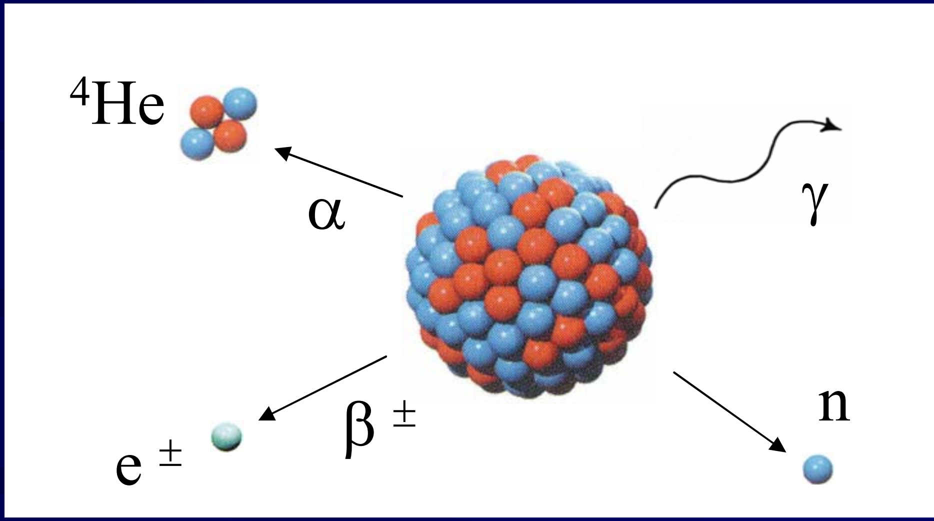
The metrology of radioactivity

C. Michotte, BIPM

SUMMARY

- Introduction
- Nuclear data of interest in activity measurements
- Activity measurement methods (primary and secondary)
- Monte Carlo simulations
- The role of the BIPM

Introduction



ACTIVITY / becquerel = Total number of decays per second for a given sample

Natural radioactivity of 1 litre of water : 15 Bq

ACTIVITY CONCENTRATION / ($Bq\ g^{-1}$) = activity per unit mass of the sample

Typical primary standard : 500 Bq / mg

Application (non exhaustive)	Source type	Activity level	Relative uncertainty (user need)
Nuclear medicine (imaging, therapy, research) *see next class*	Liquid, gas or solid β, γ	kBq to GBq	$(2 \text{ to } 10) \times 10^{-2}$
Nuclear physics (detector calibrations)	Point source α, β, γ, n	MBq	$(1 \text{ to } 5) \times 10^{-2}$
Environment (air, soil, food contamination and radioactive waste)	Spiked material α, β, γ, n	mBq to kBq	20×10^{-2}

Nuclear data : Half life $T_{1/2}$

Exponential decay :

$A(t) = A(t = t_{ref}) \exp(-\Delta t \ln(2)/T_{1/2})$ with $\Delta t = t - t_{ref}$

any activity value must be given with a date t_{ref}

- Measurement as close as possible to t_{ref} is essential for short half lives
- $T_{1/2}$ value and uncertainty are crucial :

$$A(t = t_{ref}) = A(t) \exp(+\Delta t \ln(2)/T_{1/2})$$

↓ propagation of uncertainties

$$u(A(t = t_{ref})) = u(T_{1/2})\ (A(t)\Delta t \ln(2) / T_{1/2}^{\ 2}) \exp(\Delta t \ln(2)/T_{1/2})$$

$$u(A(t = t_{ref}))/A(t = t_{ref}) = (\Delta t \ln(2) / T_{1/2})\ (u(T_{1/2})/T_{1/2})$$

Measurement of $T_{1/2}$

- Measure the activity during the decay, for a duration of several half lives
  - highly stable instrument
  - behaviour of the detector as a function of the count rate must be well-known

