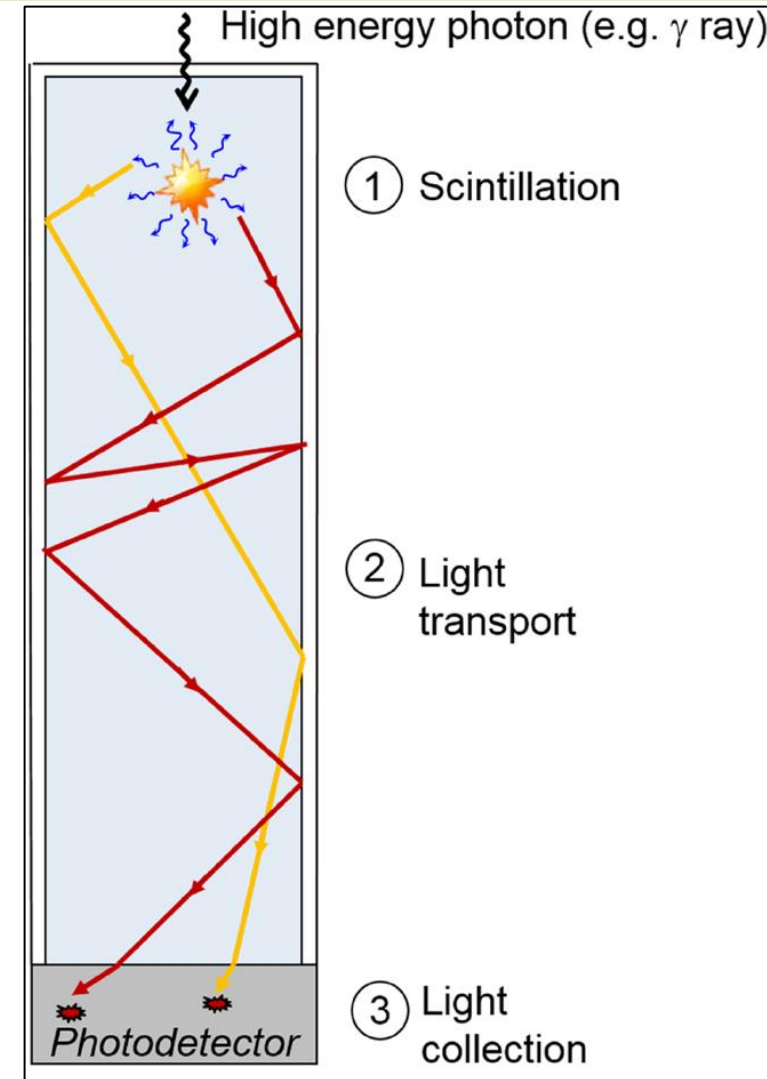
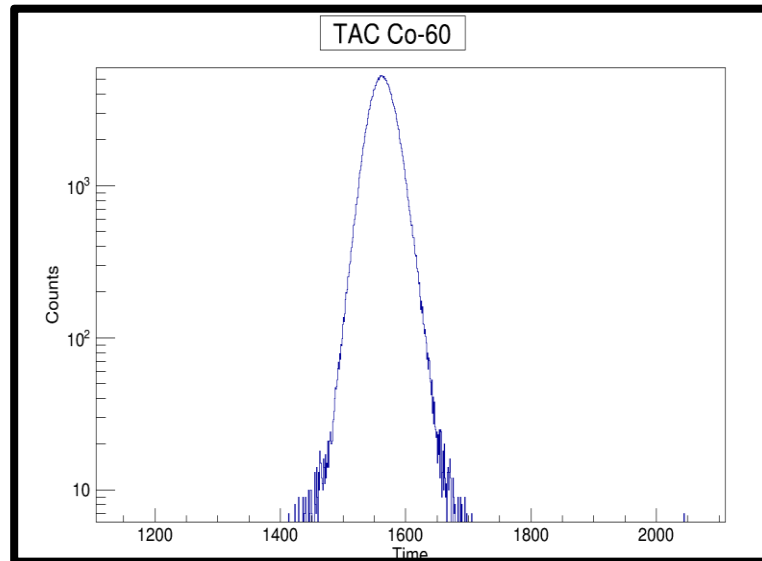
- Scintillators (like  $\text{LaBr}_3(\text{Ce})$ ) are the fastest detectors nowadays
- The incident photon excites the crystal molecules
- They quickly de-excite emitting UV (usually)
- Secondary photons absorbed by the photocathode
- The electrons from the photoelectric effect give us the signal
- Photoelectric effect is more likely with UV rays





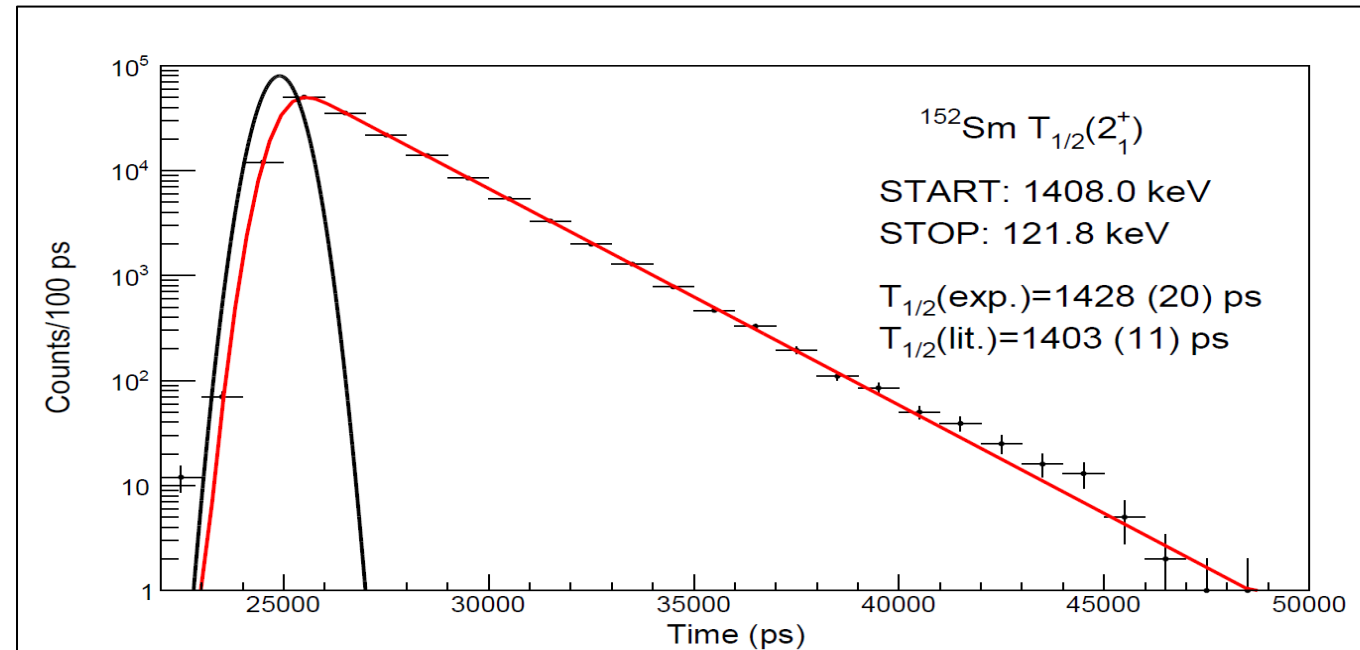
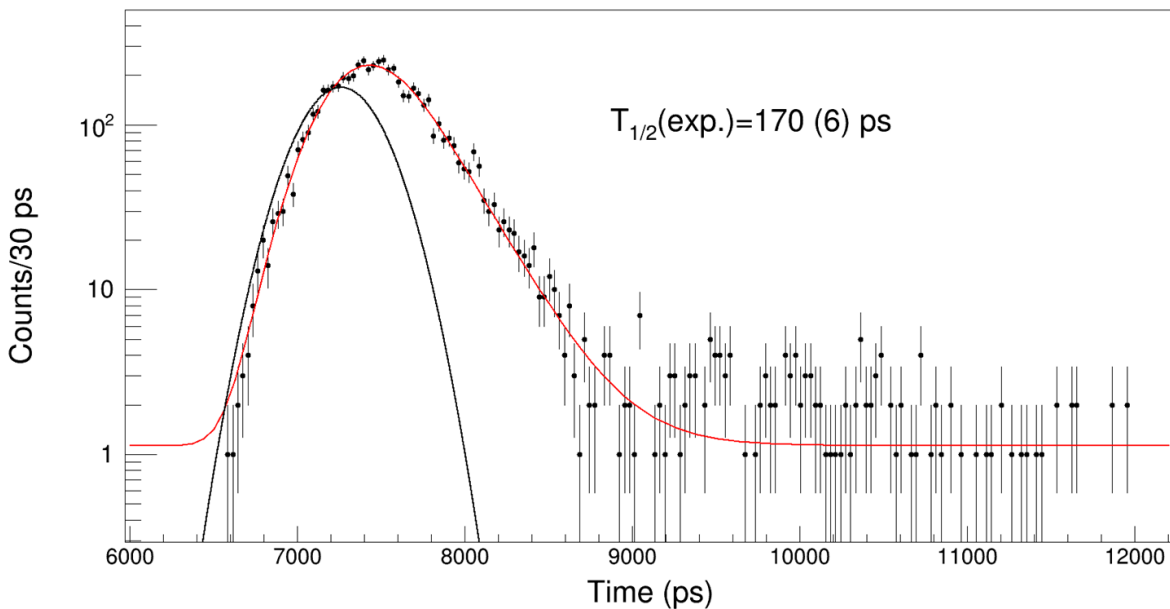
# Timing resolution

- Timing resolution is the time-width of a prompt ( $\tau \sim 0$  ps) signal
- Scintillators crystals are a few cm long and wide
- Light takes time bouncing inside the crystal (1 mm  $\sim$  3.3 ps)
- In meta-materials, timing resolution mainly depends on crystal size

