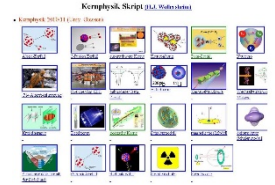


# Outline: Two-photon decay

Lecturer: Hans-Jürgen Wollersheim

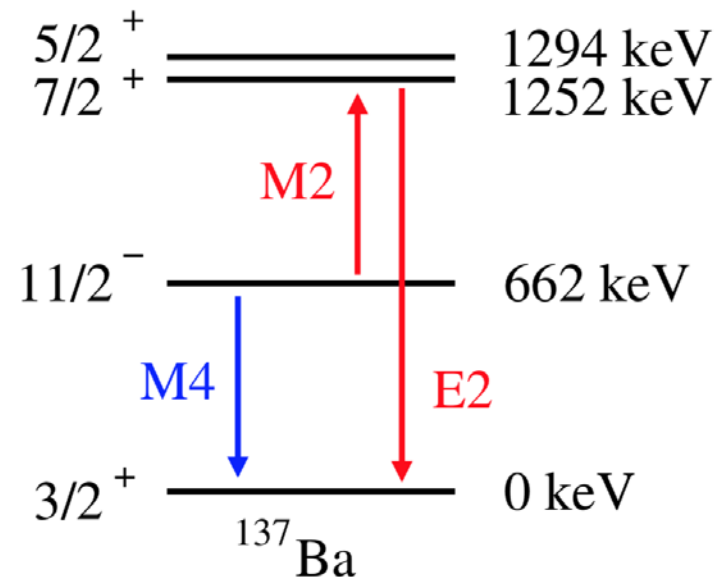
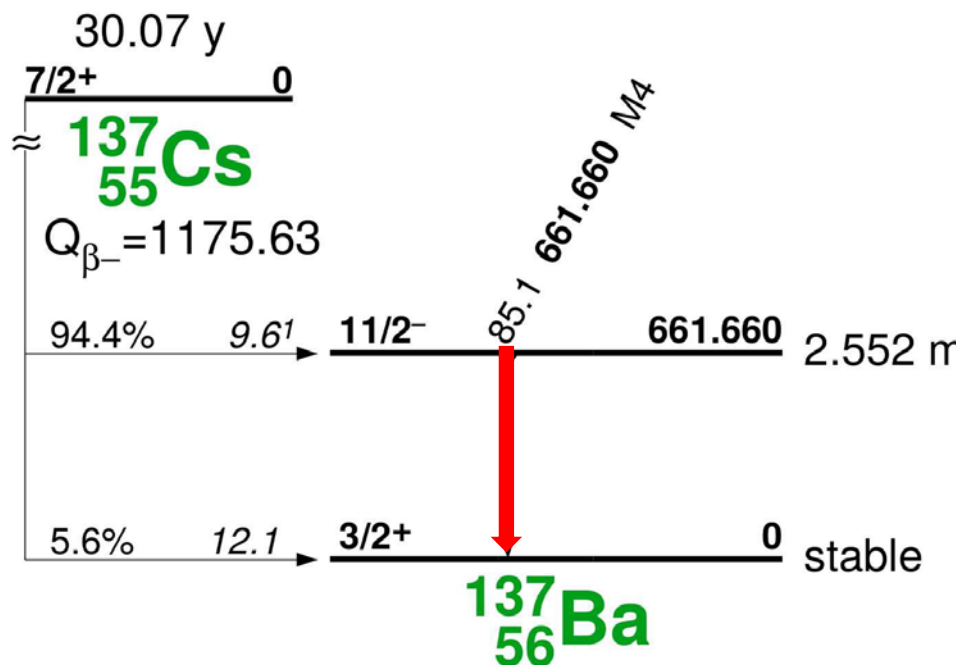
e-mail: [h.j.wollersheim@gsi.de](mailto:h.j.wollersheim@gsi.de)

web-page: <https://web-docs.gsi.de/~wolle/> and click on



1. 2-photon atomic transitions
2. double- $\gamma$  decay in nuclear physics
3. positronium decay

# Second order photon emission in nuclei

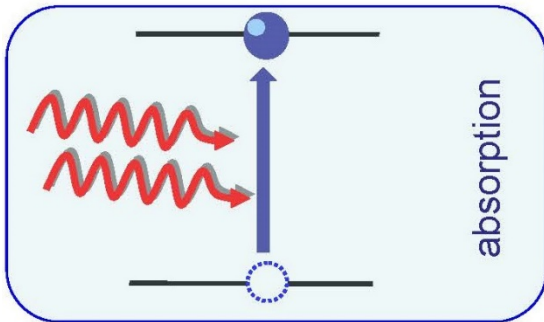
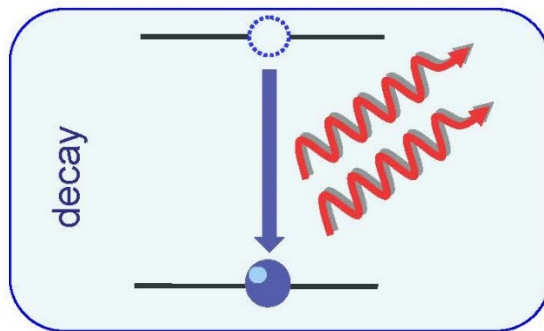


$$\tau(11/2) = [T(ME4; 11/2 \rightarrow 3/2) \cdot (1 + \alpha_T(M4))]^{-1}$$

$$T(M4; 11/2 \rightarrow 3/2) = 1.877 \cdot 10^{-6} \cdot E_\gamma^9 \cdot B(M4; 11/2 \rightarrow 3/2)$$

# Two-photon atomic transitions

- In 1929 in her PhD thesis Maria Göppert-Mayer suggested that bound-bound transitions can undergo under simultaneous absorption/emission of two correlated photons.



## *Über Elementarakte mit zwei Quantensprüngen*

*Von Maria Göppert-Mayer*

(Göttinger Dissertation)

(Mit 5 Figuren)

### Einleitung

Der erste Teil dieser Arbeit beschäftigt sich mit dem Zusammenwirken zweier Lichtquanten in einem Elementarakt. Mit Hilfe der Diracschen Dispersionstheorie<sup>1)</sup> wird die Wahrscheinlichkeit eines dem Ramaneffekt analogen Prozesses, nämlich der Simultanemission zweier Lichtquanten, berechnet. Es zeigt sich, daß eine Wahrscheinlichkeit dafür besteht, daß ein angeregtes Atom seine Anregungsenergie in zwei Licht-

- For many years two-photon decay/excitation of atoms and ions has attracted much of experimental and theoretical interest.

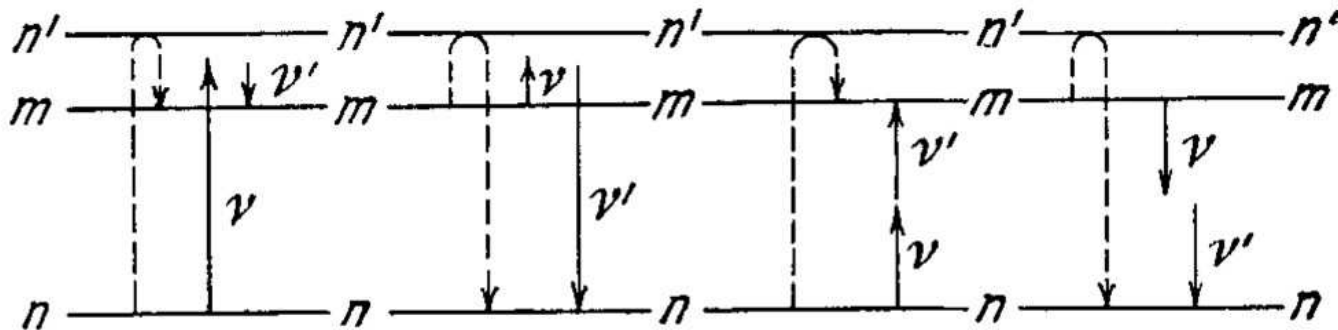
# Two-photon atomic transitions

- In 1929 in her PhD thesis Maria Göppert-Mayer suggested that bound-bound transitions can undergo under simultaneous absorption/emission of two correlated photons.
- Second order process ( $10^{-6}$  weaker)

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



- not only two-photon **emission**, but also **absorption** and Raman scattering



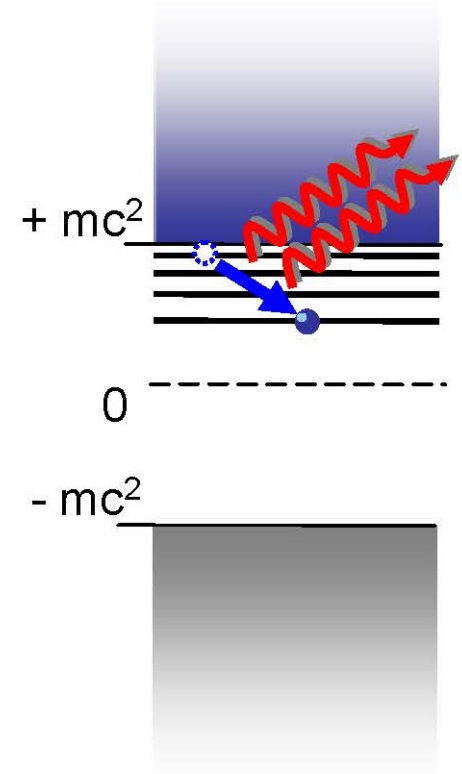
# Two-photon atomic transitions

- Analysis of the two-photon decay requires knowledge about complete spectrum of the

$$\tilde{M}_{fi} \propto \sum_{\nu} \frac{\langle \psi_f | \boldsymbol{\alpha} \boldsymbol{\varepsilon}_2 e^{-i\mathbf{k}r_2} | \psi_{\nu} \rangle \langle \psi_{\nu} | \boldsymbol{\alpha} \boldsymbol{\varepsilon}_1 e^{-i\mathbf{k}r_1} | \psi_i \rangle}{E_{\nu} - E_i + \hbar \omega_1}$$

- The summation in the second-order transition amplitude includes a summation over the discrete part of the spectrum as well as an integration over the positive and negative-energy continuum.

- A large number of studies have been performed over the last decades to investigate
  - Total decay rates
  - Energy (spectral) distributions



PRL 104, 033001 (2010)

PHYSICAL REVIEW LETTERS

week ending  
22 JANUARY 2010

## Spectral Shape of the Two-Photon Decay of the $2^1S_0$ State in He-Like Tin

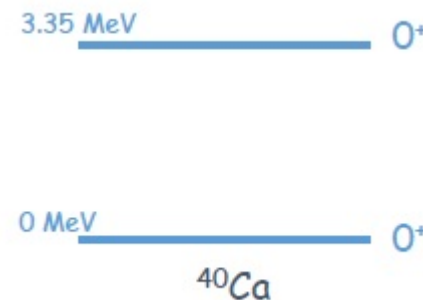
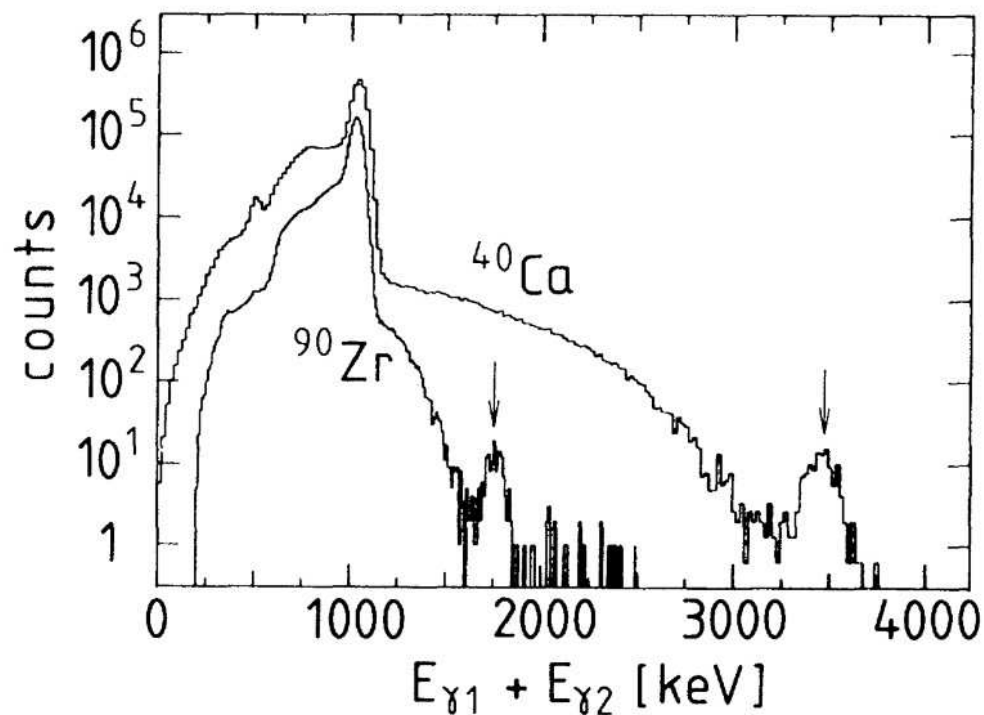
S. Trotsenko,<sup>1,2,3</sup> A. Kumar,<sup>2</sup> A. V. Volotka,<sup>4</sup> D. Banaś,<sup>5</sup> H. F. Beyer,<sup>2</sup> H. Bräuning,<sup>2</sup> S. Fritzsche,<sup>2</sup> A. Gumberidze,<sup>6</sup> S. Hagmann,<sup>1,2</sup> S. Hess,<sup>1,2</sup> P. Jagodziński,<sup>5</sup> C. Kozhuharov,<sup>2</sup> R. Reuschl,<sup>7</sup> S. Salem,<sup>1,2</sup> A. Simon,<sup>8</sup> U. Spillmann,<sup>2</sup> M. Trassinelli,<sup>7</sup> L. C. Tribedi,<sup>9</sup> G. Weber,<sup>2,10,\*</sup> D. Winters,<sup>2</sup> and Th. Stöhlker<sup>2,3,10,\*</sup>