=> using an ionization chamber : rel. uncert. of 10^{-4} at best

- Not applicable to long half lives ($T_{1/2}$ > 50 a)

  other method : $A(t) = \ln(2)\, N(t) \,/\, T_{1/2}$

  └→ Number of radioactive atoms

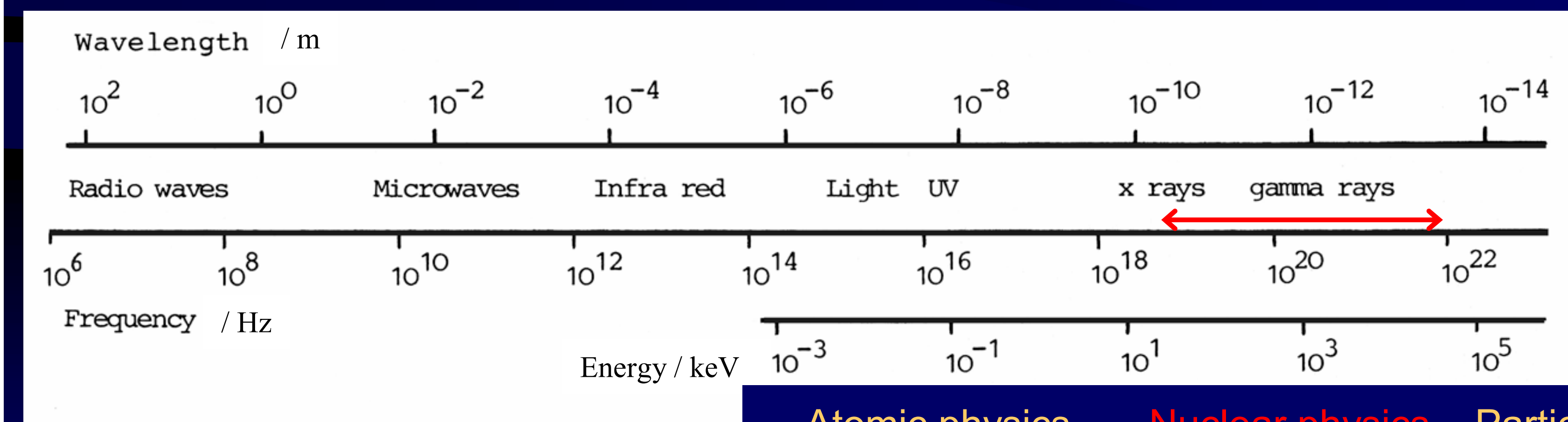
Analysis of published values => $T_{1/2}$ values and uncertainties evaluated and recommended to the users by NMIs or IAEA

Nuclear data : energies

The measured energies are used to identify a radionuclide

- Particle ($e^{\pm}$, α) energies : using magnetic spectrometers

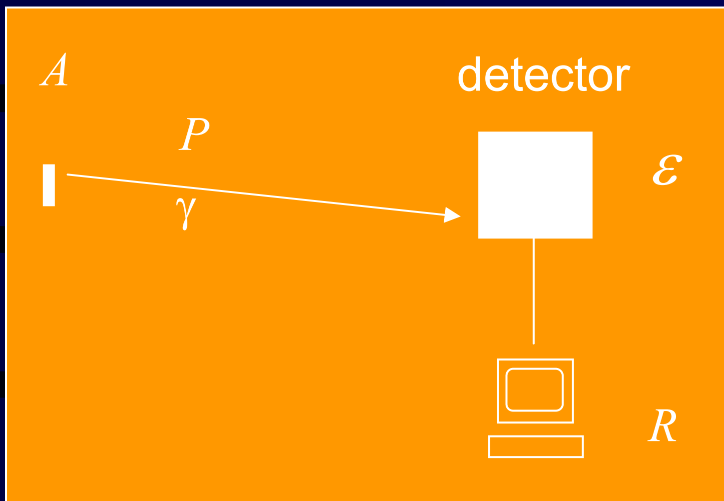
- Photon energies : related to optical wavelength for some γ-rays used as references (rel . uncert. 5×10^{-6})
[Bragg diffraction on Si calibrated using a He-Ne laser]