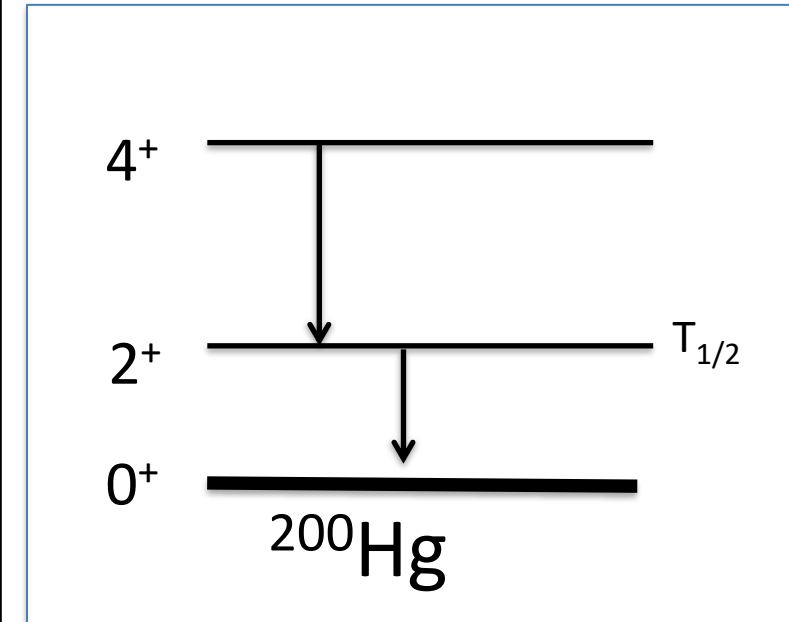
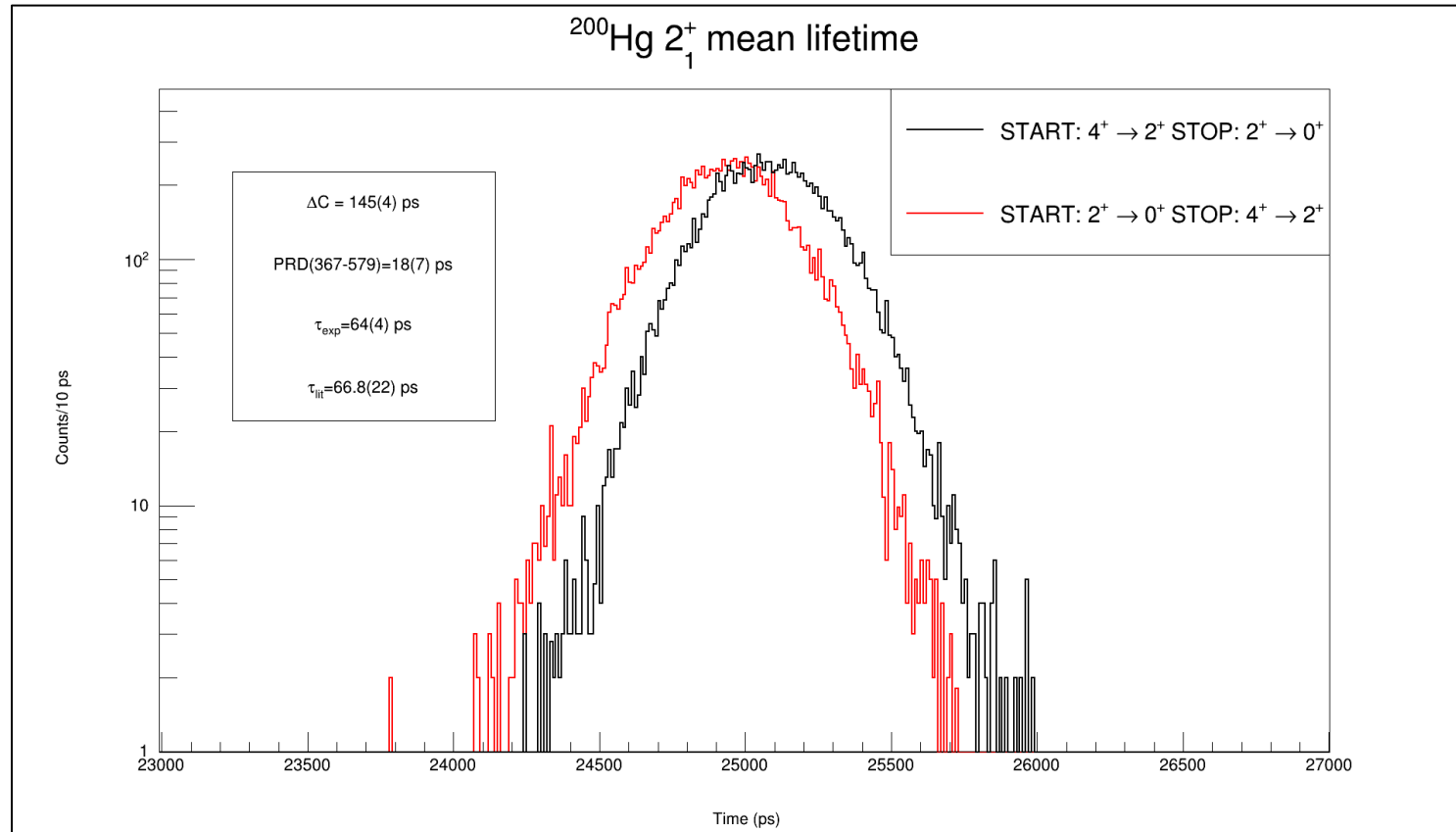


# Convolution method

- Prompt part may be approximated to a Gaussian shape, down to 3 orders of magnitude
- Slope method may be used for  $T_{1/2} \sim$  timing resolution and above
- 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  $^{200}\text{Hg}$   
 $T_{1/2}(\text{literature})=46.4(4)$  ps  
 $T_{1/2}(\text{experiment})=44(3)$  ps

# Generalized centroid difference method

The centroid of the time distribution will be given by:

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

$\tau_0$  = a constant delay of the setup, cannot be determined (in general)

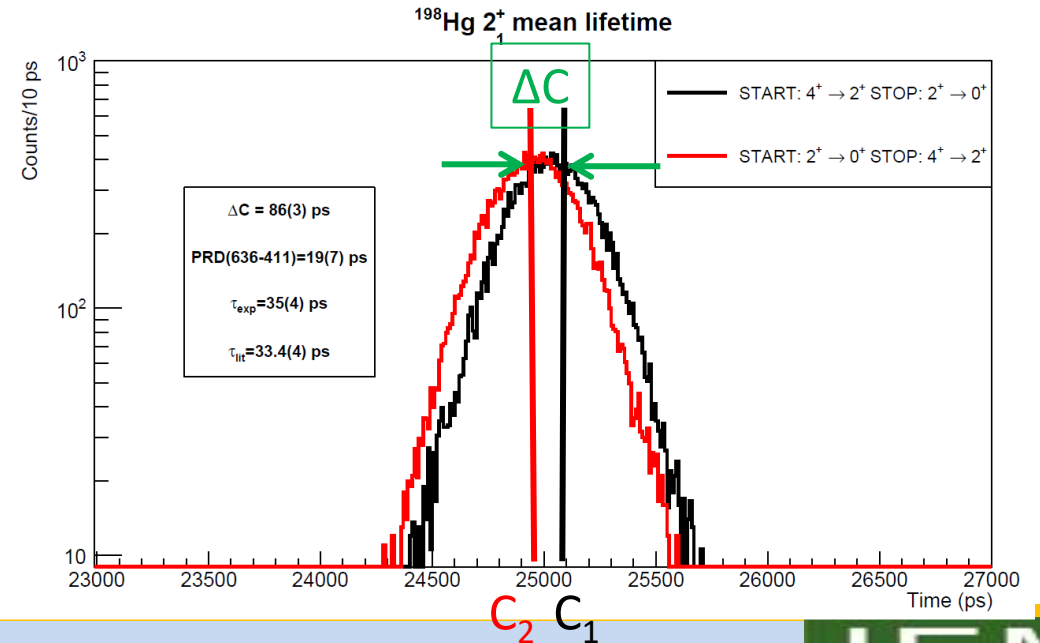
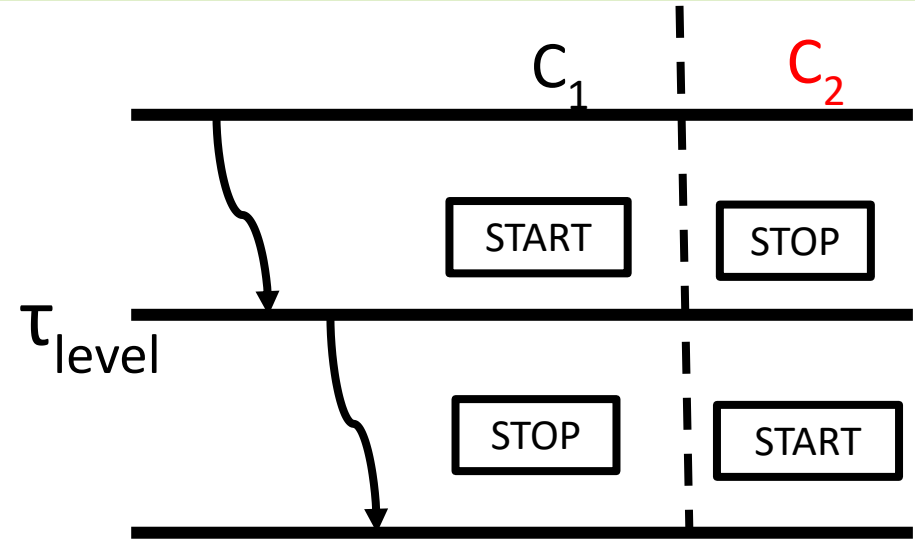
$\tau_{\text{level}}$  = Lifetime we want to measure

$$C_1 = \tau_{\text{level}} + \tau_0$$

$$C_2 = -\tau_{\text{level}} + \tau_0$$

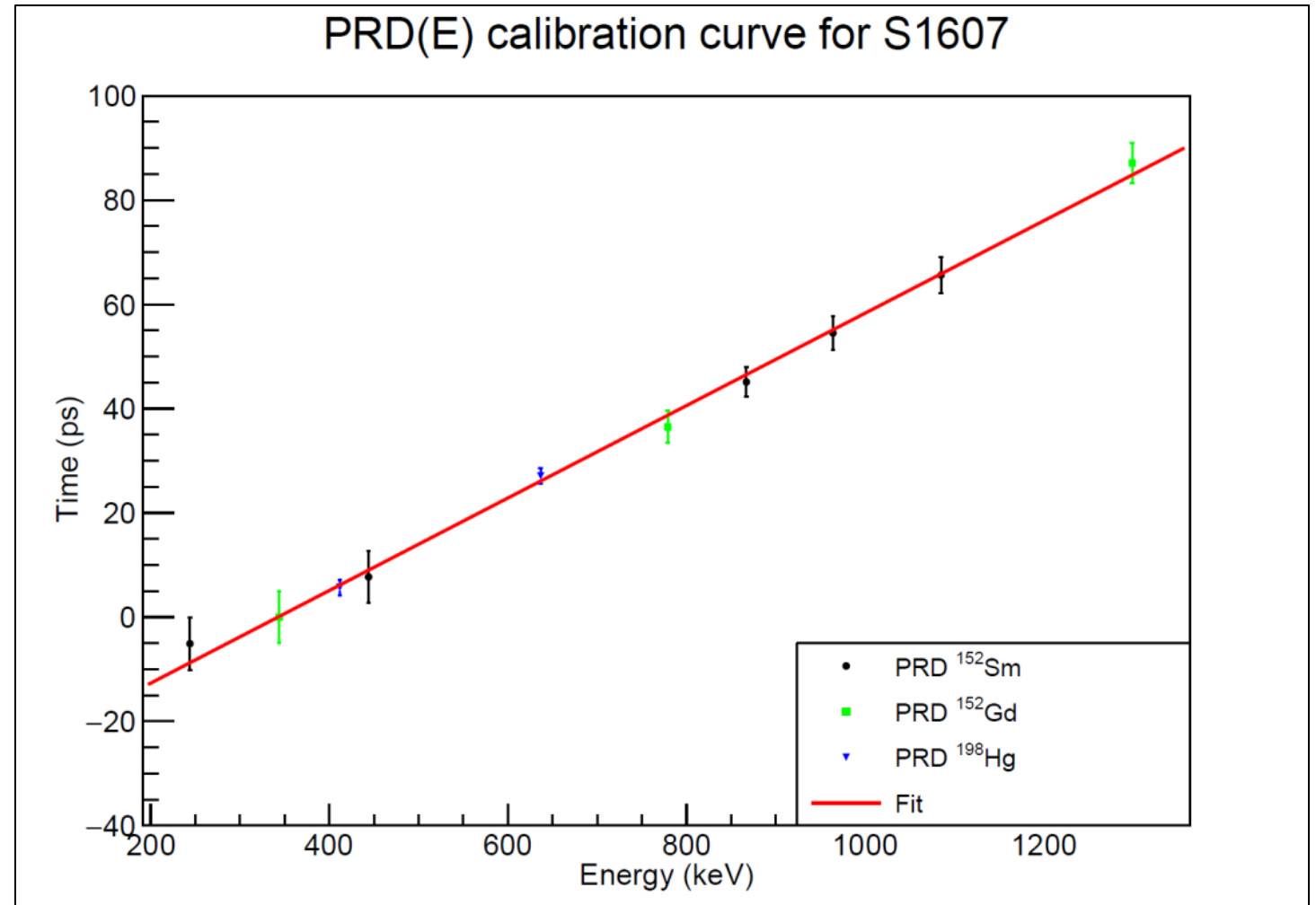
$$\Delta C = C_1 - C_2 = \tau_{\text{level}} + \cancel{\tau_0} - (-\tau_{\text{level}} + \cancel{\tau_0}) = 2\tau_{\text{level}}$$