# Double-gamma decay in nuclear physics

- first unambiguous observation
- $\gamma\gamma$ -decay only known in a special case:  
 $0^+ \rightarrow 0^+$  ( $^{90}\text{Zr}$ ,  $^{40}\text{Ca}$ ,  $^{16}\text{O}$ )

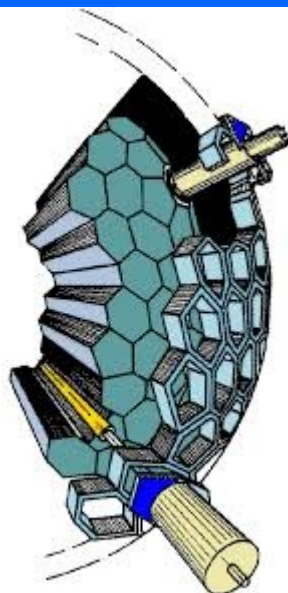
Heidelberg – Darmstadt Crystal ball ( $4\pi$ , 162 NaI)

single photon decay strictly forbidden

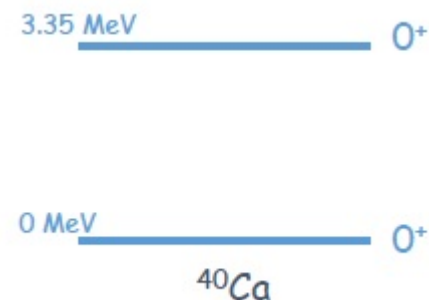


Sum-energy spectrum in coincidence with protons populating the states of interest

# Double-gamma decay in nuclear physics



Heidelberg – Darmstadt Crystal ball ( $4\pi$ , 162 NaI)  
angular resolution  $\pm 9^\circ$

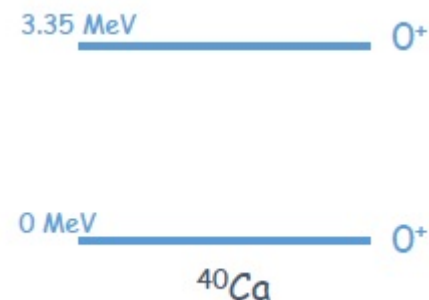
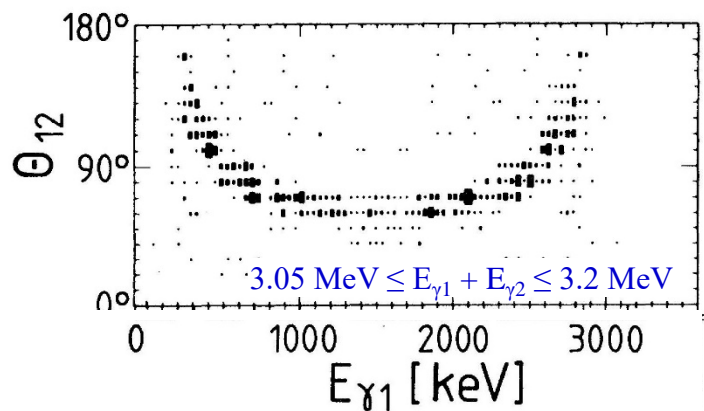


$\gamma$ -background: internal pair conversion and subsequent positron annihilation in flight

$$\frac{1}{E_{\gamma 1}} + \frac{1}{E_{\gamma 2}} = \frac{(1 - \cos\theta_{12})}{m_e c^2}$$

$$E_{\gamma 1} + E_{\gamma 2} = E_{e^+} + 2m_e c^2 \cong E_0$$

# Double-gamma decay in nuclear physics



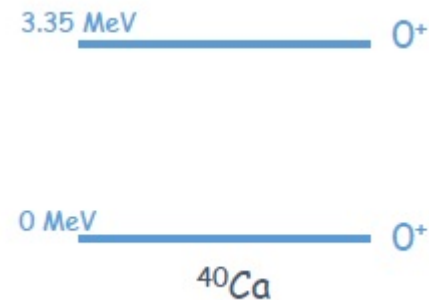
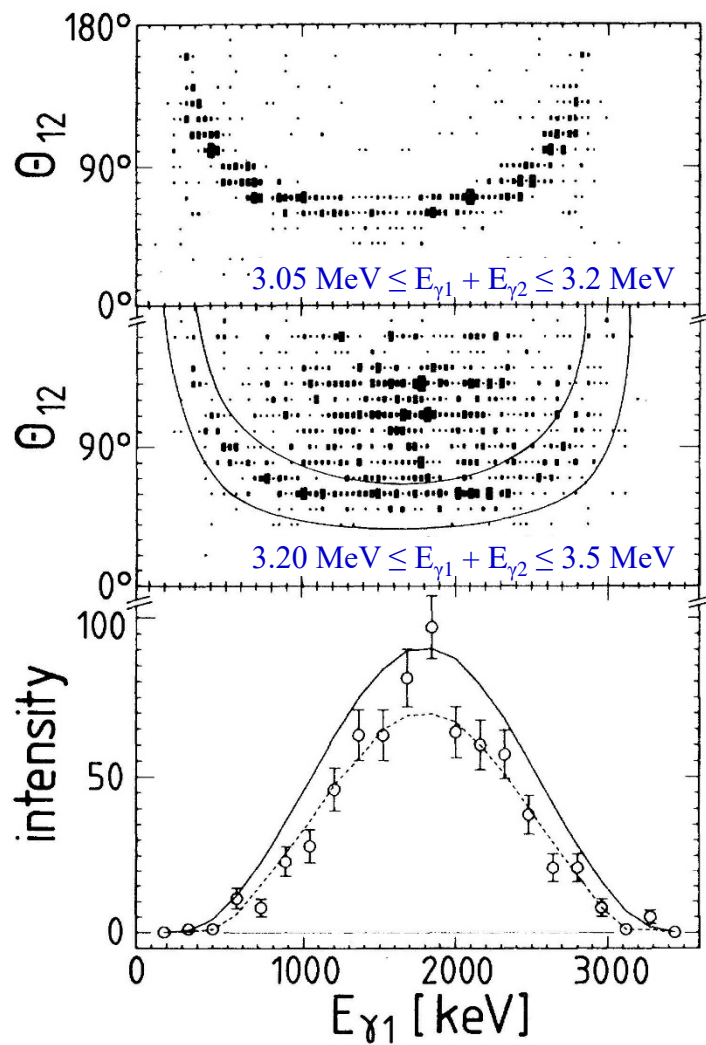
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$$\frac{1}{E_{\gamma 1}} + \frac{1}{E_{\gamma 2}} = \frac{(1 - \cos\theta_{12})}{m_e c^2}$$

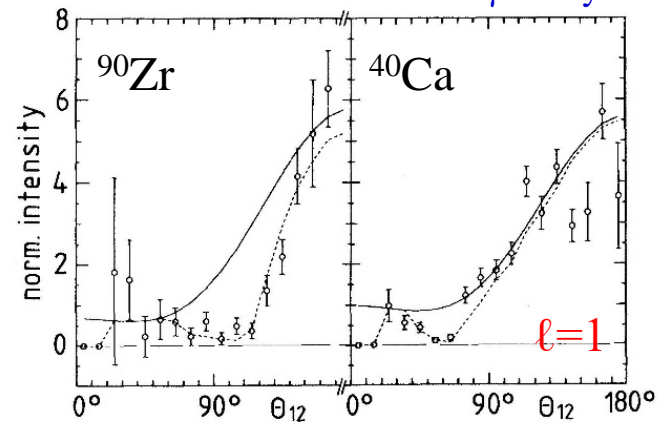
$$E_{\gamma 1} + E_{\gamma 2} = E_{e^+} + 2m_e c^2 \cong E_0$$



# Double-gamma decay in nuclear physics

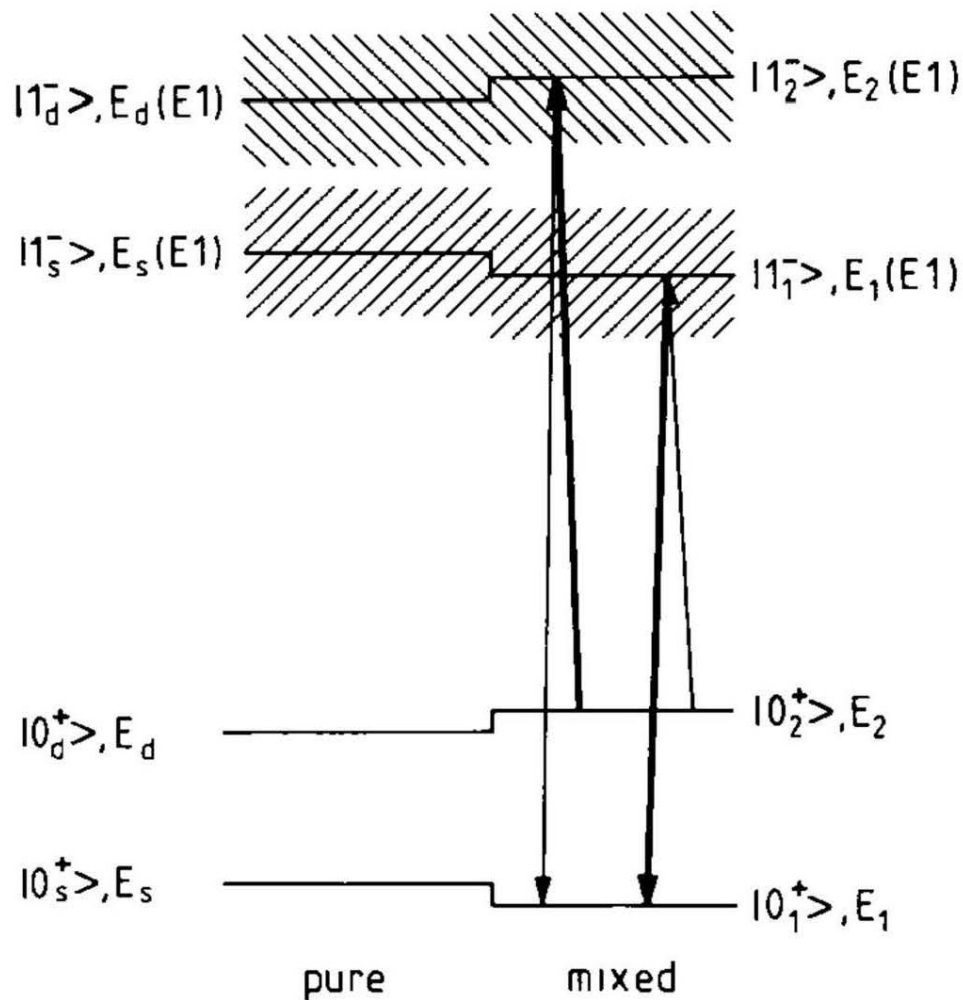


directional correlation of  $2\gamma$ -decay



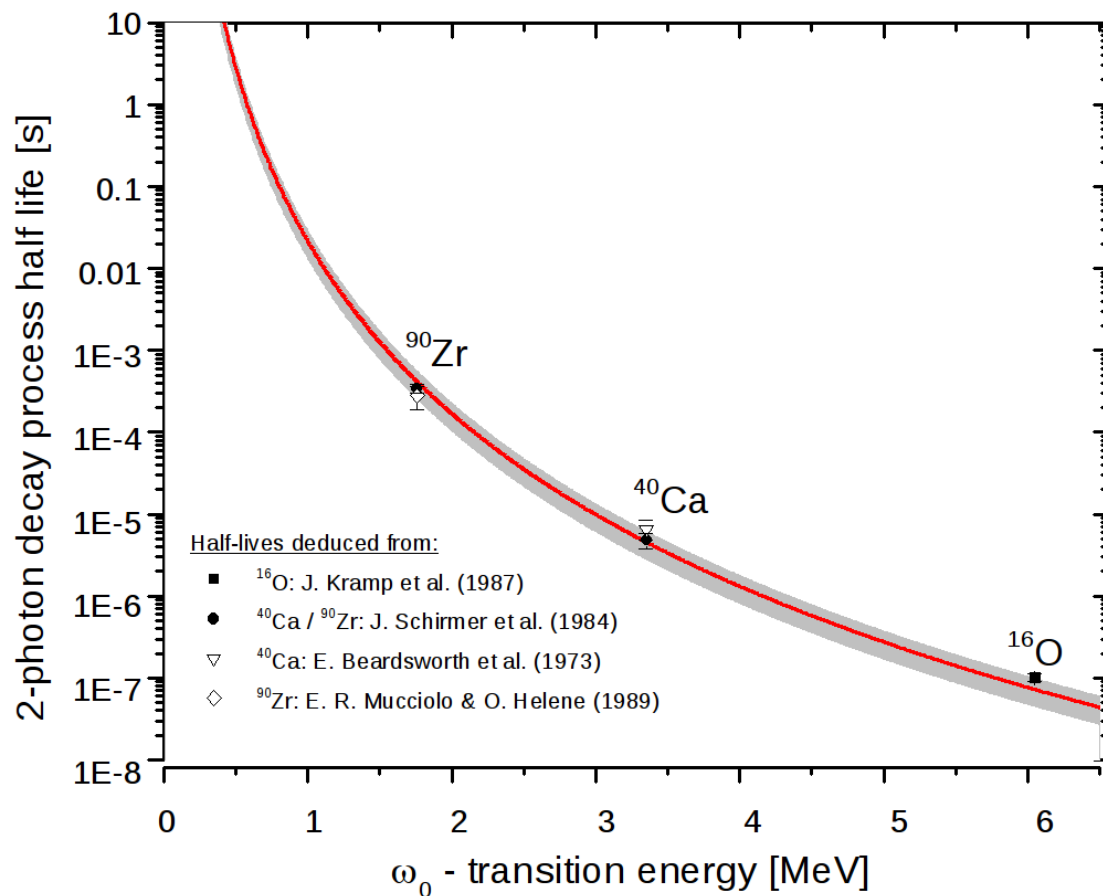
angular correlation not symmetric around  $90^\circ$