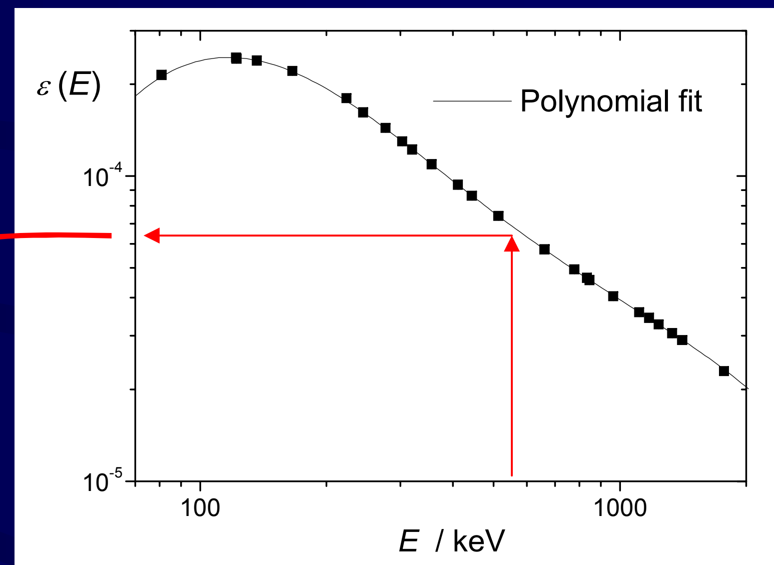
Atomic physics Nuclear physics Particle physics

Nuclear data : emission probabilities P

The case of γ rays



Calibration of detection efficiency using other radionuclides



$R = A\,P\,\varepsilon \Rightarrow P = R\,/\,(A\;\varepsilon\,)$

Primary meas.

$\varepsilon\,(E_i) = R_i\,/\,(A_i\,P_i)$

ITERATION

Other primary meas.

Hundreds of radionuclides / thousands of γ transitions

=> huge iterative process slowly converging since the 1960's

Presently : relative $u(P_\gamma)$ is 10^{-3} to 10^{-2} in general

- Many national recommended databases published
- More and more international coordination to share the work and produce internationally recommended data
- Correlations in the data need to be considered

Primary measurement methods

- primary standards are the measurement methods (radioactive sources are decaying !)
- primary = independent of other standards
  and of emission probabilities
- the decay scheme determines the choice of method
  often, the choice is not unique
  => compare the methods
  => robust results !