$$\Delta C = 2\tau_{\text{level}}$$

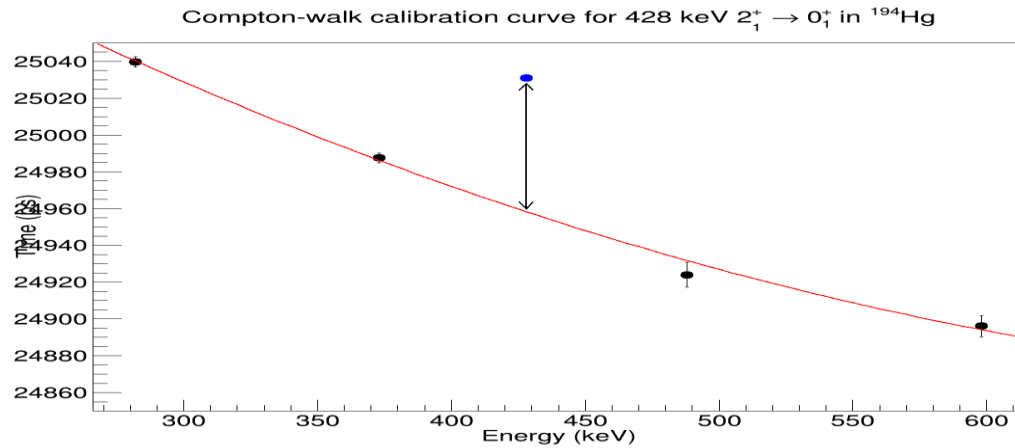


# Relative PRD calibration curve

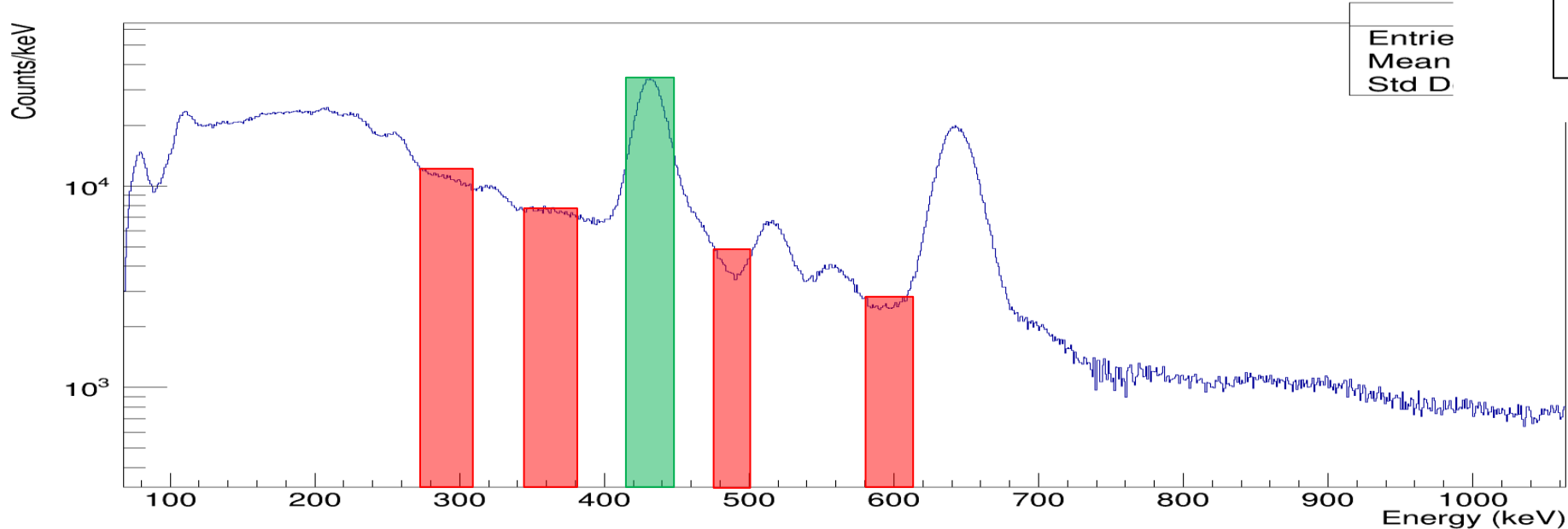
- Timing response calibration
- $^{152}\text{Eu}$  commercial source
- Lifetimes are precisely measured
- Values are corrected by the literature lifetimes
- Uncertainty  $\sim 5$  ps mainly from Compton background



# Compton correction



START  $\text{LaBr}_3(\text{Ce})$  energy with HPGe gate in  $5_1^- / 6^+ \rightarrow 4^+$  749.0/735.0 keV transition

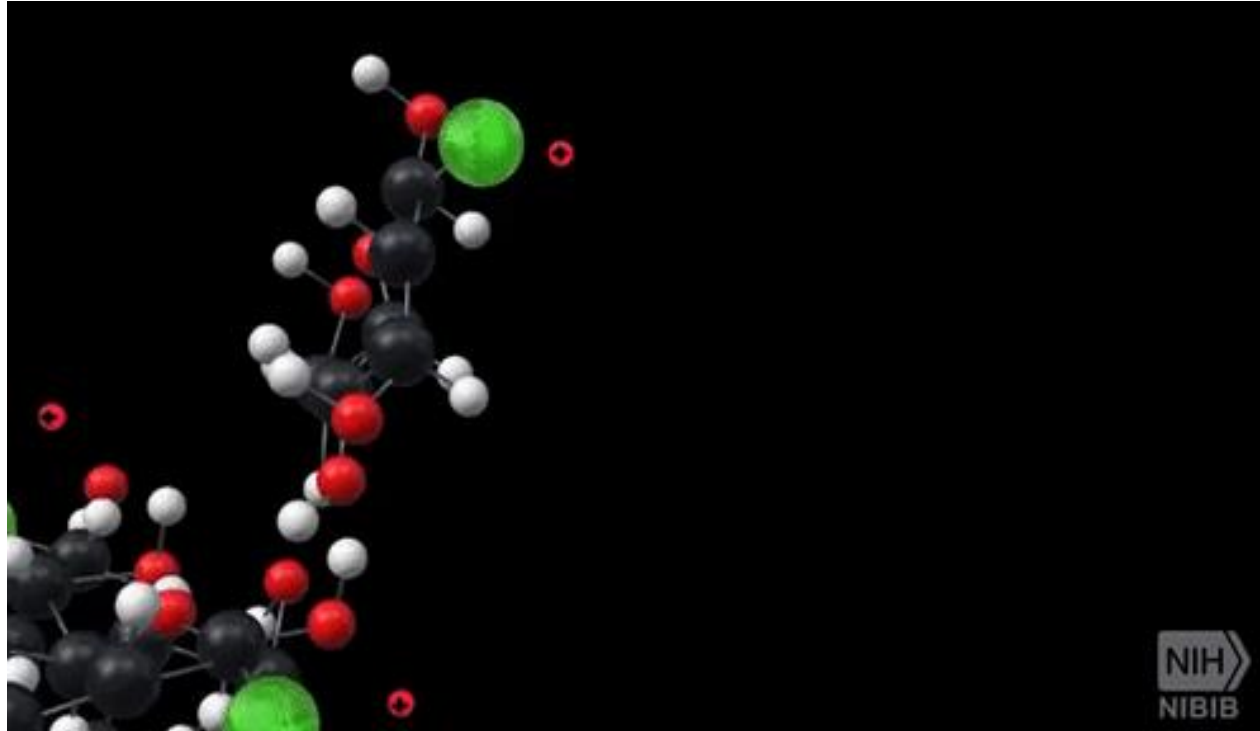


$$A_T C_T = A_P C_P + A_C C_C$$

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

# Practical applications: PET-TOF

# Positron-electron annihilation

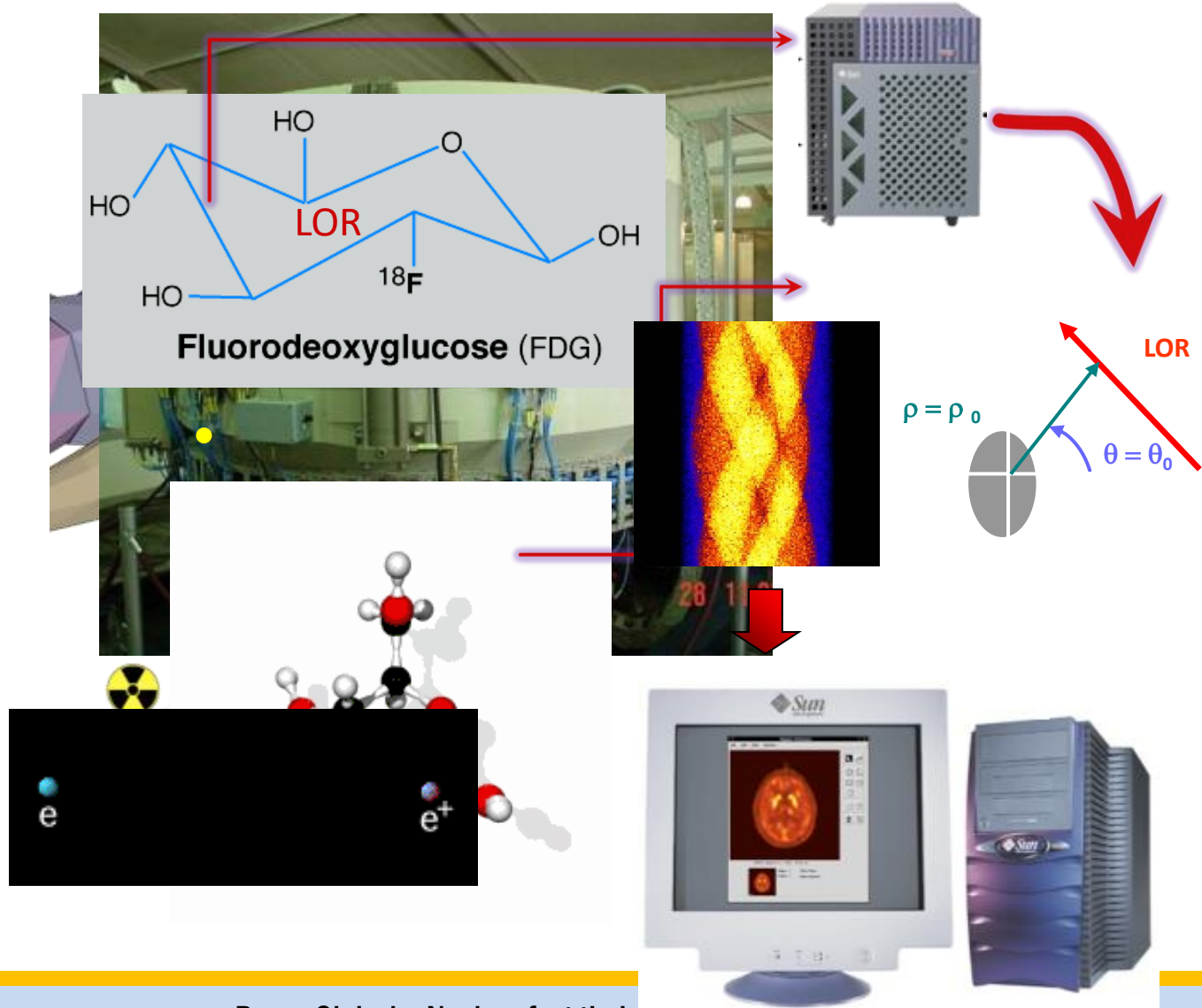
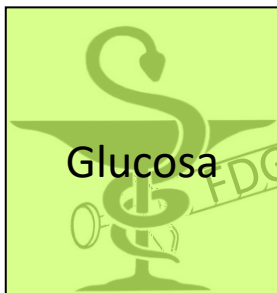
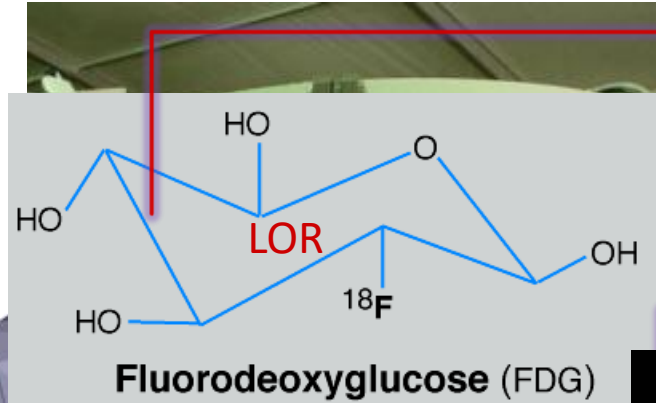
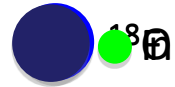


- Positron ( $e^+$ ) is the anti-particle of the electron
- If they touch, they annihilate  $E=mc^2$
- Mass of  $e^-/e^+$  is  $511 \text{ keV}/c^2$
- Momentum conservation, **two** 511-keV photons are emitted in **opposite directions**