# Double-gamma decay in nuclear physics



2-state mixing model for  $0^+$  state and  $1^-$  GDR states  
 states couple only via GDR states

# Double-gamma decay in nuclear physics



$2\gamma$ nucleus	$\Gamma_{\gamma\gamma}/\Gamma_{total}$
$^{16}\text{O}$	$6.6(5) \cdot 10^{-4}$
$^{40}\text{Ca}$	$4.5(10) \cdot 10^{-4}$
$^{90}\text{Zr}$	$1.8(1) \cdot 10^{-4}$

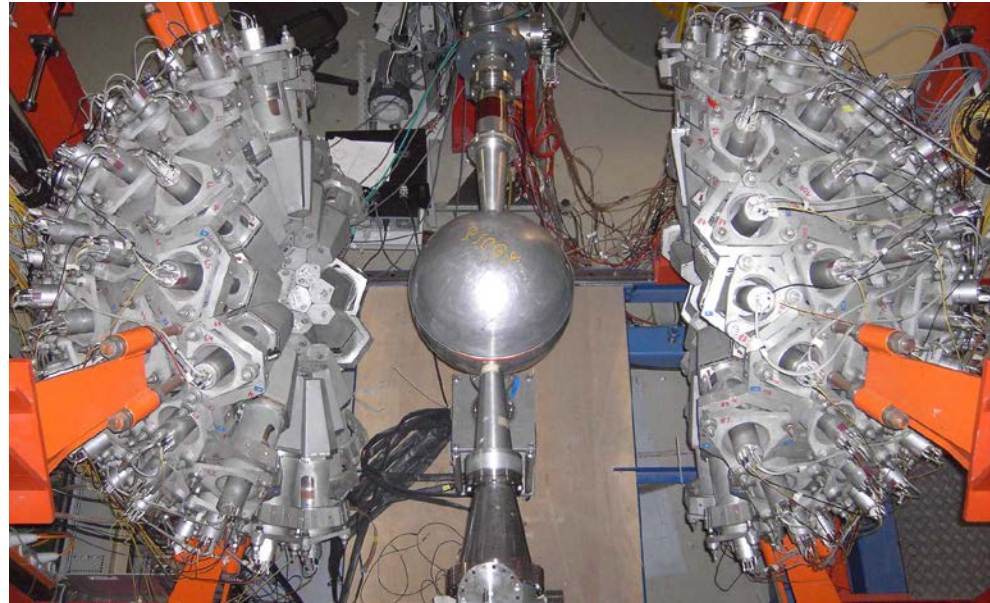
$0^+ \rightarrow 0^+$  ( $^{68}\text{Ni}$ ,  $^{72}\text{Ge}$ ,  $^{80}\text{Ge}$ ,  $^{96}\text{Zr}$ ,  $^{98}\text{Zr}$ ,  $^{98}\text{Mo}$ )

# Recent experimental advance: $\text{LaBr}_3(\text{Ce})$ detectors

- so far:  $\text{NaI}(\text{Tl})$  detectors
  - standard detectors, if **high efficiency** is crucial
  - but: poor **time** and **energy resolution**

Heidelberg-Darmstadt Crystal-ball

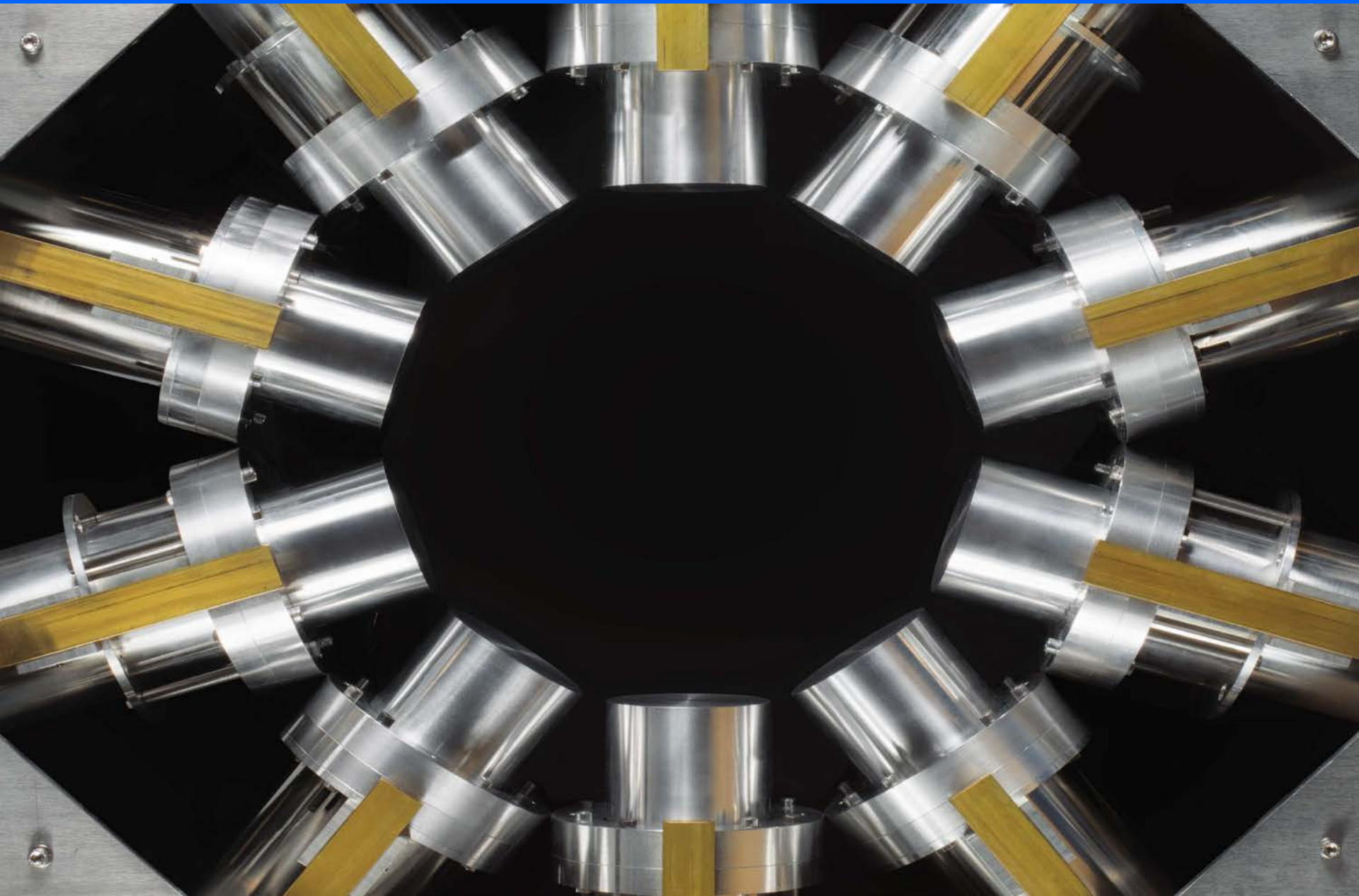
full solid angle  $4\pi$     162  $\text{NaI}(\text{Tl})$  detectors



- **large volume  $\text{LaBr}_3(\text{Ce})$  detectors** available:
  - better energy resolution by a factor 2-3
  - better time resolution by a factor 5-10
  - very fast  $\rightarrow$  high rate measurement

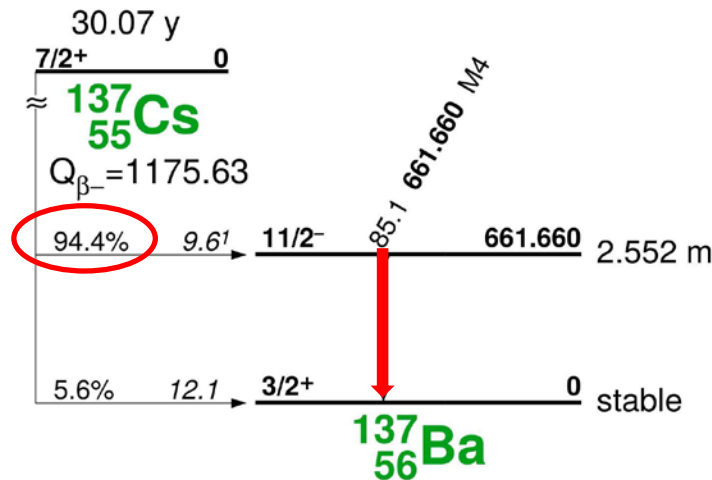


# GALATEA array: 18 $\text{LaBr}_3(\text{Ce})$ detectors (3"x3")



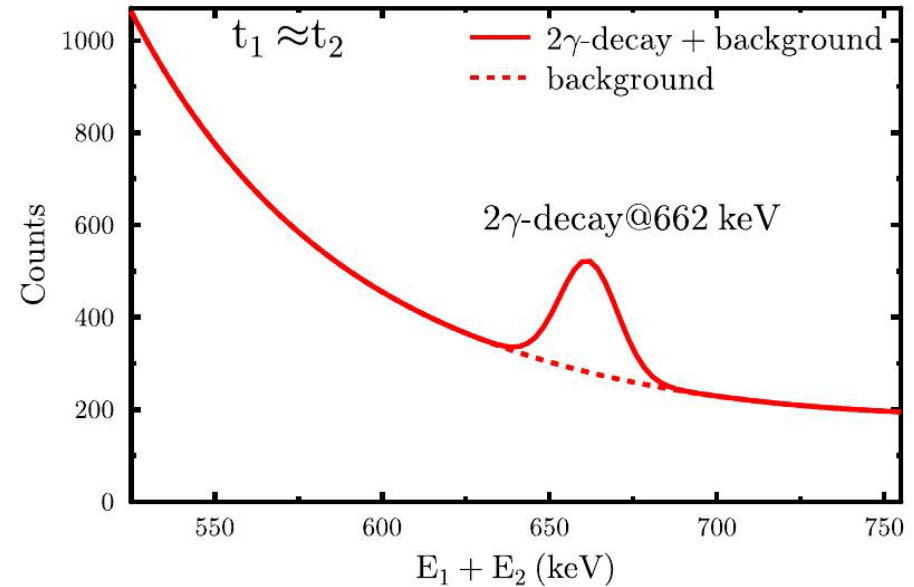
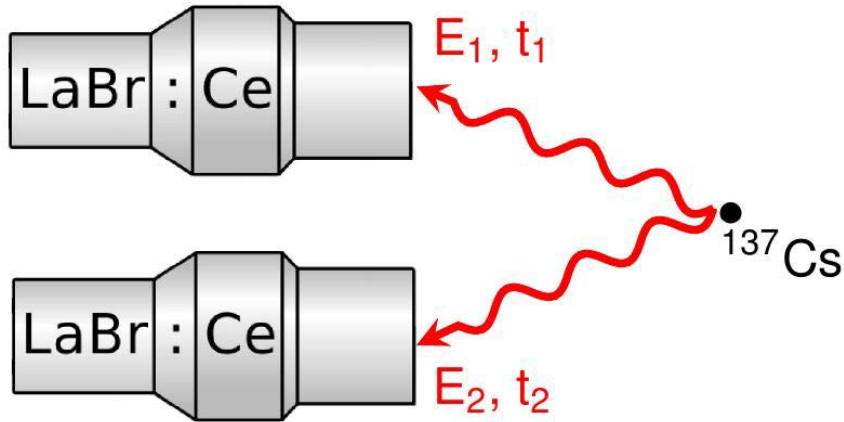
# Competitive double-photon decay: $\gamma\gamma/\gamma$

- for  $0^+ \rightarrow 0^+$  transitions:
  - **single** photon decay **strictly forbidden**
  - $\Gamma_{\gamma\gamma}/\Gamma \sim 10^{-4}$
  - $\Gamma/\Gamma_{IP}$  (internal pair production)
- **Competitive** double-gamma decay ( $\gamma\gamma/\gamma$ )
  - $\gamma\gamma$  decay **competing** with **allowed** single gamma decay
  - $\Gamma \approx \Gamma_\gamma$
  - $\Gamma_{\gamma\gamma}/\Gamma_\gamma \ll 10^{-4}$
  - has never been observed, despite a few searches in last 30 years