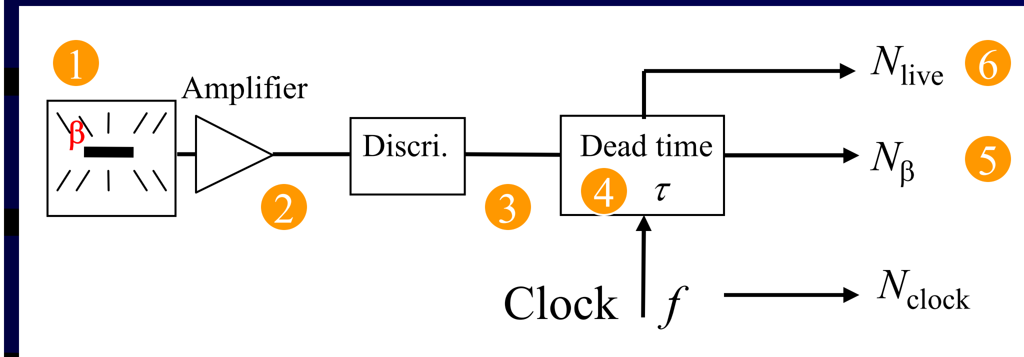
Primary measurement methods

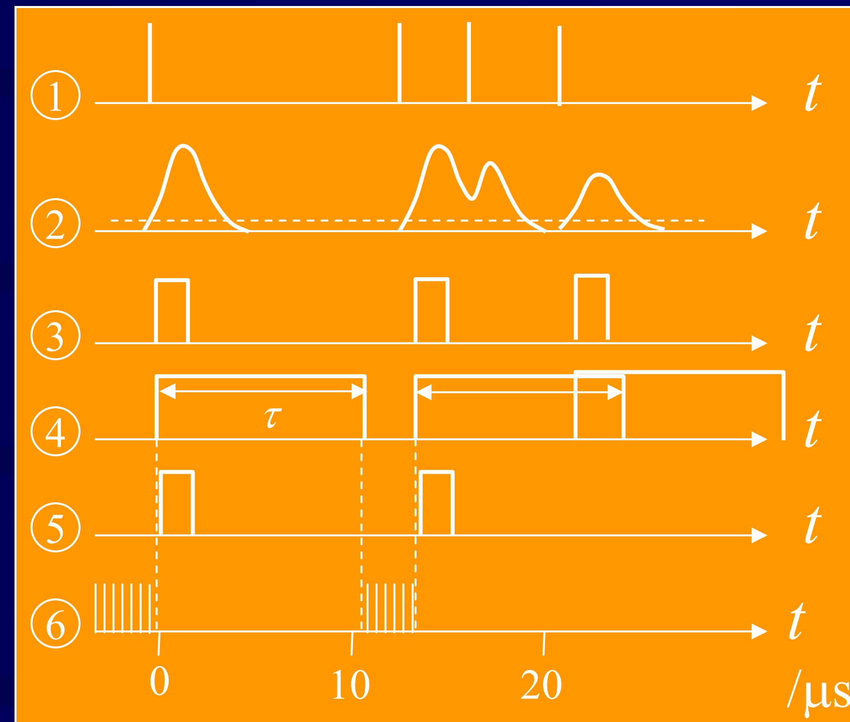
- CORRECTIONS COMMON TO ALL METHODS
- EXAMPLES of PRIMARY MEASUREMENTS METHODS

1. Coincidence method
2. $4\pi\gamma$ counting