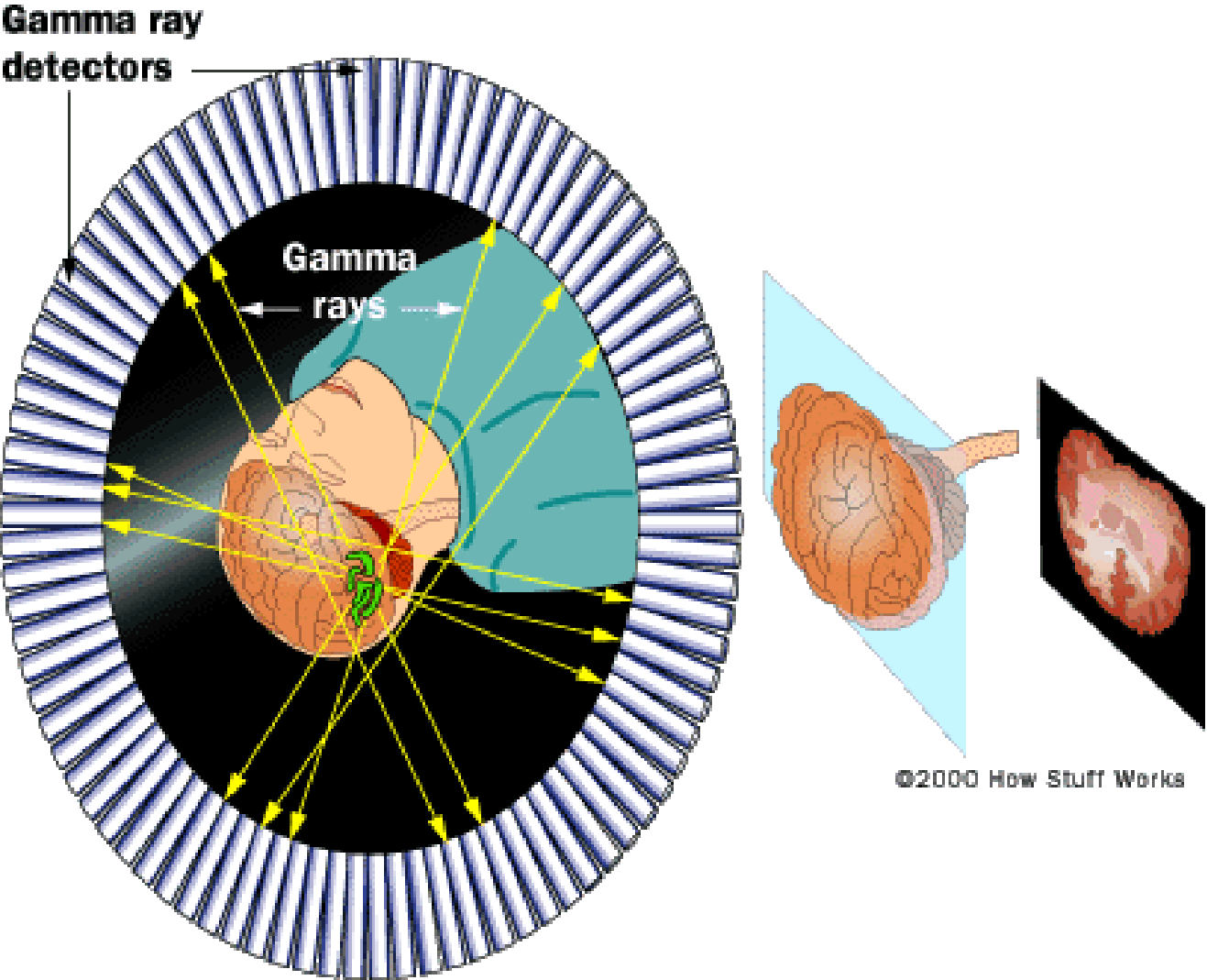
# Positron Emission Tomography (PET)

Positron Emission Tomography (PET)



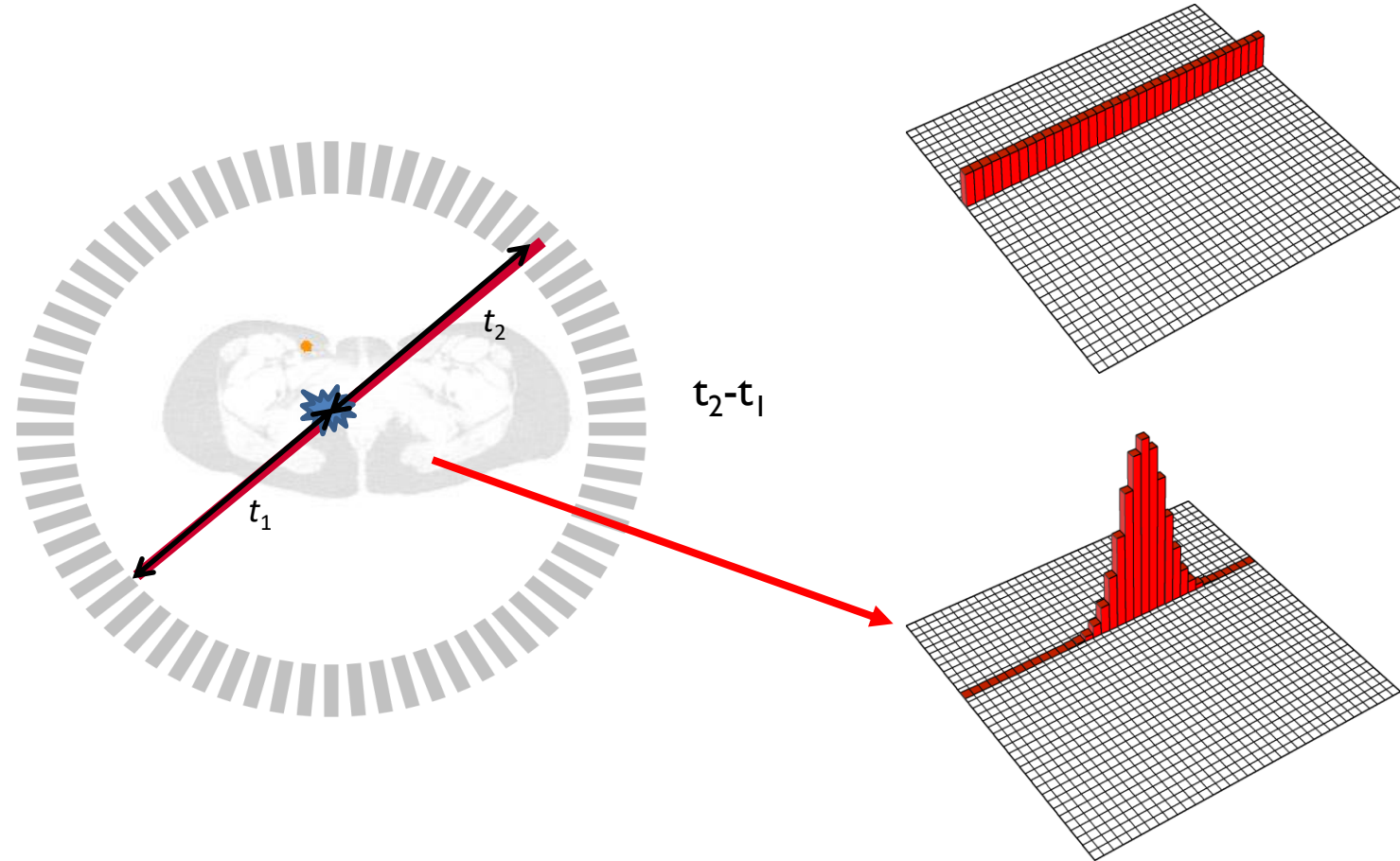
Courtesy of K. Abushab UCM-Spain

# Positron Emission Tomography (PET)



©2000 How Stuff Works

# PET – Time of Flight (ToF)

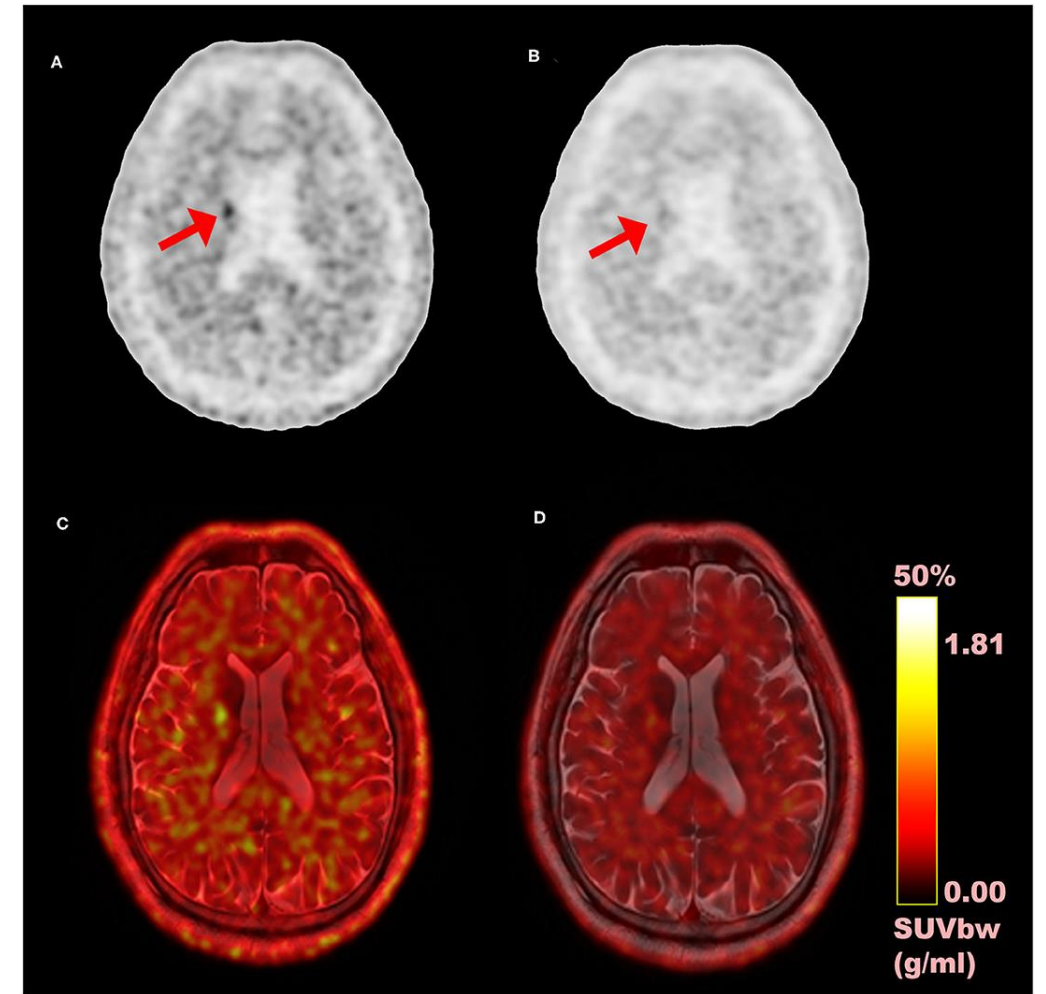


# Increased resolution

- ToF greatly increases resolution
- Allows for earlier detection
- Development of detectors and electronics
- Goal of 10 ps resolution
- Lunabrain Project in Madrid