# Basic principle of the experiment

- use radioactive  $^{137}\text{Cs}$ -source: 16.3(5)  $\mu\text{Ci}$

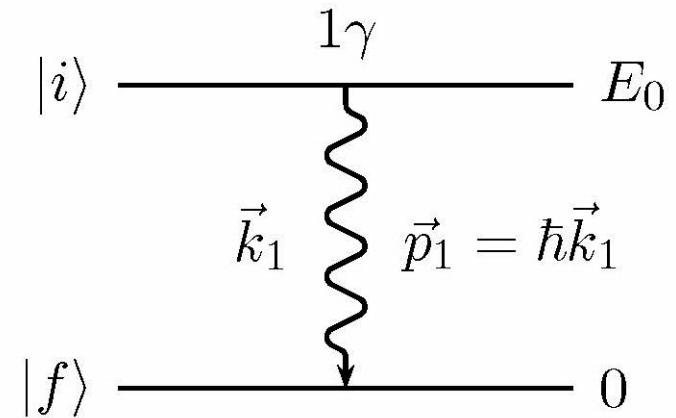


- background  $\leftrightarrow$  small decay probability ( $\sim 1$  event per day)
  - direct Compton scattering
  - random coincidences
  - cosmic rays, sequential Compton scattering, internal radioactivity

# Emission of electromagnetic one and two photon(s)

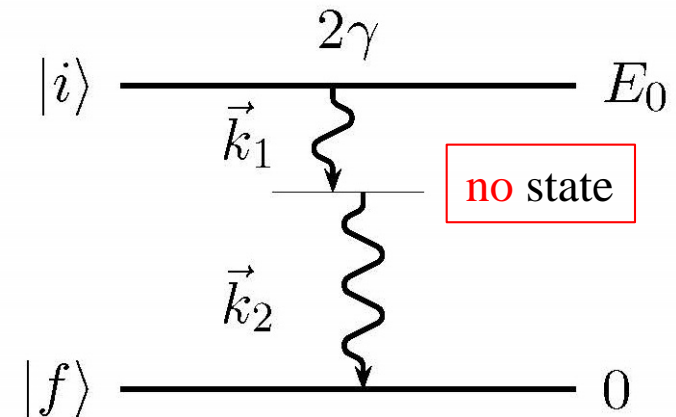
- **single** photon emission

$$E_0 = E_1 = \hbar\omega_1$$



- **double-gamma** decay  
two photons emitted  
**simultaneously**

$$E_0 = E_1 + E_2$$





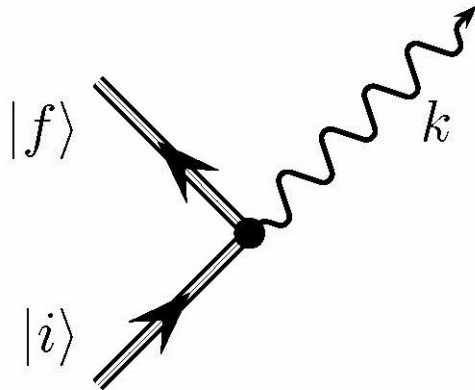
# Decay width: first order perturbation theory

- Interaction of a nucleus with free EM radiation field:

$$H_{int} = -\frac{1}{c} \int \vec{j}_N(\vec{r}, t) \vec{A}(\vec{r}, t) d^3 r$$

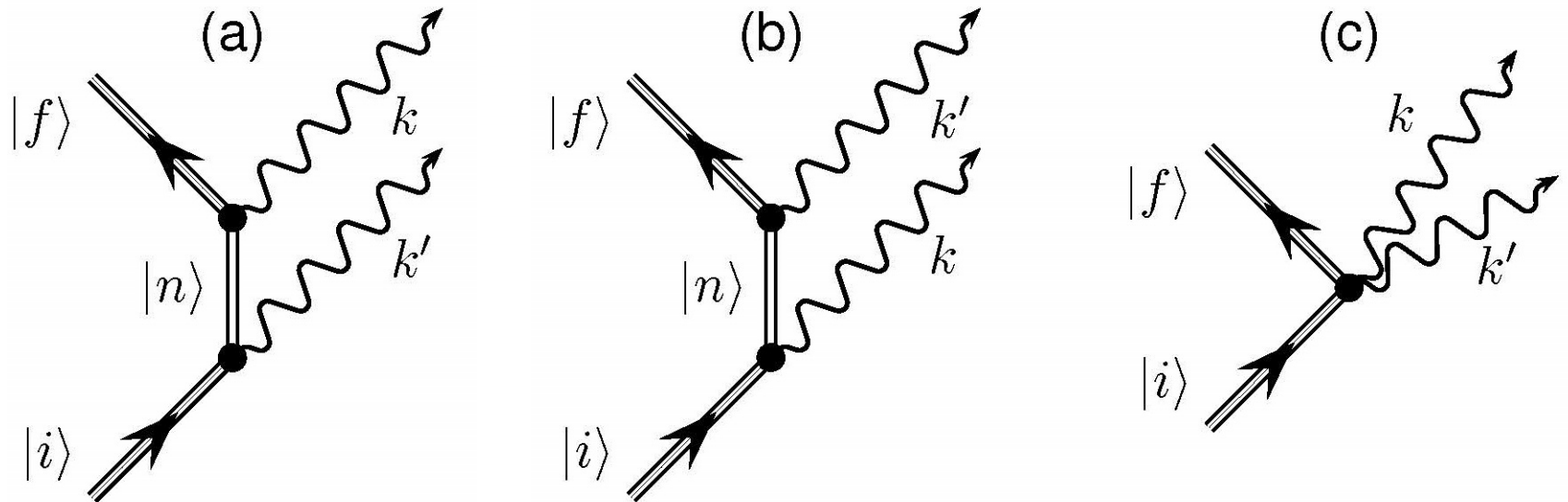
- **Fermi's Golden Rule**

$$\Gamma_\gamma = 2\pi |\langle f | H_{int} | i \rangle|^2 \rho_f$$



$\rho_f$ : density of final states;  $H_{int}$ : interaction Hamiltonian;  $\vec{j}_N(\vec{r}, t)$ : nuclear current density;  $\vec{A}(\vec{r}, t)$ : EM vector potential

## Second order



- a,b) **resonance** amplitudes (second order in  $\vec{j} \cdot \vec{A}$  interaction)
  - sum over **all** intermediate states  $|n\rangle$
  - usual selection rules apply at each vertex
- c) **Seagull** amplitude (introduced as a correction to the non relativistic treatment of the nuclear Hamiltonian): first order, but quadratic in the radiation field  $A^2$
- theory is fully developed

J. Kramp, ..., **D. Schwalm** et al., NPA 474, 412 (1987)

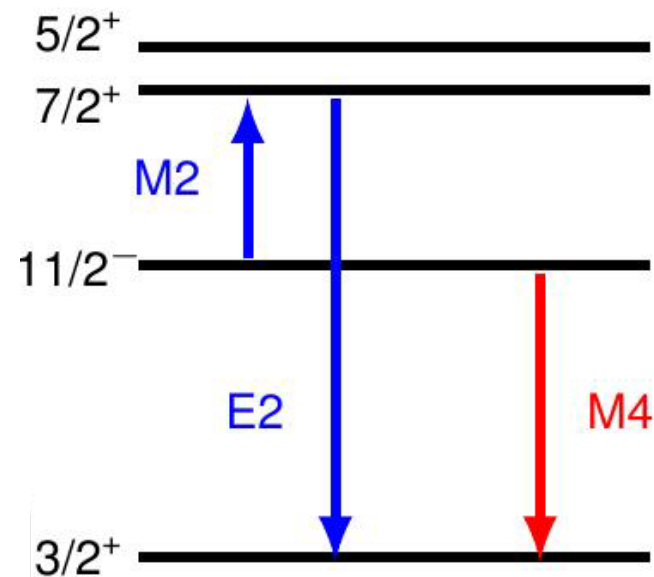
# Matrix element of the double-gamma decay

$$\frac{d\Gamma_{\gamma\gamma}^2}{d\omega d\cos\theta}(\alpha_{00'}, \dots)$$

$$\alpha_{00'} = \sum_n \frac{\langle f || \mathbf{O} || n \rangle \cdot \langle n || \mathbf{O}' || i \rangle}{E_n}$$

$$\alpha_{M2E2} = \frac{\langle 3/2^+ || \mathbf{E2} || 7/2^+ \rangle \cdot \langle 7/2^+ || \mathbf{M2} || 11/2^- \rangle}{E_{7/2^+}} + \dots$$

$^{137}\text{Ba}$

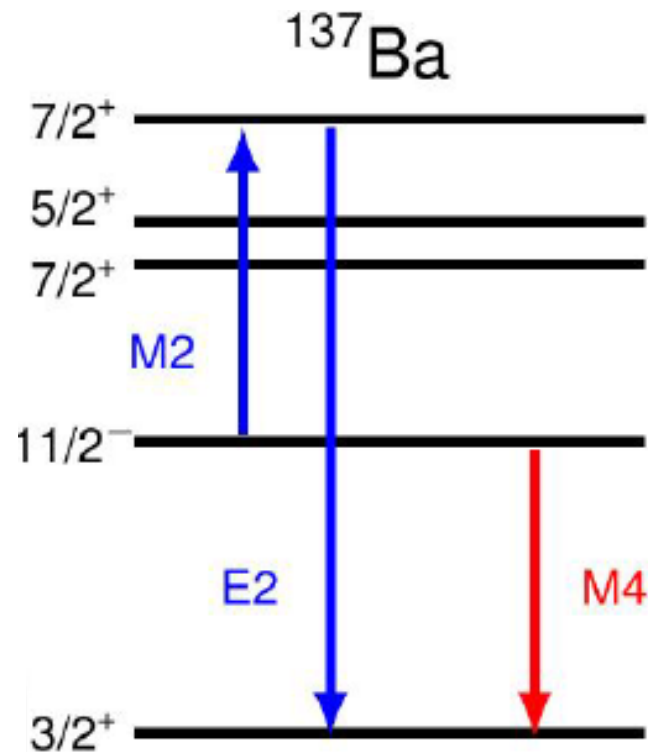


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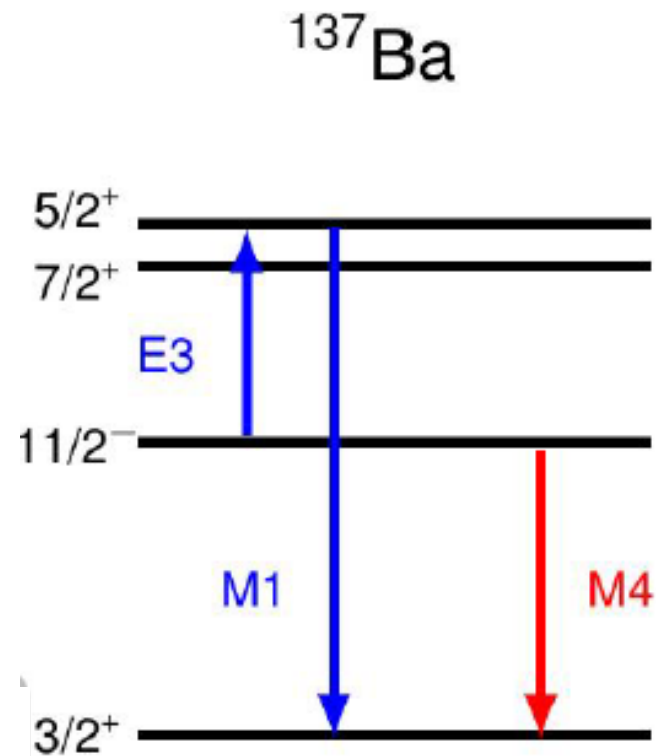


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$$\alpha_{E3M1} = \frac{\langle 3/2^+ || \mathbf{M1} || 5/2^+ \rangle \cdot \langle 5/2^+ || \mathbf{E3} || 11/2^- \rangle}{E_{5/2^+}} + \dots$$