3. Defined solid angle counting for α emitters

Corrections common to all methods

- Dead-time loss : basic principle



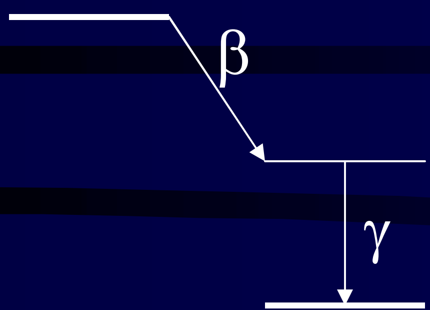


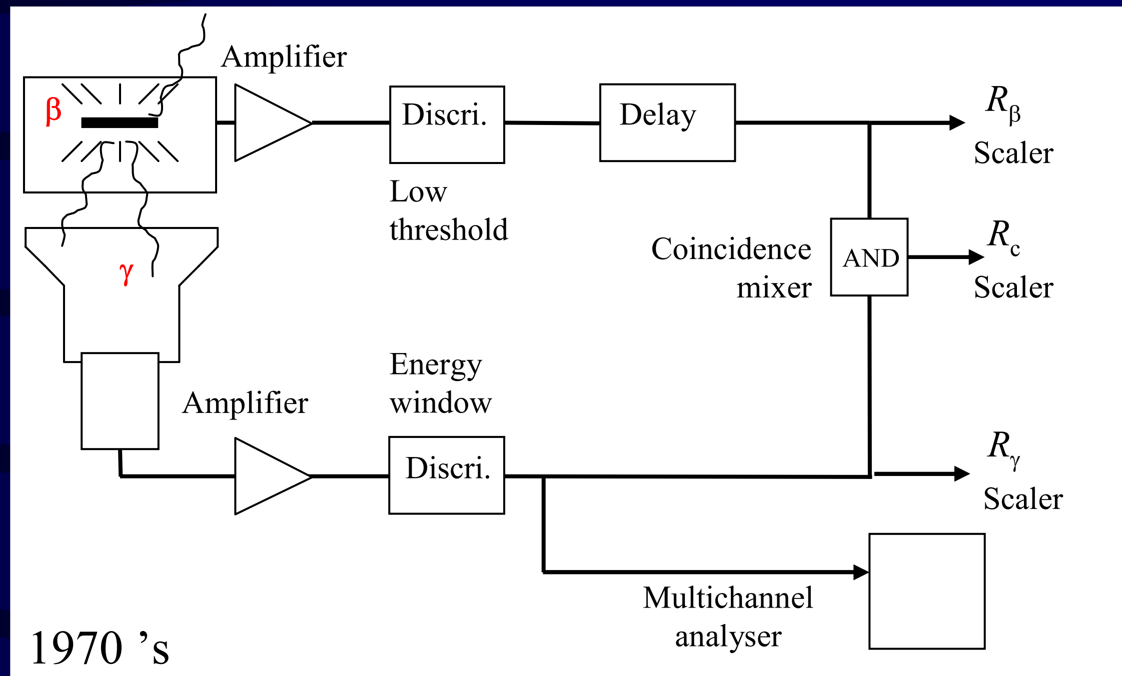
$$R_{\beta} = (N_{\beta} / d)\ \underline{(N_{clock}/N_{live})}$$