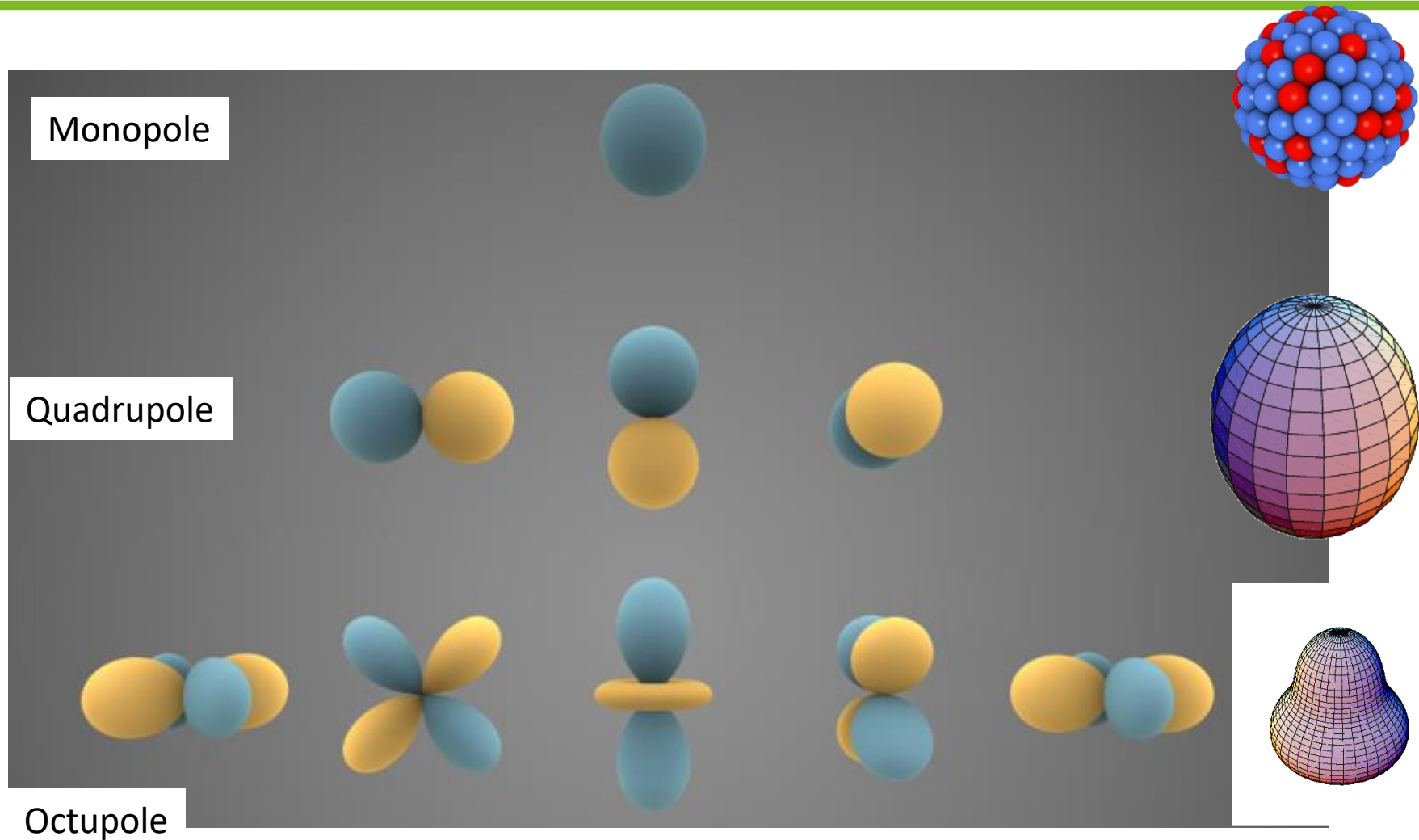
With ToF

Without ToF



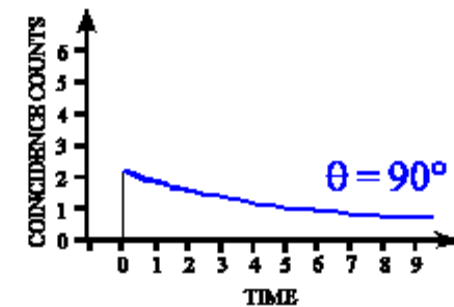
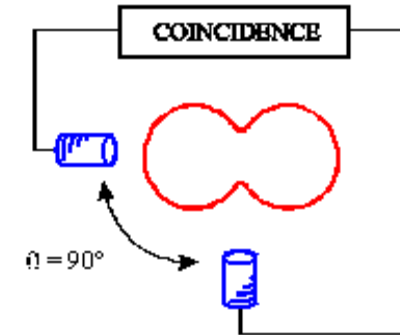
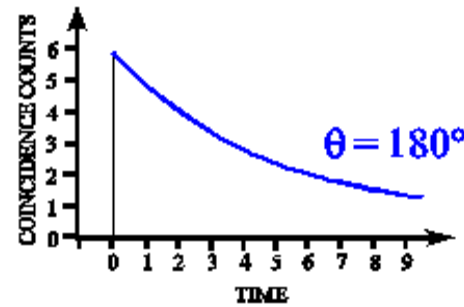
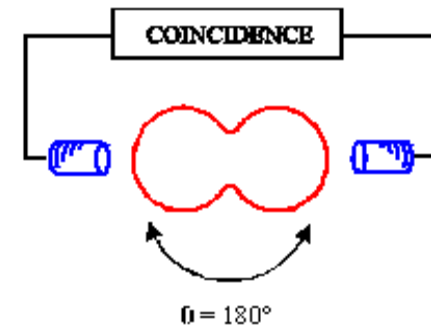
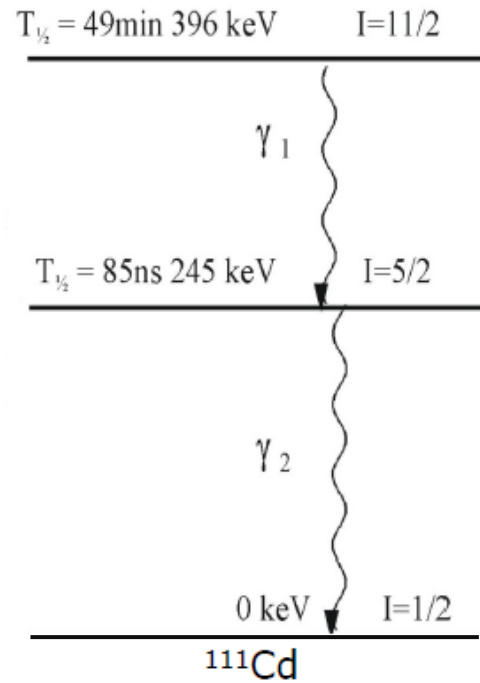
# Practical applications: Perturbed Angular Correlations

# Multipole radiation



# PAC Spectroscopy

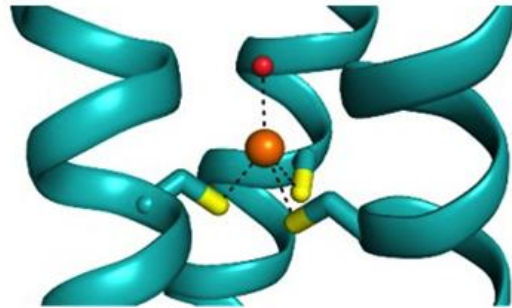
$\gamma$  -  $\gamma$  angular correlation



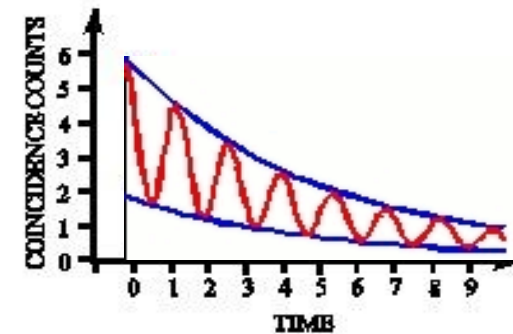
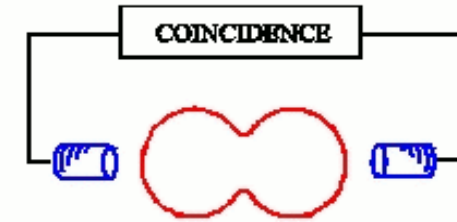
(www.uni-leipzig.de)

# Perturbed angular correlation

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



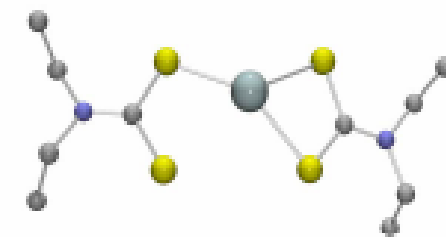
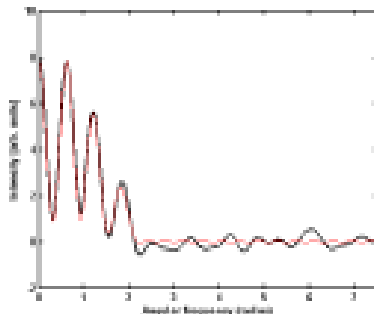
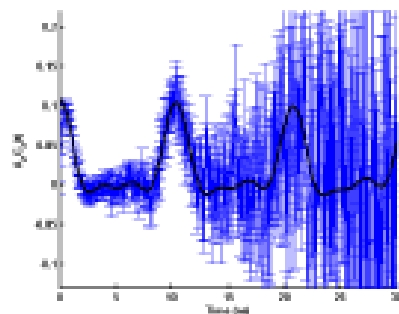
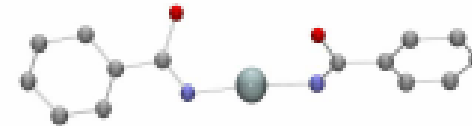
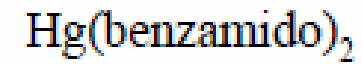
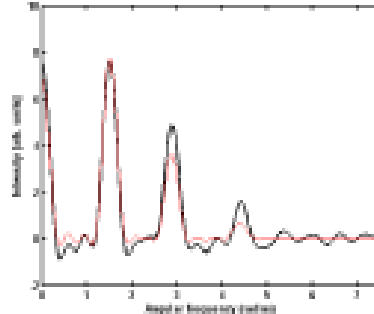
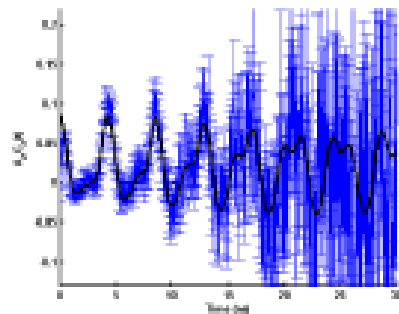
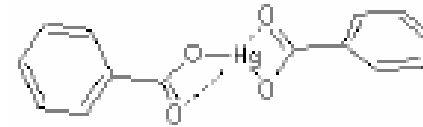
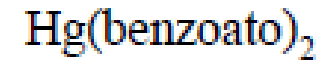
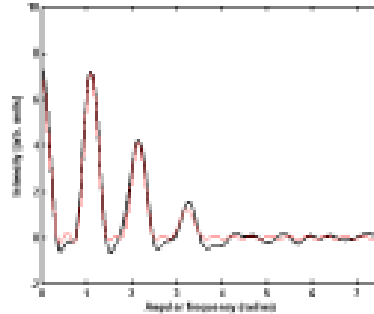
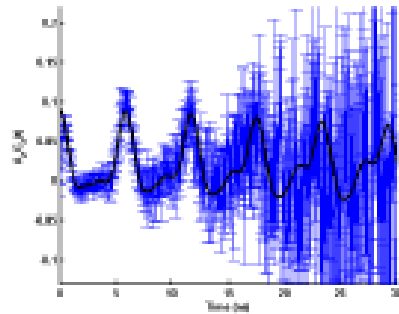
- This technique requires detectors with good energy resolution and excellent timing resolution.
- $\text{LaBr}_3$  scintillators are the ideal choice.