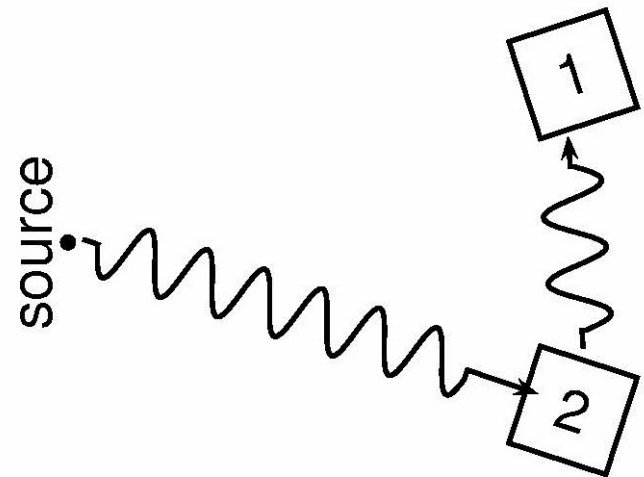
# Experimental Obstacle

- very small branching ratio  $\Gamma_{\gamma\gamma}/\Gamma_{\gamma} \ll 10^{-4}$
- **Compton scattering**  
energy of **single**  $\gamma$ -ray deposited in two detectors
  - exact same signature for **energy sum**

$$E_0 = E_1 + E_2$$

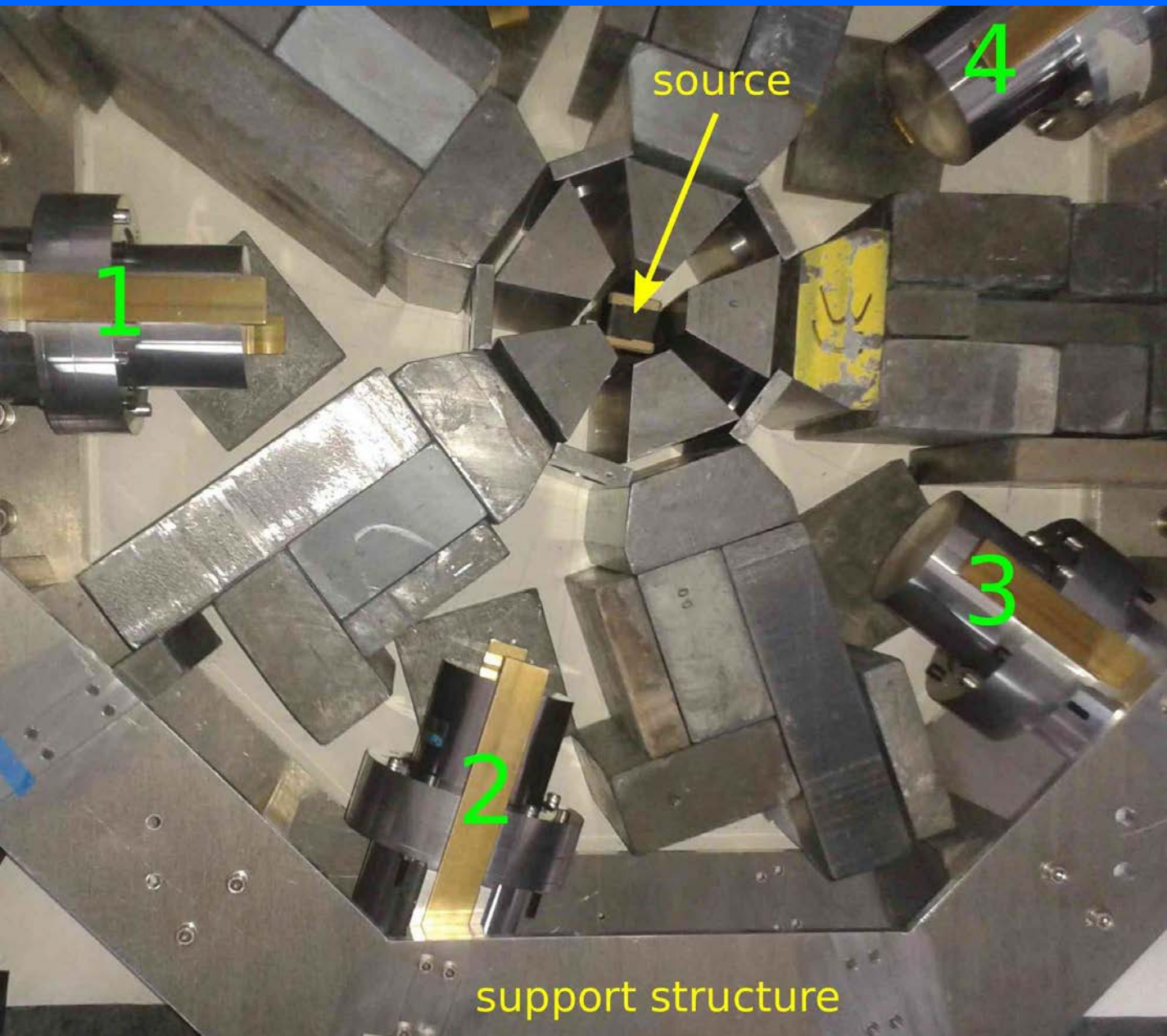
but

- different energy distribution
  - different path of photons: shielding
- 
- almost same **timing** ( $\Delta t \sim 1ns$ )  
but:
    - $\Delta t \neq 0$
- 
- *no problem for  $0^+ \rightarrow 0^+$*



$$E'_{\gamma} = \frac{E_{\gamma}}{1 + \frac{E_{\gamma}}{m_e c^2} (1 - \cos\theta)}$$

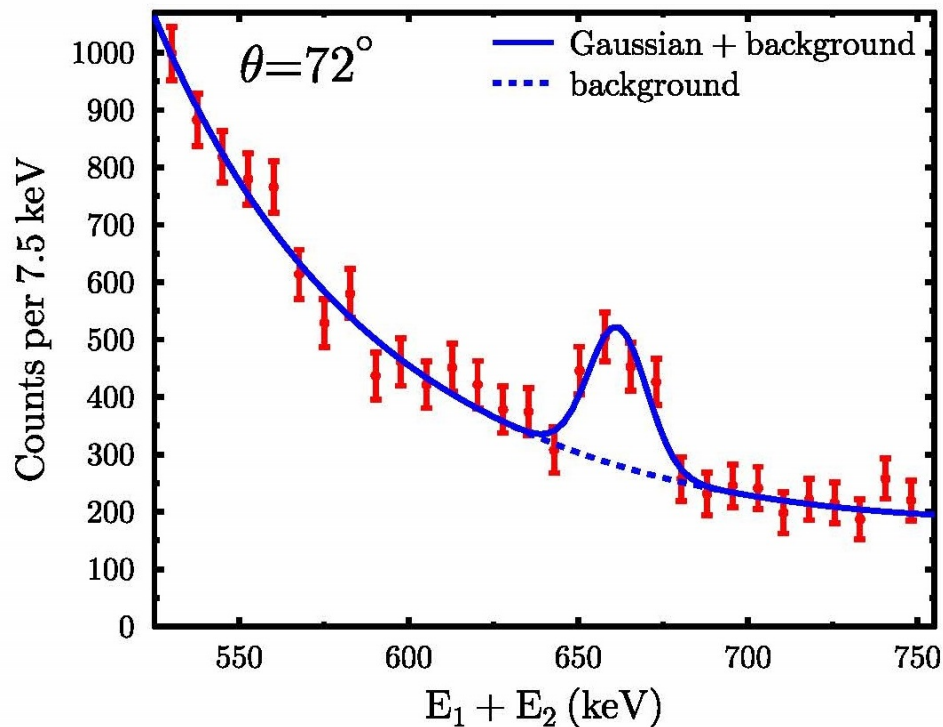
# Experimental setup



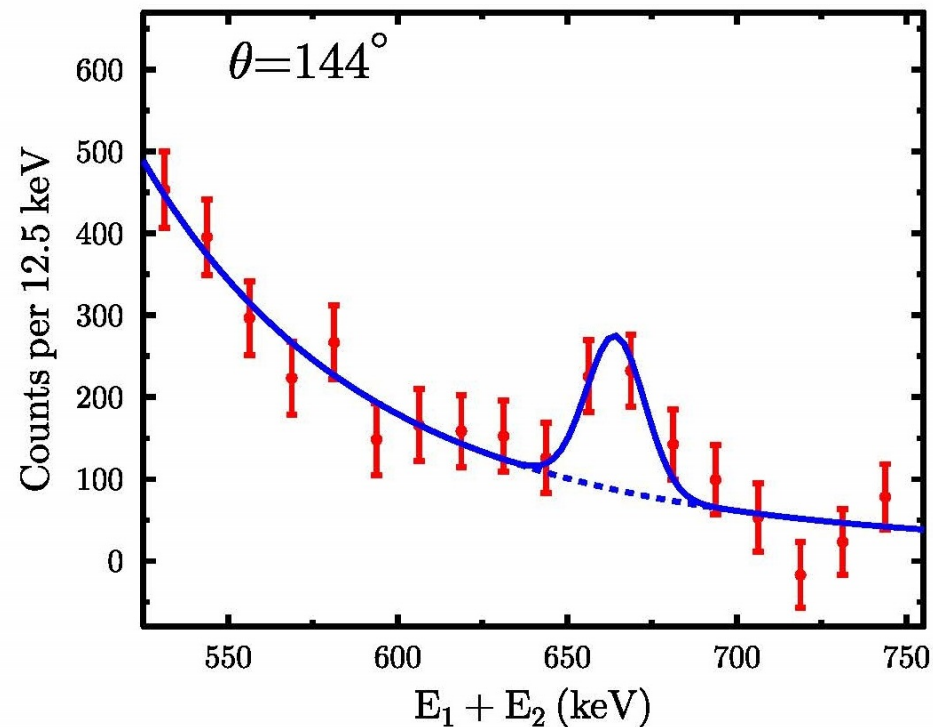
- $72^\circ$ : 5 detector pairs
- $144^\circ$ : 5 detector pairs
- thick **Pb blocks** between detectors
- $\varepsilon_{\text{FE}}(662 \text{ keV}) = 1.5 \%$
- $\varepsilon_{\gamma\gamma} \approx 4 \cdot 10^{-4}$
- $\Delta E = 3\%$  (FWHM)
- $\Delta t = 1 \text{ ns}$  (FWHM)
- on disk: **53 days**
- source:  $^{137}\text{Cs}$  (600 kBq)



# Results – random subtracted energy spectra



693(95) counts ( $\sigma = 7.3$ )  
 $\Gamma_{\gamma\gamma}/\Gamma_\gamma = 1.56(23) \cdot 10^{-6}$

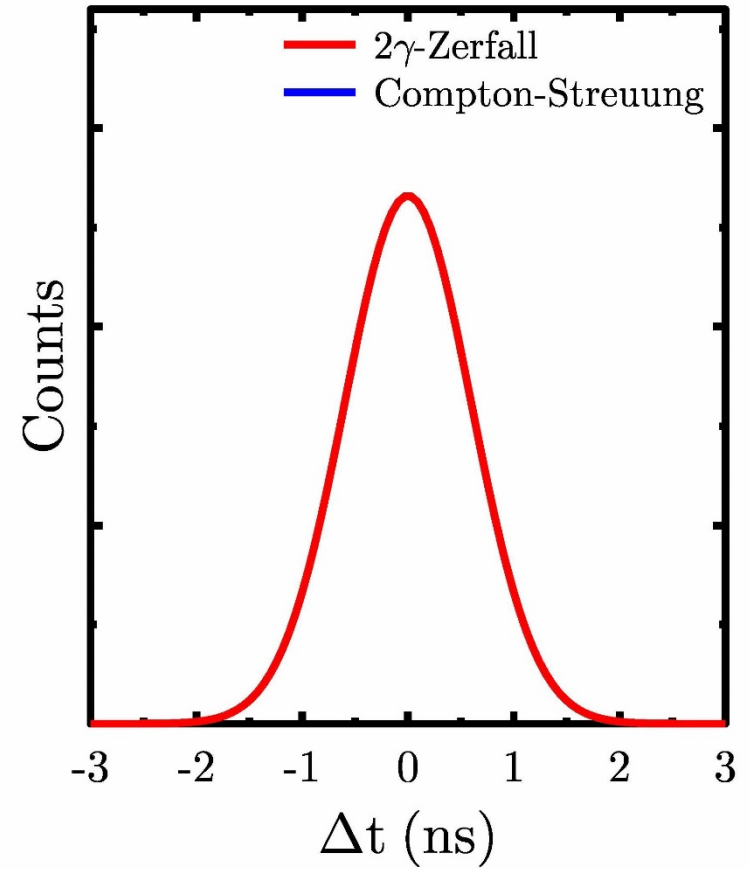
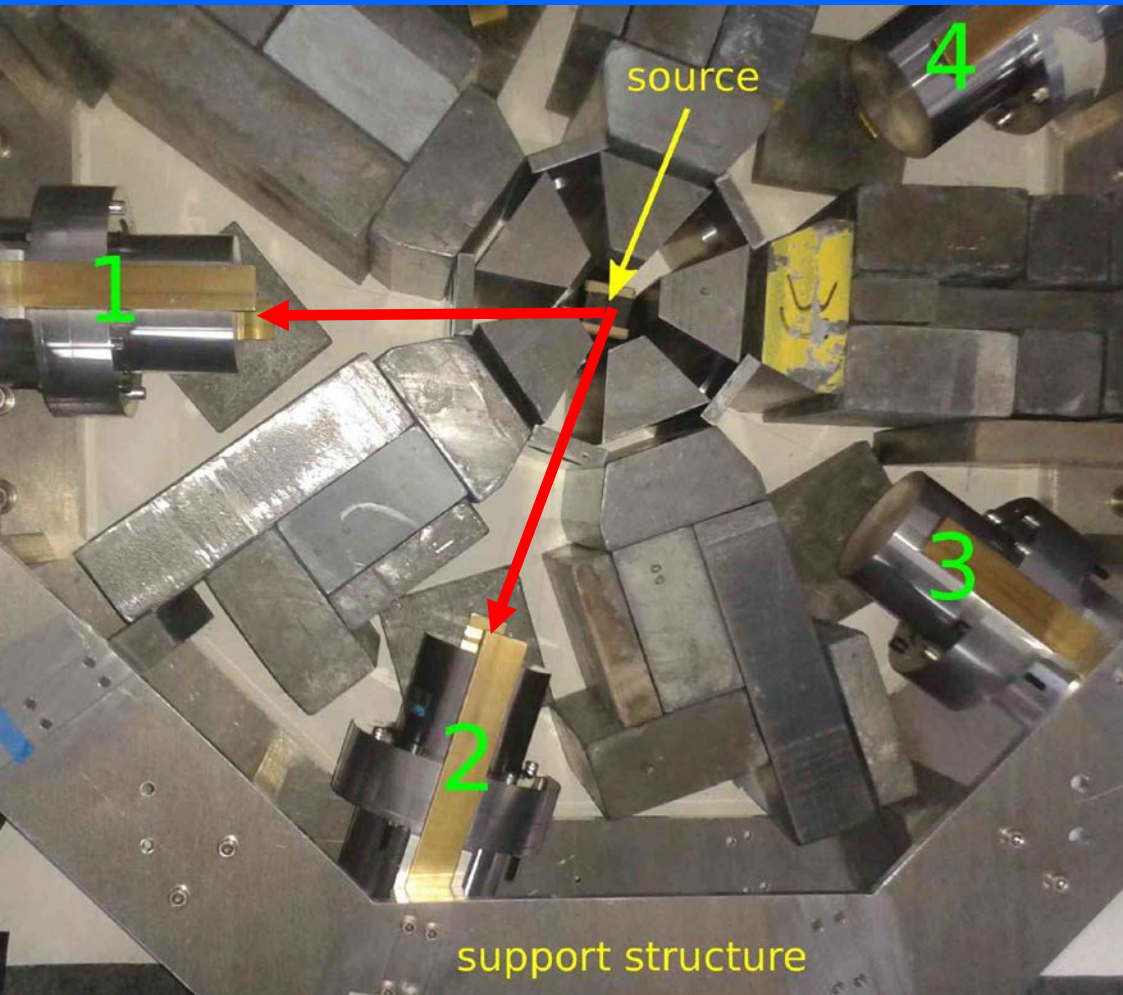