(www.uni-leipzig.de)

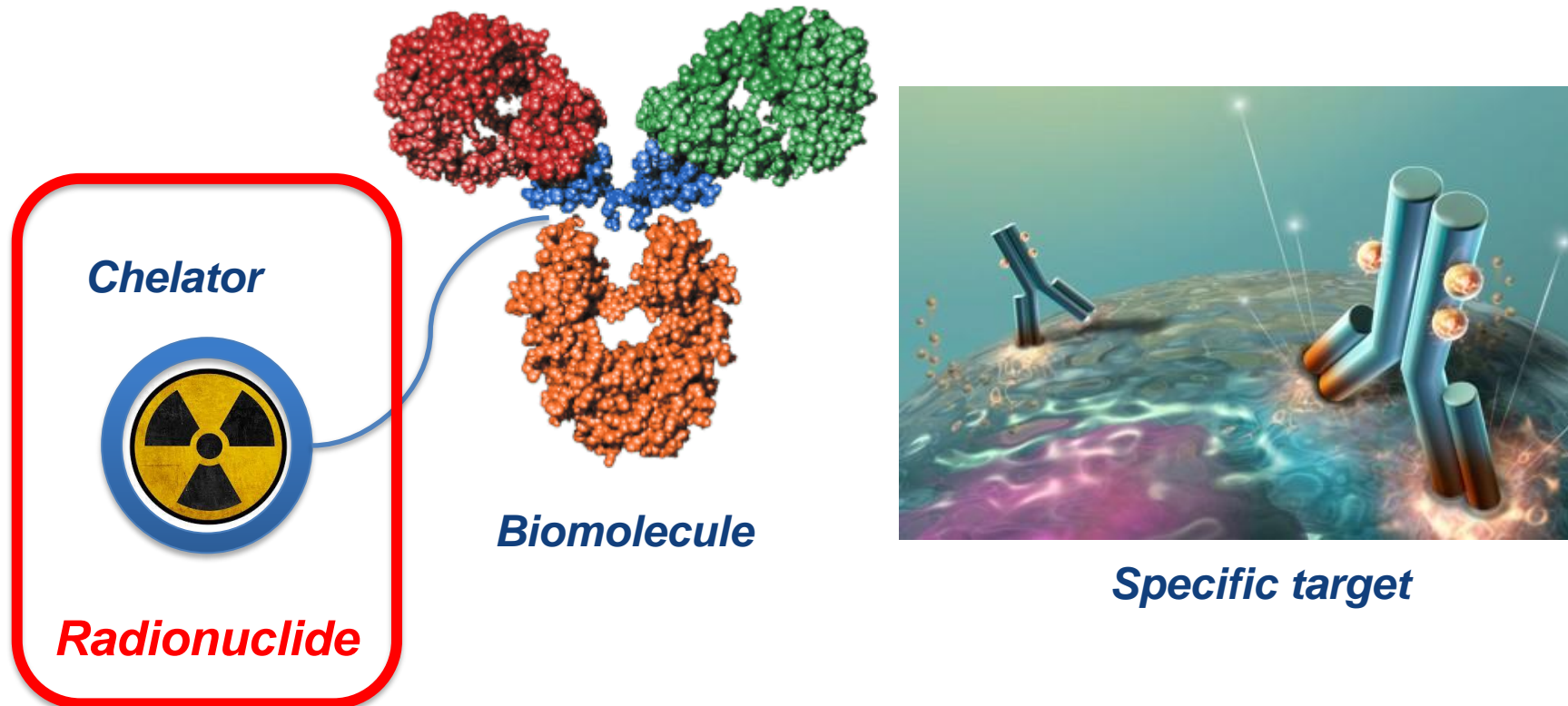


# PAC Spectroscopy reveals coordination chemistry

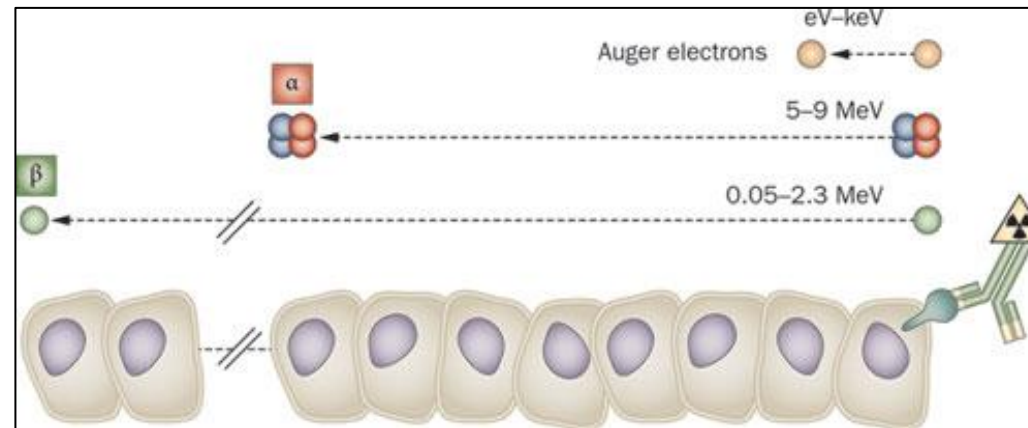
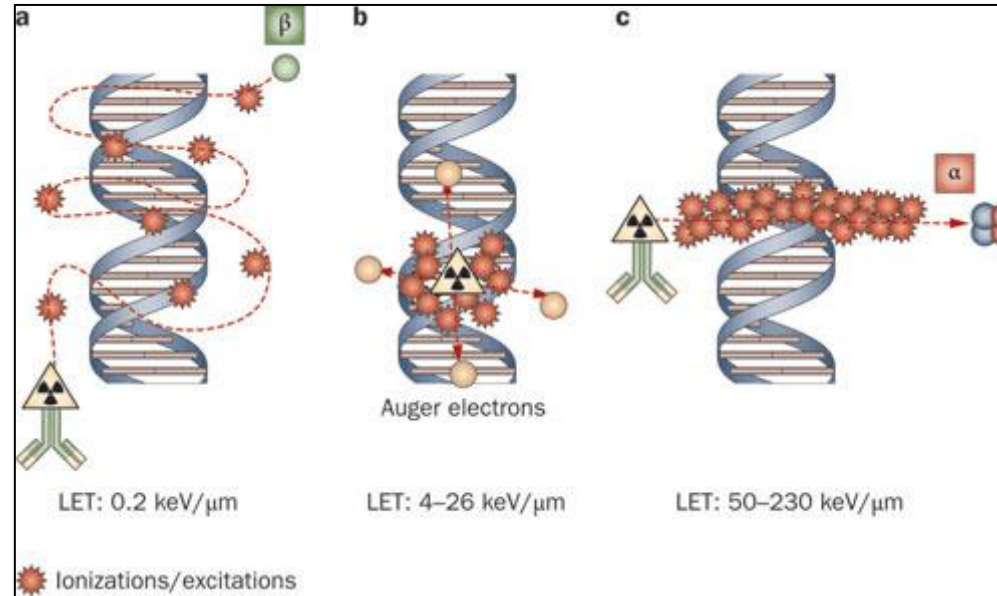


# Radiopharmaceuticals

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”



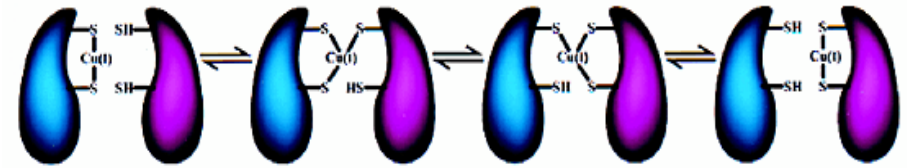
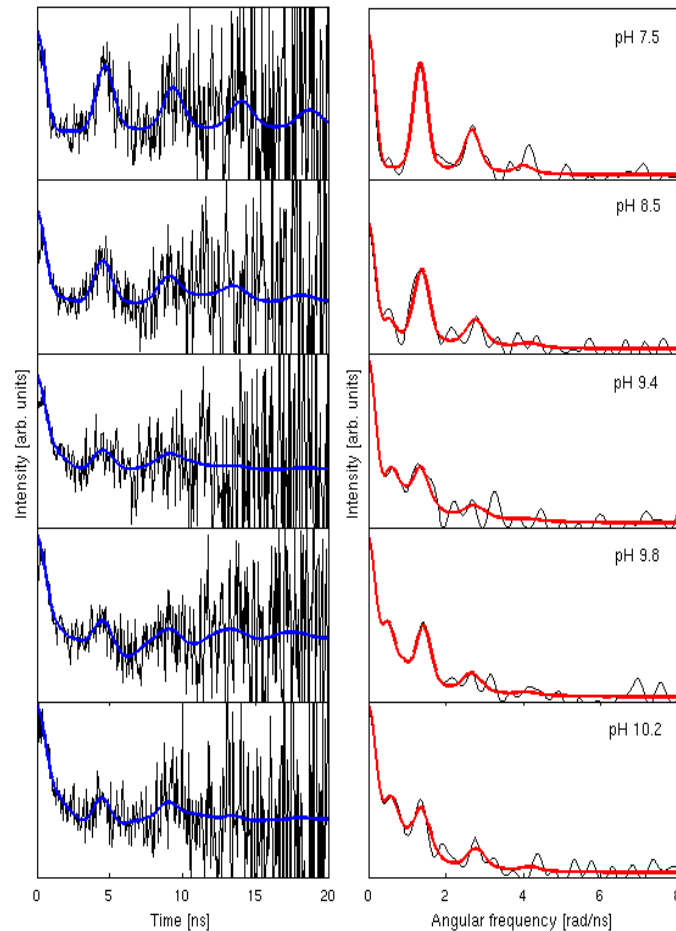
# Targeted Radionuclide Therapy (TRT)



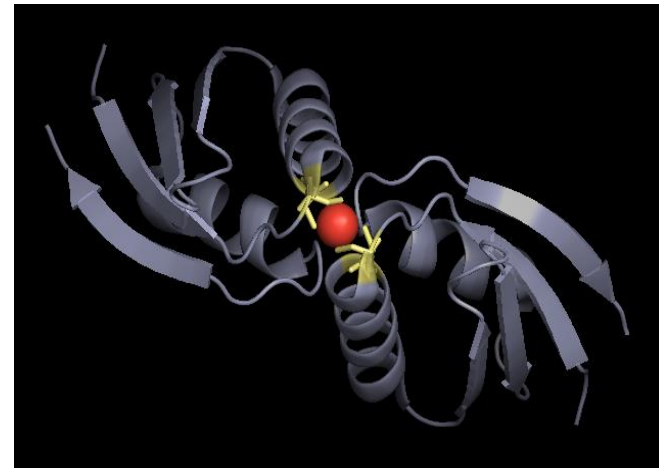
- *Pouget J.-P. et al. (2011) Clinical radioimmunotherapy—the role of radiobiology Nat. Rev. Clin. Oncol.*

# PAC Spectroscopy characterizes protein-protein interactions

The metal ion binding site changes with pH level

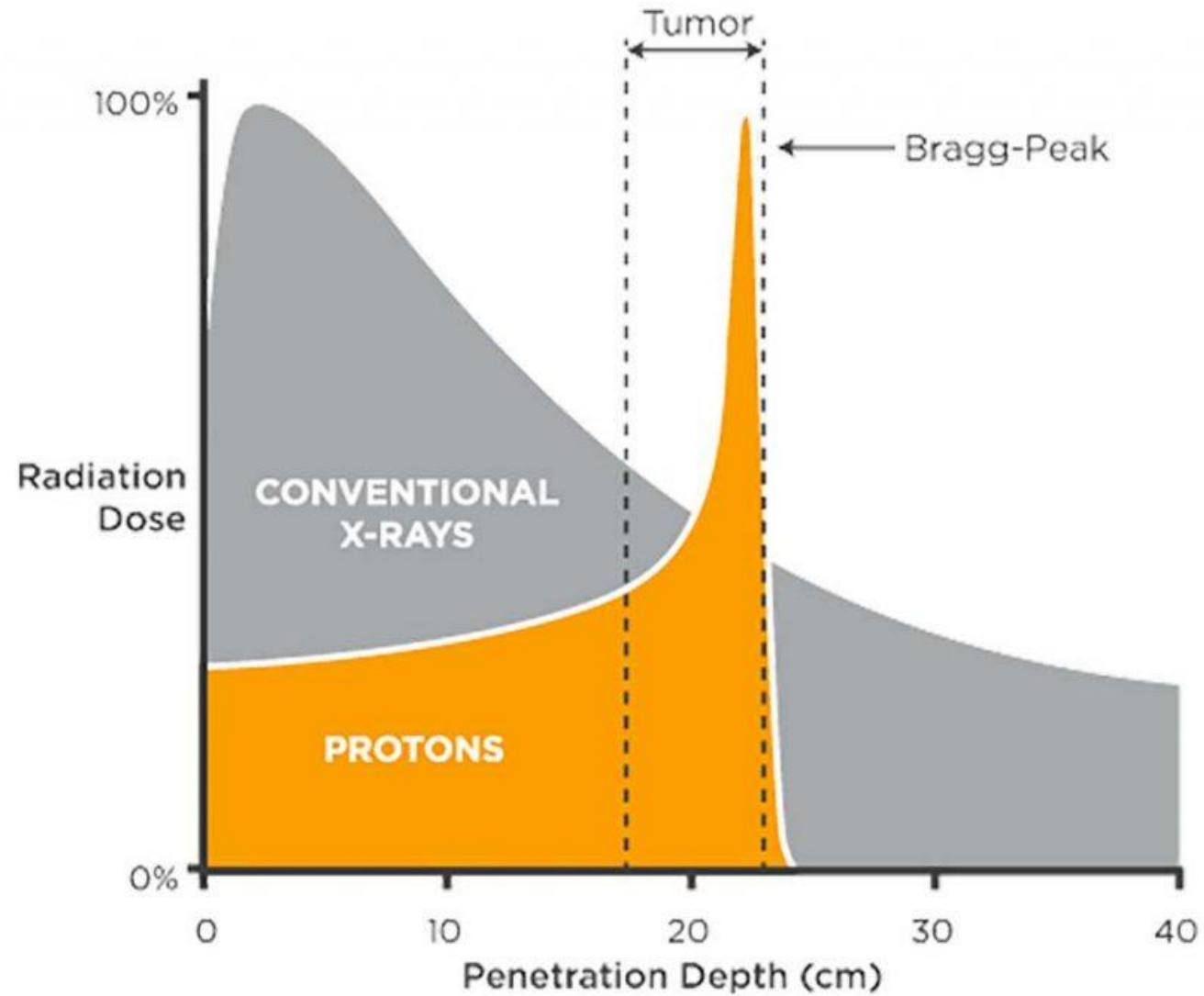


Wernimont et al. Nature Structural Biology 7, 766 - 771 (2000)