325(76) counts ( $\sigma = 4.3$ )  
 $\Gamma_{\gamma\gamma}/\Gamma_\gamma = 0.70(18) \cdot 10^{-6}$

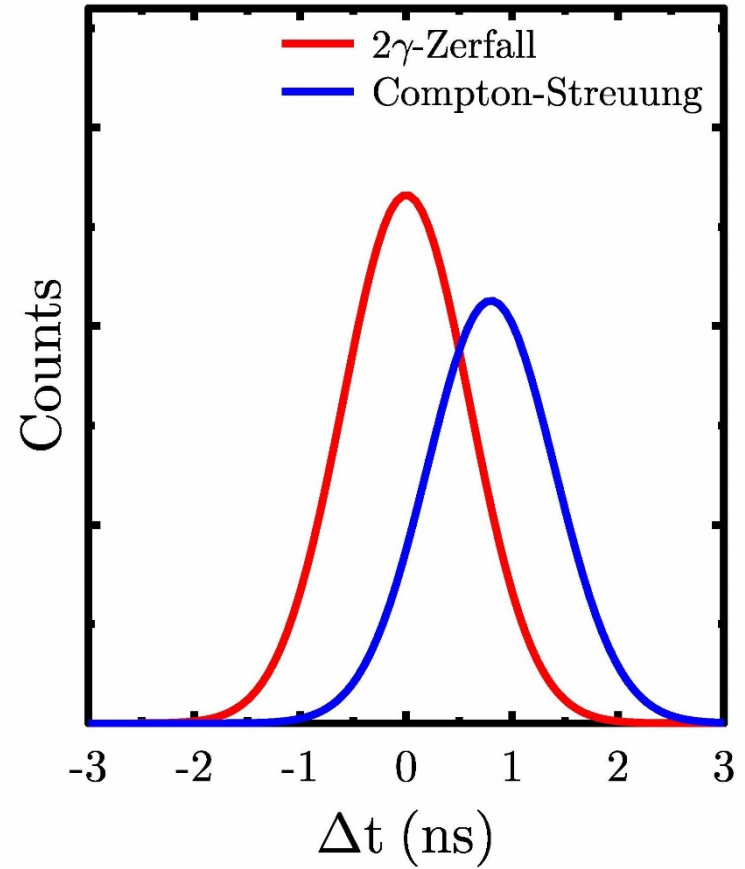
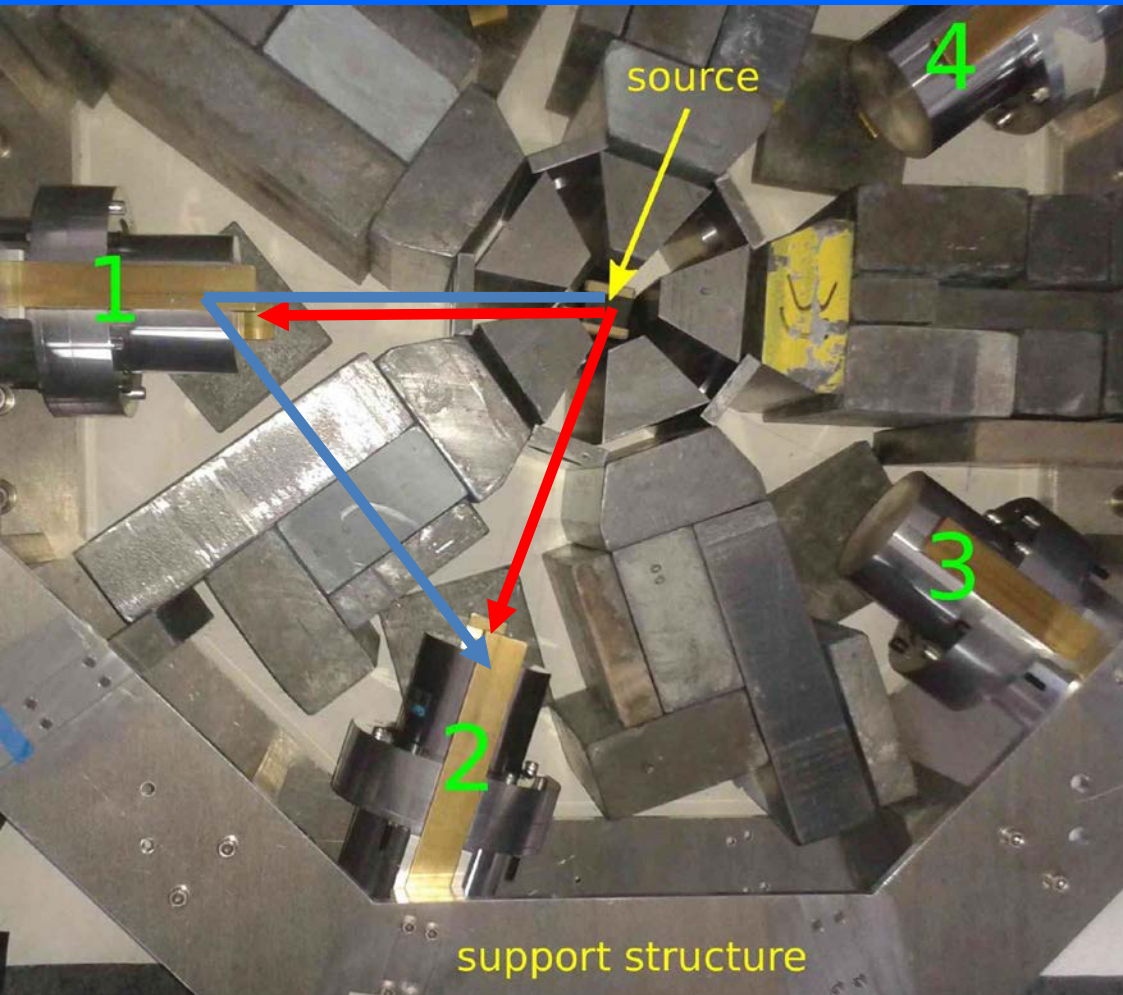
**observation** of the competitive double-gamma decay  
very pronounced **angular correlation**



# Time analysis

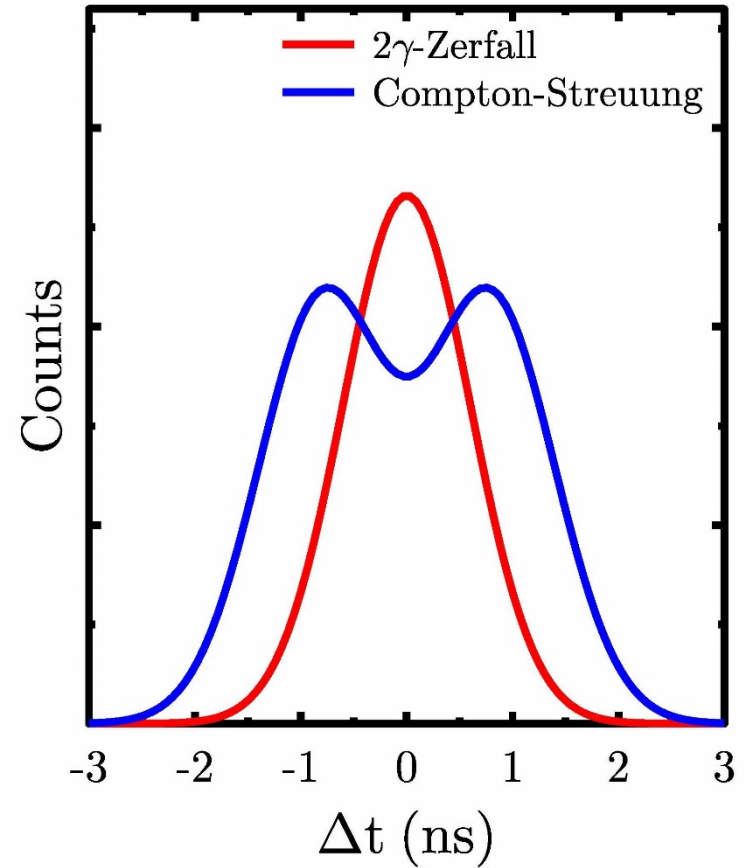
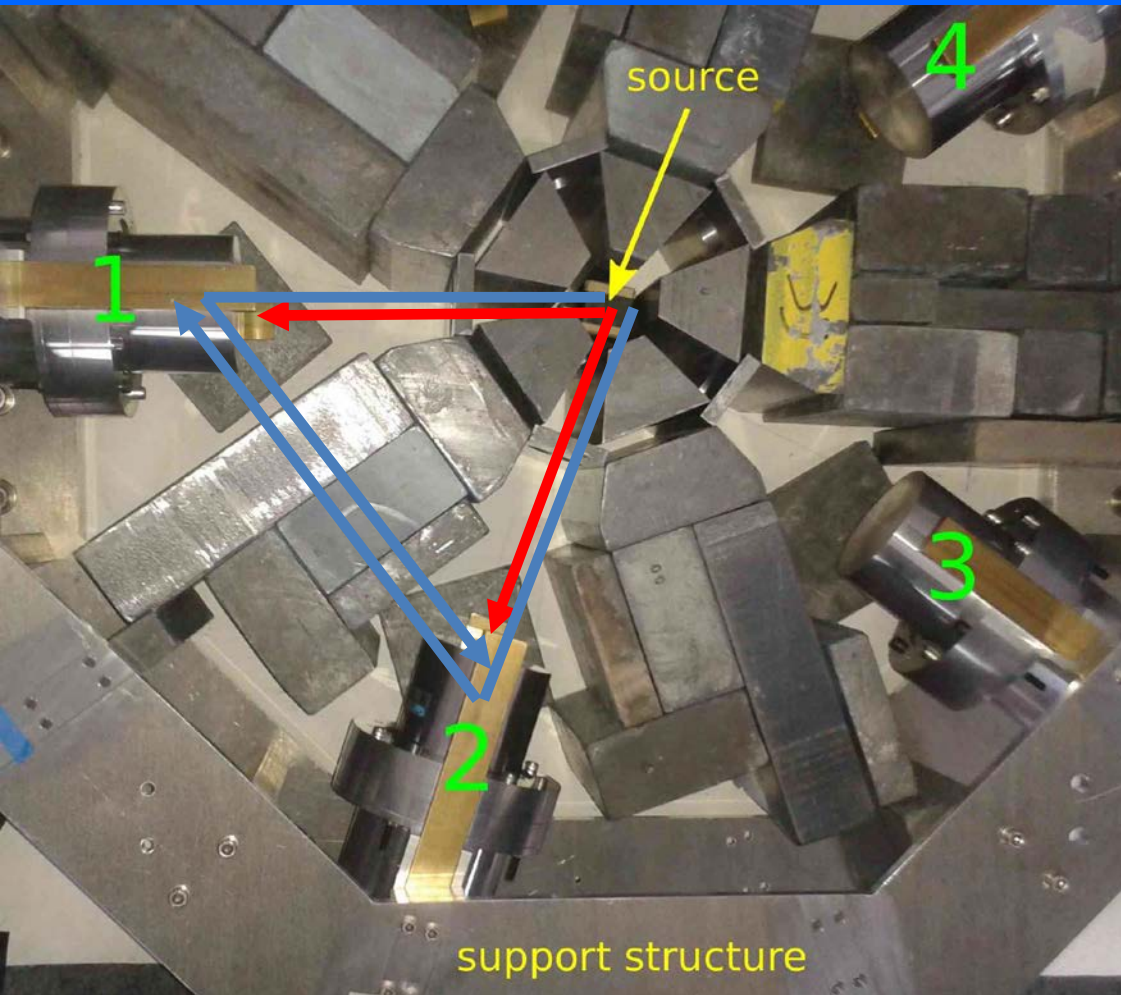