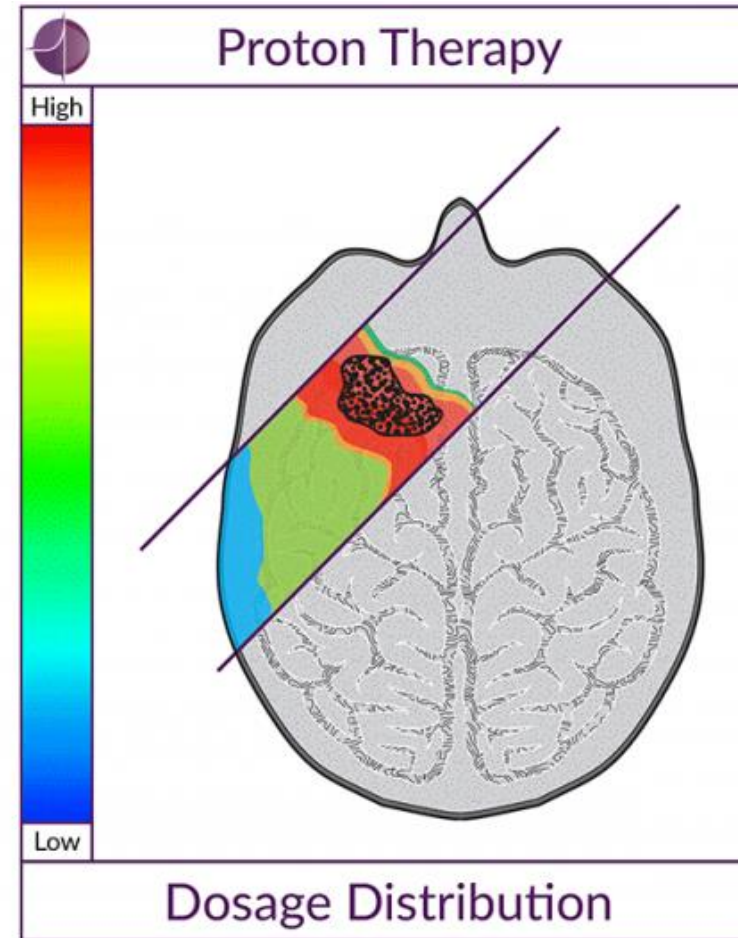
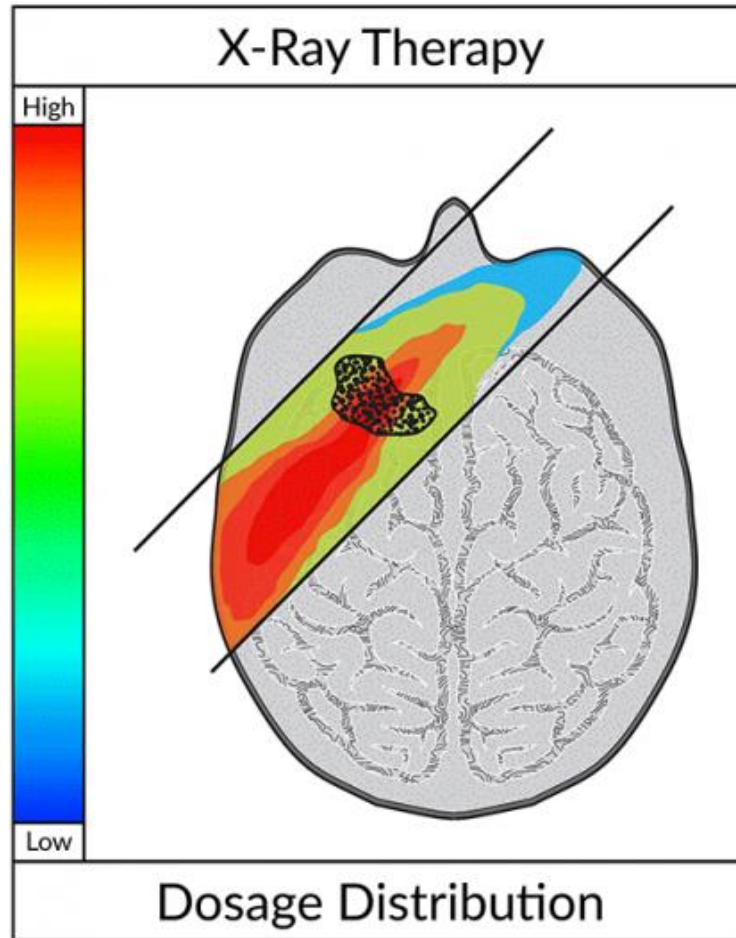


# Practical applications: Proton therapy range verification

# Bragg curve



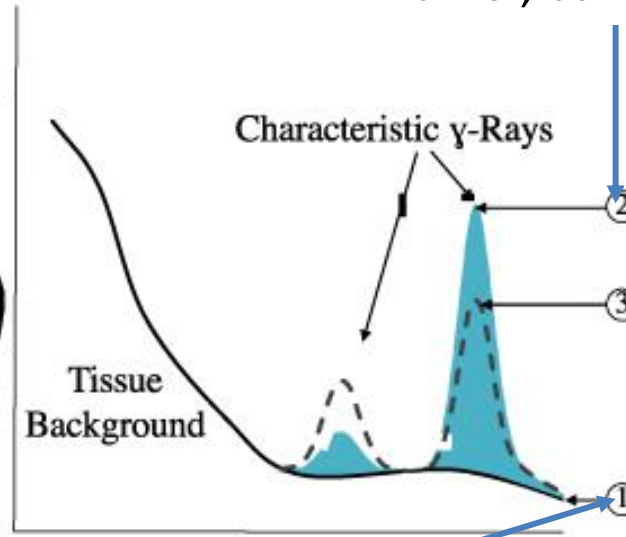
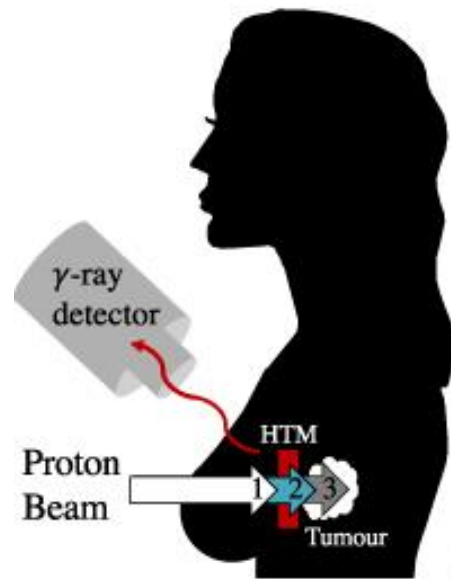
# Protontherapy



# 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  $\rightarrow$  Bragg peak next metal marker, correct energy

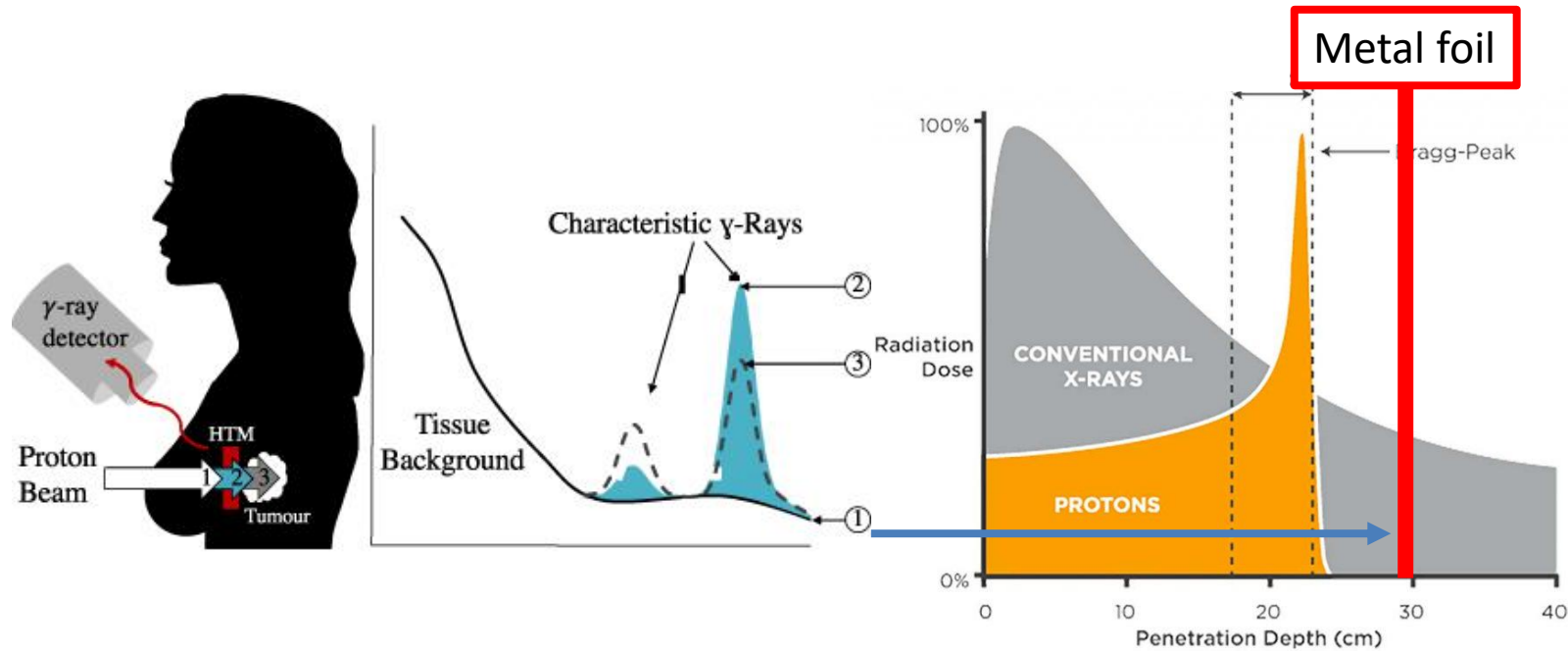


1- No peaks  $\rightarrow$  proton energy too low

3- Wrong peak ratio  $\rightarrow$  Bragg peak beyond metal marker, too high energy