# Time analysis



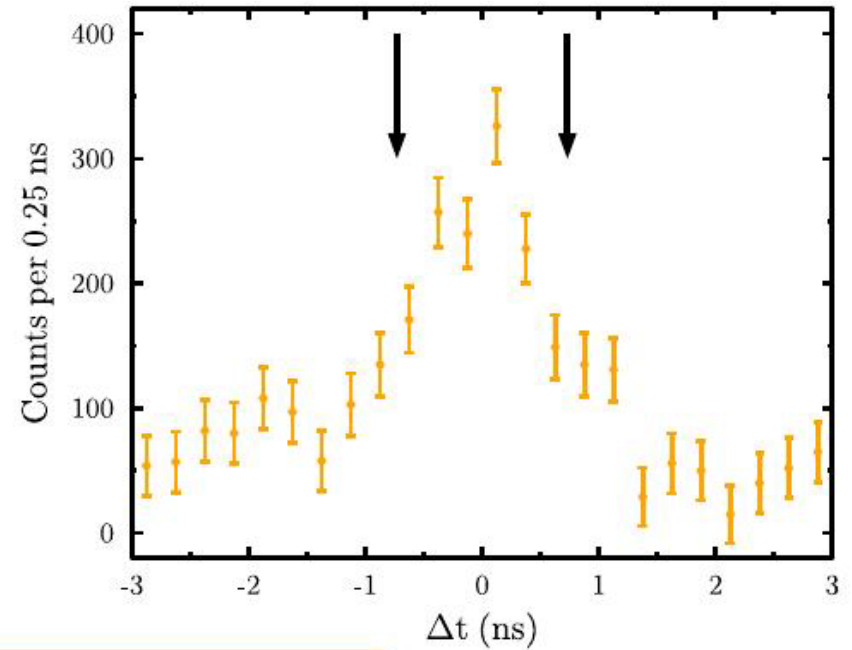
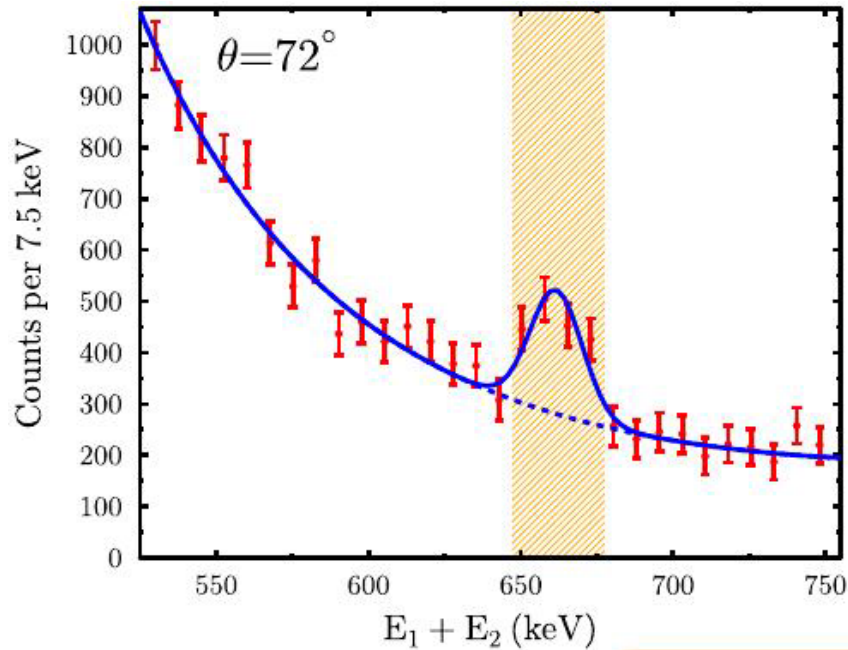


# Time analysis



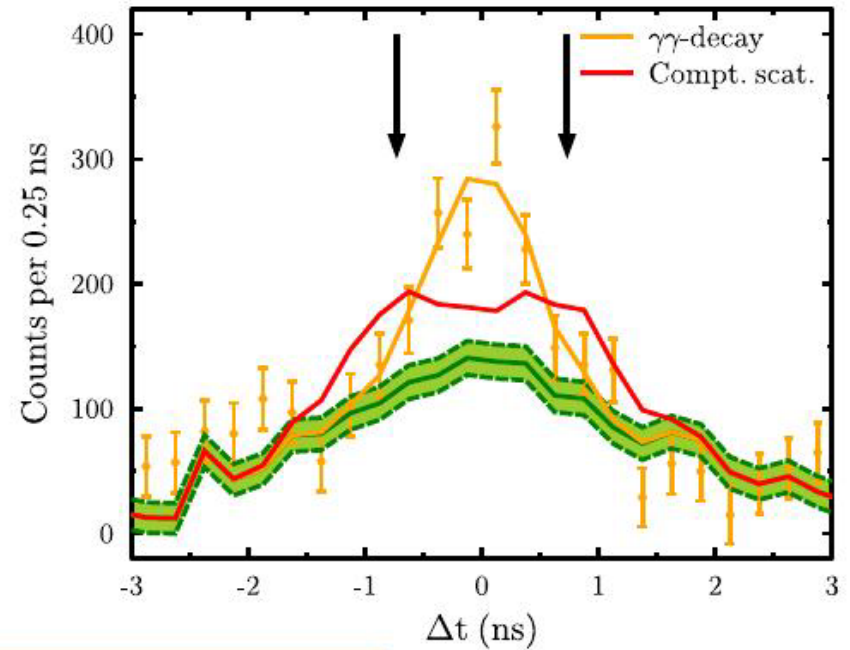
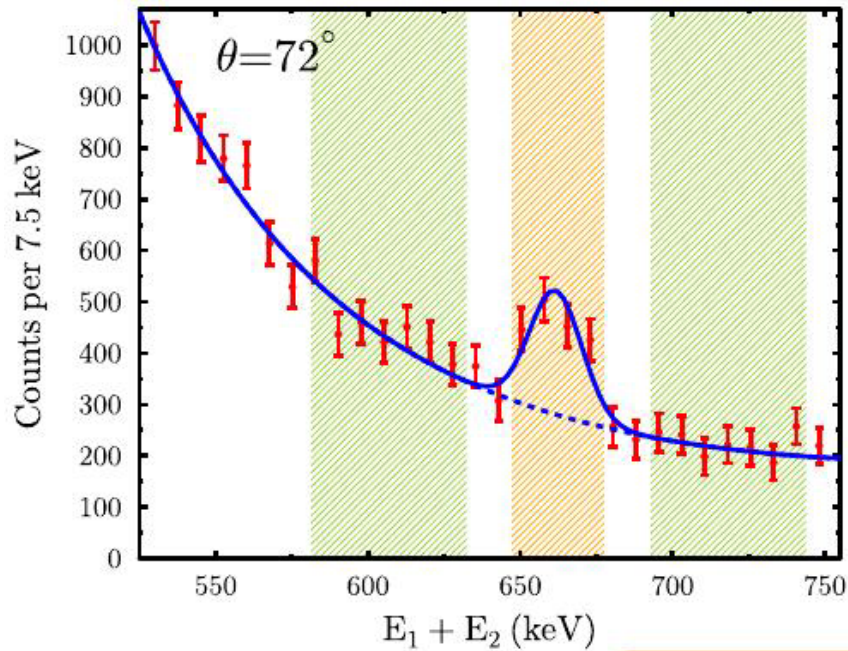
- Compton scattering should be visible in time spectrum

# Time analysis



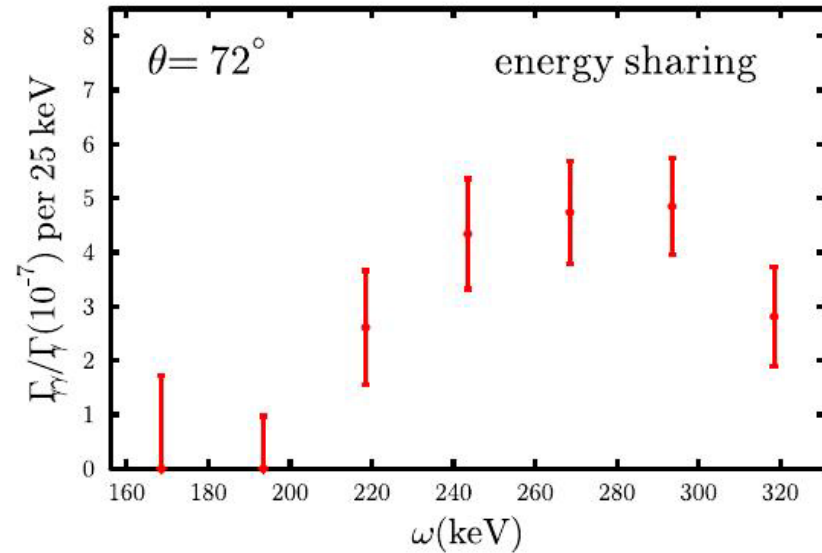
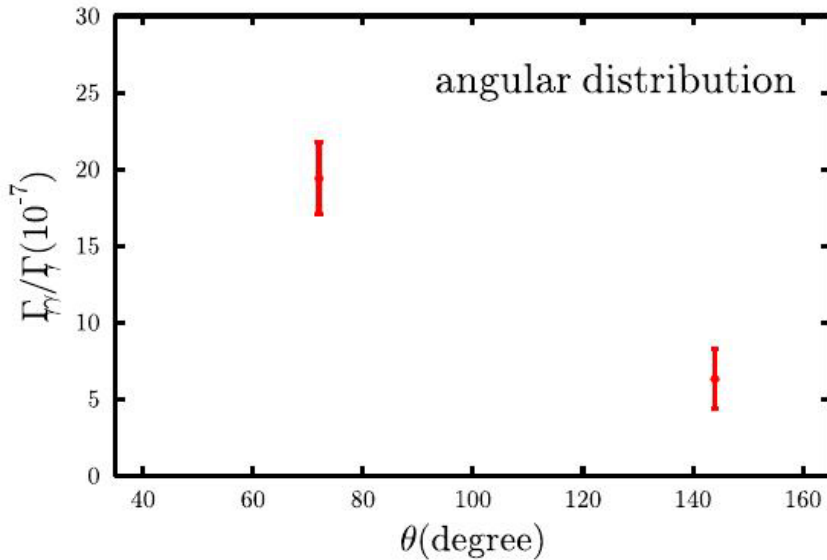
Shape of the time spectrum proves that Compton scattering is not the origin of the peak at 662 keV

# Time analysis



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

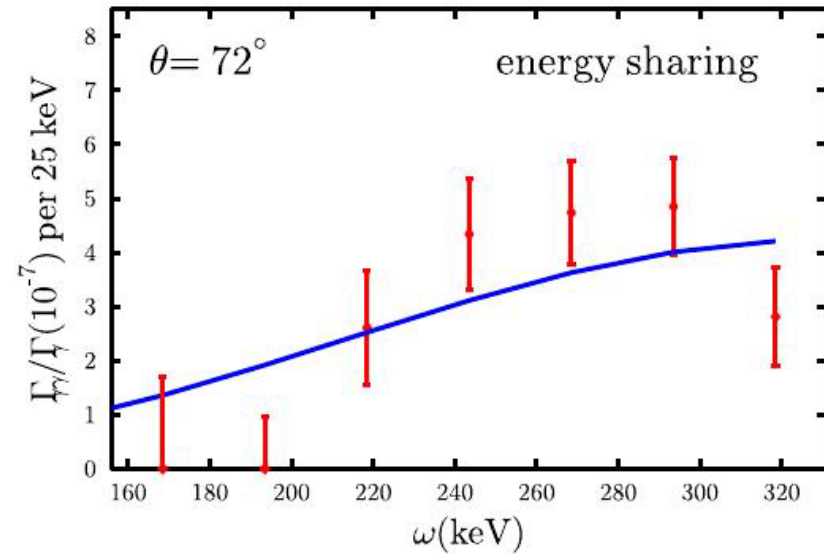
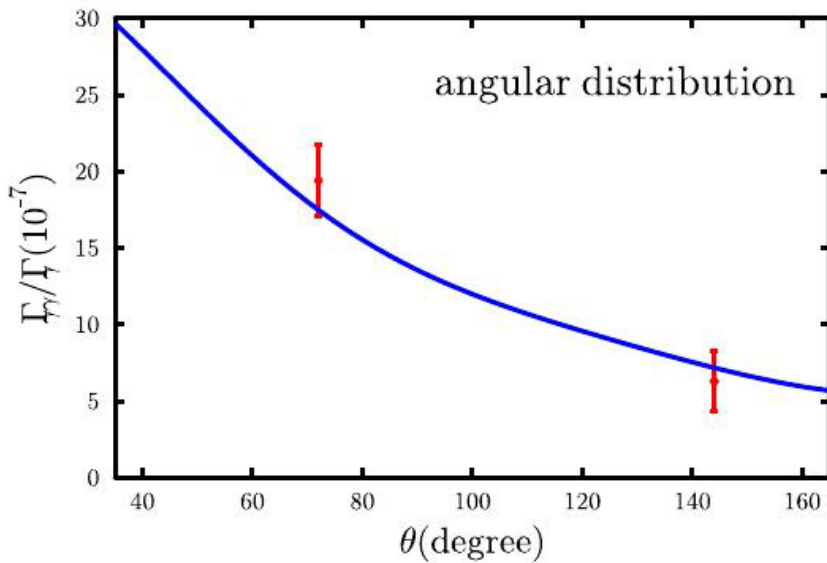
# Results



$$\frac{d\Gamma_{\gamma\gamma}^2}{d\omega d\cos\theta} = A_{qq}(\alpha_{M2E2}^2) + A_{od}(\alpha_{E3M1}^2) + A_x(\alpha_{M2E2} \cdot \alpha_{E3M1})$$