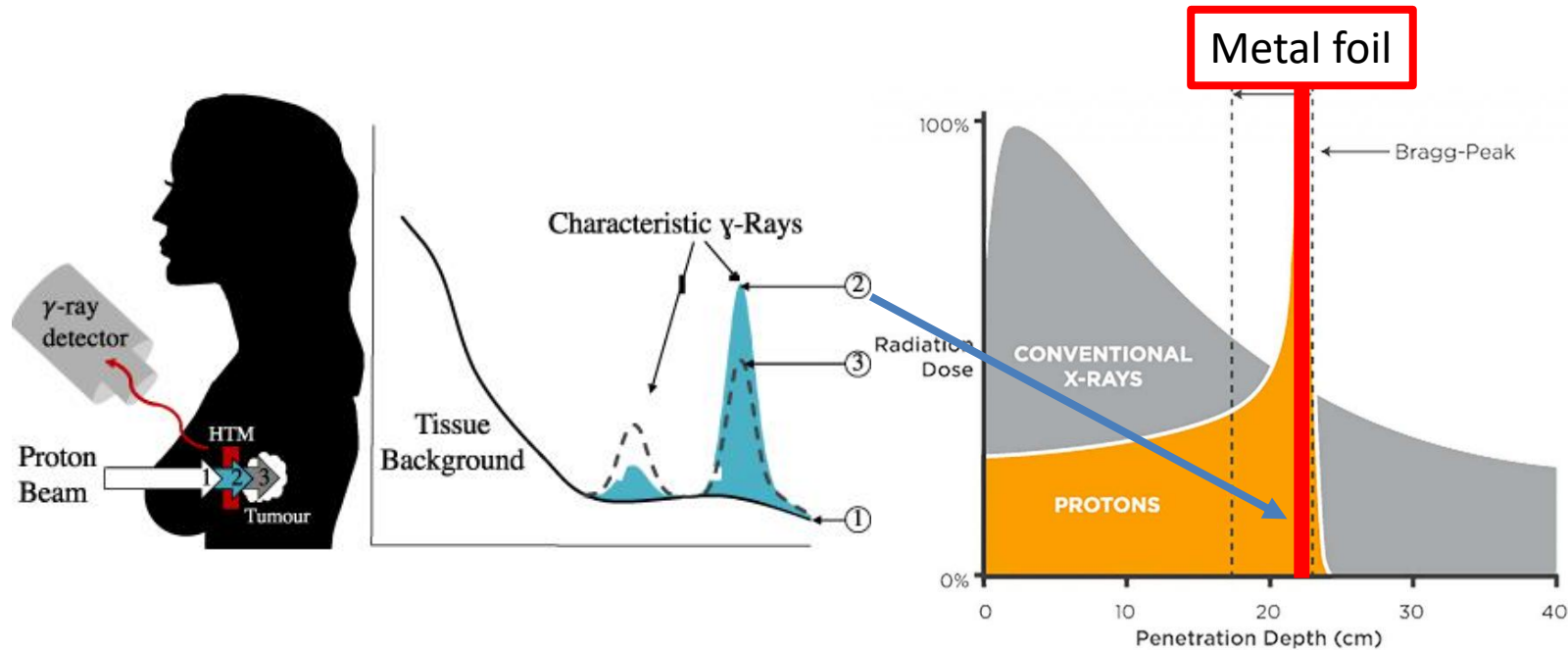


# Range verification



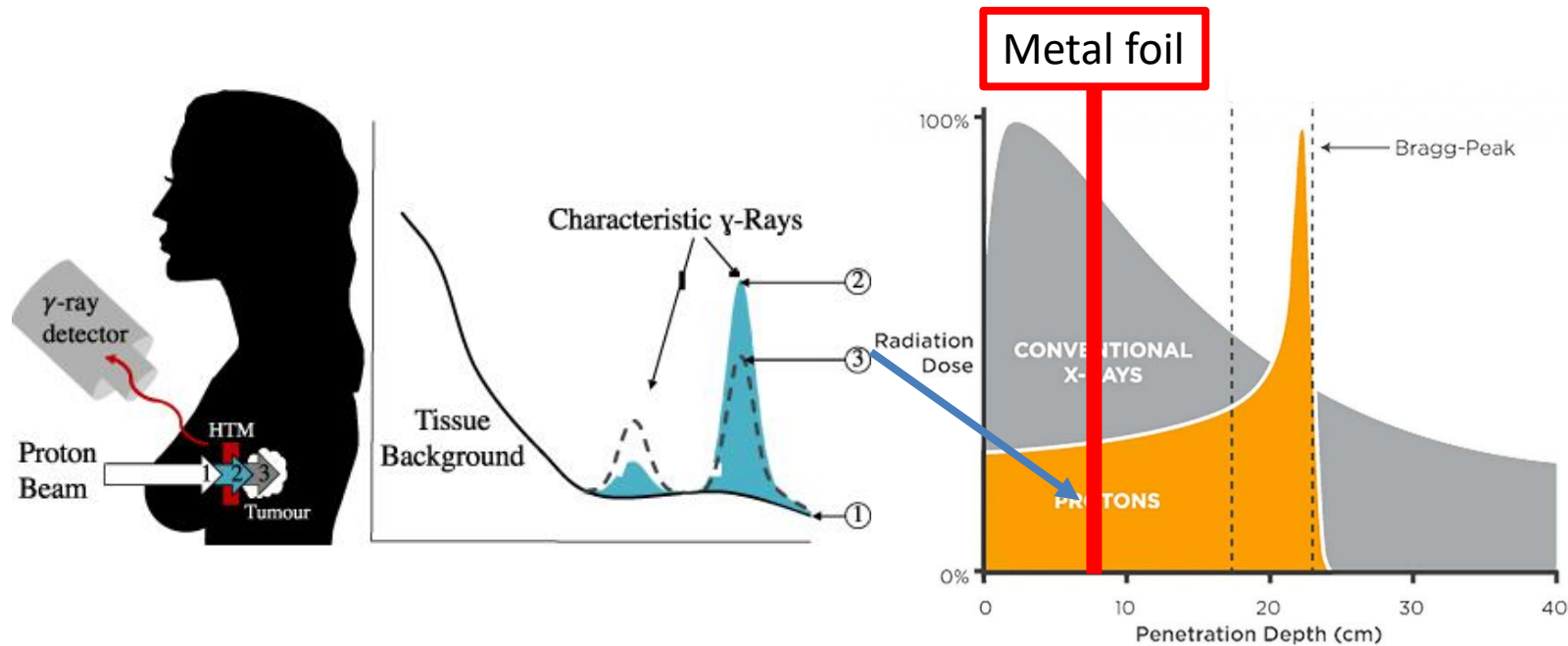
- Protons stop before metal foil
- No nuclear reaction
- Only tissue background

# Range verification



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

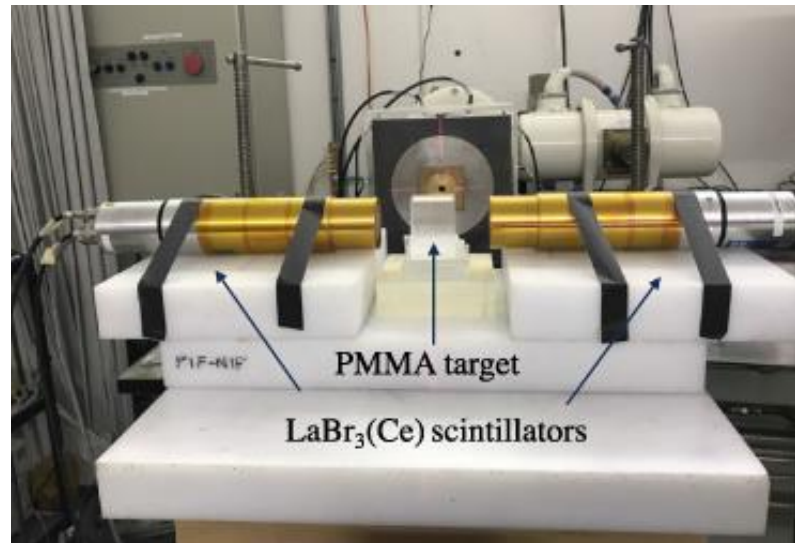
# Range verification



- 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  $\text{LaBr}_3$
- The technique allows for online range verification
- Sub-mm precision achieved



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

# Summary

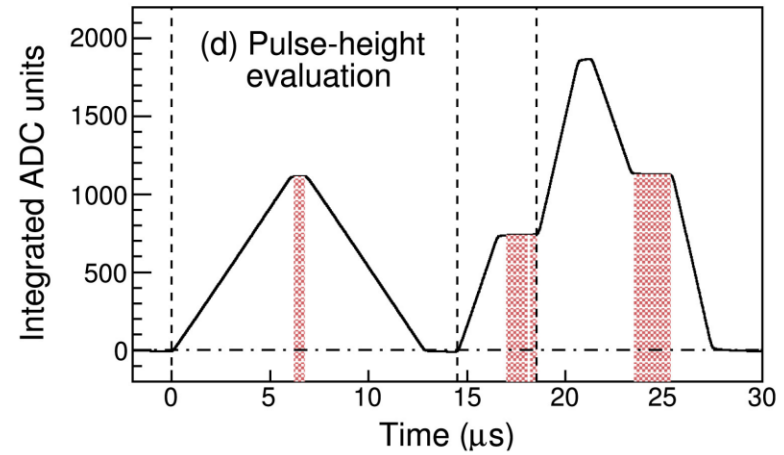
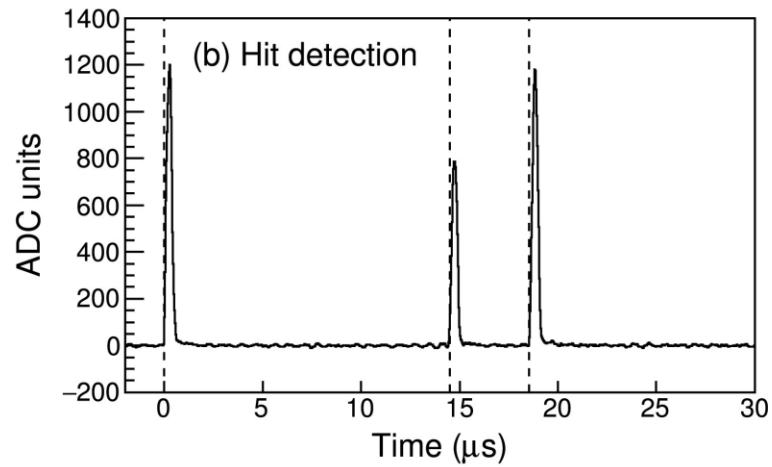
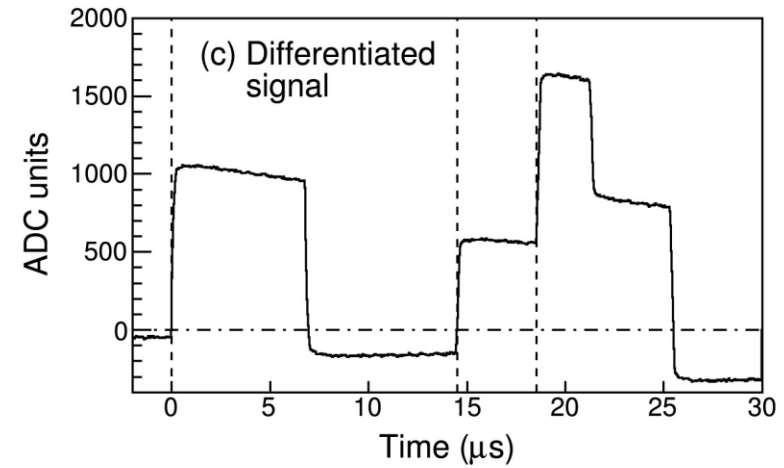
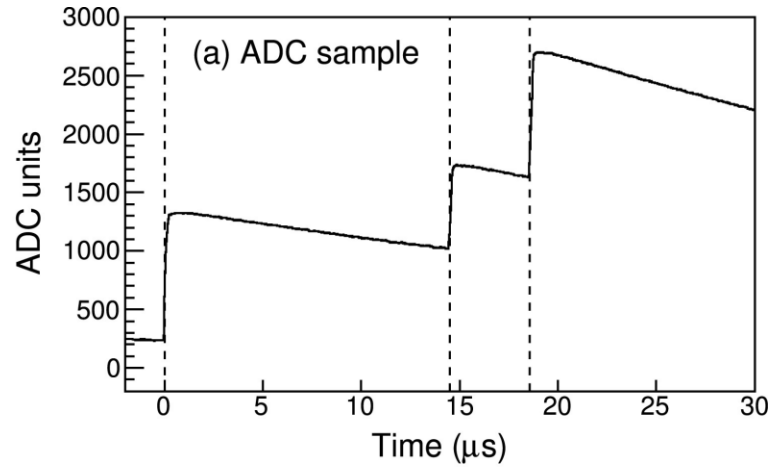
- Lifetime measurement is one of the most powerful probes we have to study nuclear structure
- Scintillators allow to measure timing down to  $\sim 10$  ps ( $10 \times 10^{-12}$  s)
- The fast-timing method has practical applications:
  - Protein structure and interaction
  - Medical imaging (PET-ToF)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



Any questions?

You can always contact me at  
[bruno.olaizola@csic.es](mailto:bruno.olaizola@csic.es)

# Pile up



# PAELLA at TRIUMF

## Perturbed Angular corrELations Labr Array





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