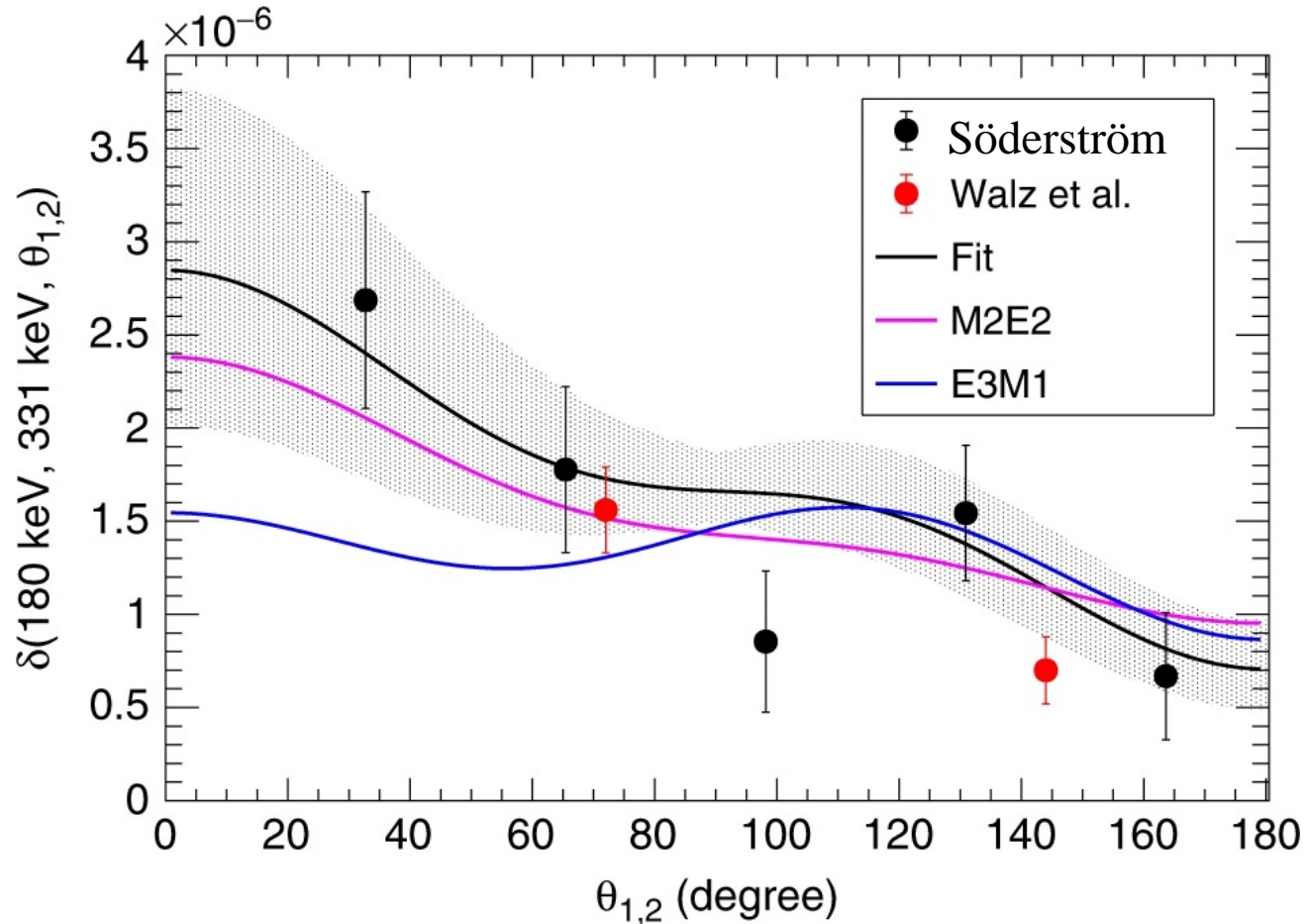


# Results



$$\frac{d\Gamma_{\gamma\gamma}^2}{d\omega d\cos\theta} = A_{qq}(\alpha_{M2E2}^2) + A_{od}(\alpha_{E3M1}^2) + A_x(\alpha_{M2E2} \cdot \alpha_{E3M1})$$

# Angular distribution



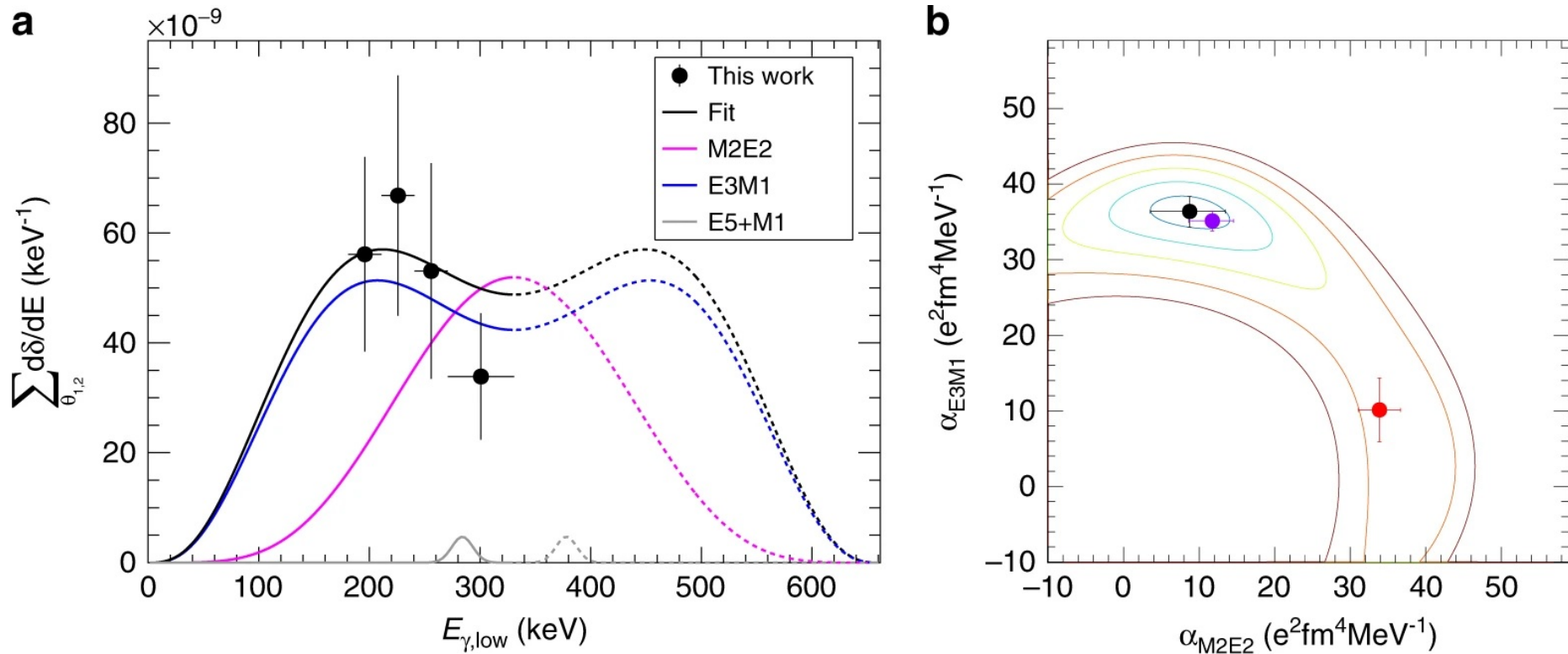
$$W_{\ell=2, \ell=2}(\theta) = 1 + \frac{1}{3}P_1(\cos\theta) + \frac{5}{49}P_2(\cos\theta) + \frac{1}{7}P_3(\cos\theta) + \frac{40}{441}P_4(\cos\theta)$$

$$W_{\ell=1, \ell=3}(\theta) = 1 - \frac{1}{8}P_1(\cos\theta) + \frac{5}{49}P_2(\cos\theta) + \frac{3}{8}P_3(\cos\theta)$$

no symmetry axis:  
odd Legendre polynomials

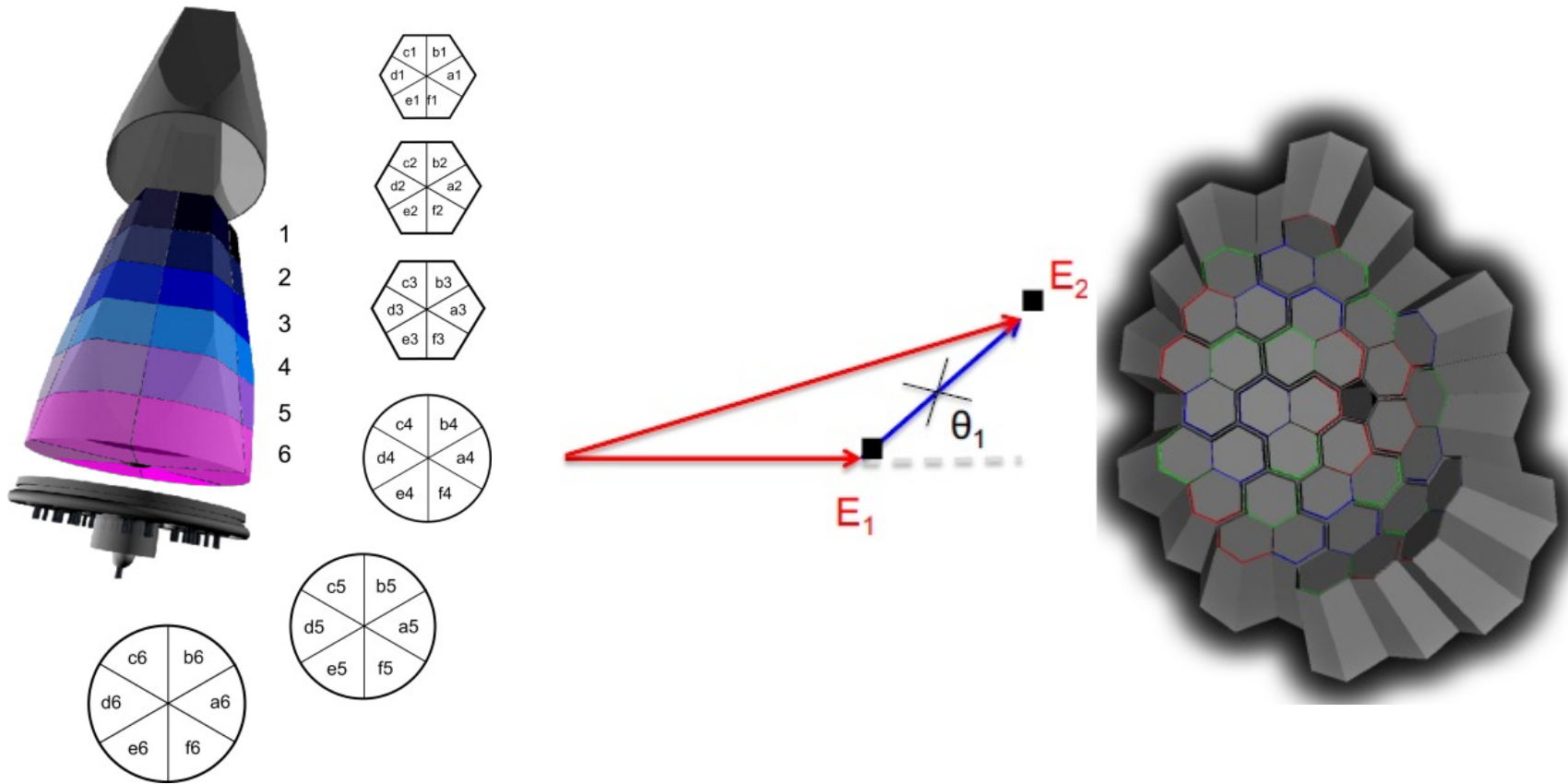


# Multipole nature of the $\gamma\gamma/\gamma$ decay



	$\Gamma_{\gamma\gamma}^{exp} / \Gamma_{\gamma}^{exp}$ ( $10^{-6}$ )	$\alpha_{M2E2}$ ( $e^2 fm^4 / MeV$ )	$\alpha_{E3M1}$ ( $e^2 fm^4 / MeV$ )
Walz	2.05(30)	33.9(28)	10.1(42)
Söderström	2.62(37)	$\pm 8.8(50)$	$\pm 36.4(20)$

# Future: AGATA for the $\gamma\gamma$ -decay



Position sensitivity and PSA to get spatially a difference between Compton scattered events and real double gamma events

# Appendix: Legendre polynomials

$$P_0(\cos\theta) = 1$$

$$P_1(\cos\theta) = \cos\theta$$

$$P_2(\cos\theta) = \frac{1}{2}(3\cos^2\theta - 1)$$

$$P_3(\cos\theta) = \frac{1}{2}(5\cos^3\theta - 3\cos\theta)$$

$$P_4(\cos\theta) = \frac{1}{8}(35\cos^4\theta - 30\cos^2\theta + 3)$$

$$P_5(\cos\theta) = \frac{1}{8}(63\cos^5\theta - 70\cos^3\theta + 15\cos\theta)$$

$$P_6(\cos\theta) = \frac{1}{16}(231\cos^6\theta - 315\cos^4\theta + 105\cos^2\theta - 5)$$