Dead-time correction

- Exponential decay
- Background (natural radioactivity, cosmic rays)
- Radioactive impurities

EXAMPLE 1 :
the coincidence method for β-γ decay (or α-γ)





$$R_\beta = A\, P_\beta\, \varepsilon_\beta$$

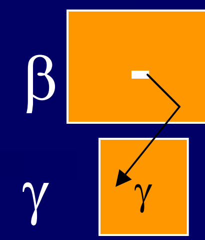
$$R_\gamma = A\, P_\gamma\, \varepsilon_\gamma \qquad R_c = A\, P_\beta\, P_\gamma\, \varepsilon_\beta\, \varepsilon_\gamma$$

$$\Rightarrow A = R_\beta\, R_\gamma \,/\, R_c$$

Random coincidences (e.g. between β and γ coming from 2 different desintegrations) : function of the resolving time of the coincidence and of the β and γ count rates

β detector (β energy spectrum is continuous : 0 to E_{max})

- low energy threshold => window-less and 4π geometry
- low *Z* and *ρ* material to minimize the γ efficiency (false coincidences : see later)

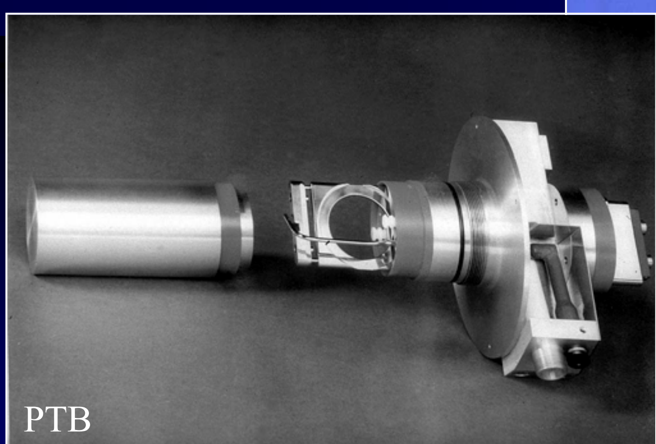


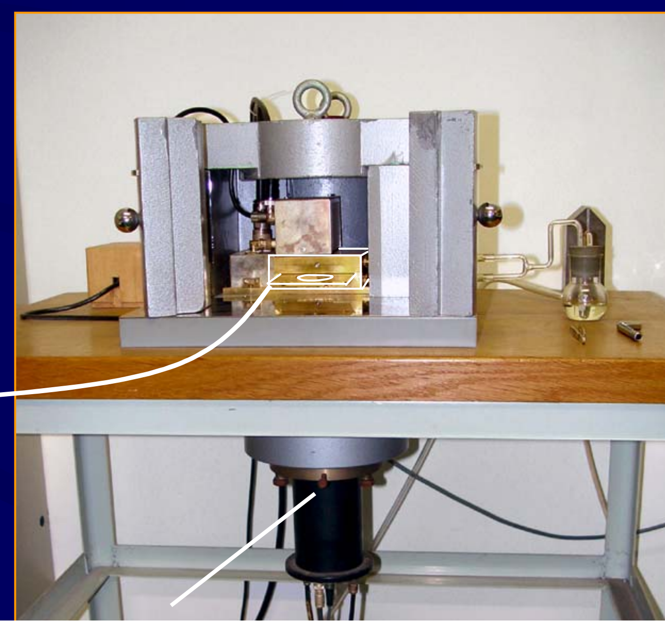
=> Proportional counters (PC), liquid or plastic scintillators

Atmospheric PC

gas = Ar , CH_4

Pressurized PC





γ detector NaI(Tl)

high *Z* material to maximize the efficiency

Extrapolation in the coincidence method

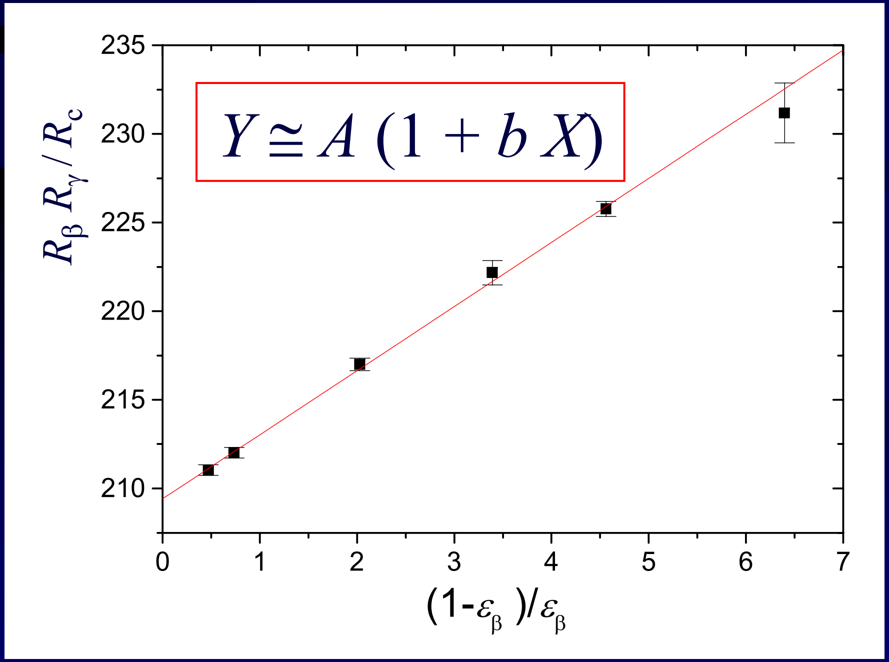
$$\begin{cases} R_\beta = A\,P_\beta\,[\varepsilon_\beta + (1-\varepsilon_\beta)\,\varepsilon_k\,] \\ R_\gamma = A\,P_\gamma\,\varepsilon_\gamma \\ R_c = A\,P_\beta\,[\varepsilon_\beta P_\gamma\,\varepsilon_\gamma + (1-\varepsilon_\beta)\,\varepsilon_c\,] \end{cases}$$

ε_k is for $\varepsilon_{\beta\gamma}$, ε_{ce}, ...

ε_c is for « false » coincidences

=> $$\frac{R_\beta R_\gamma}{R_c} \cong A\left[1 + \left(\frac{1-\varepsilon_\beta}{\varepsilon_\beta}\right)\left(\varepsilon_k - \frac{\varepsilon_c}{P_\gamma \varepsilon_\gamma}\right)\right] \xrightarrow{\varepsilon_\beta \to 1} A$$

b

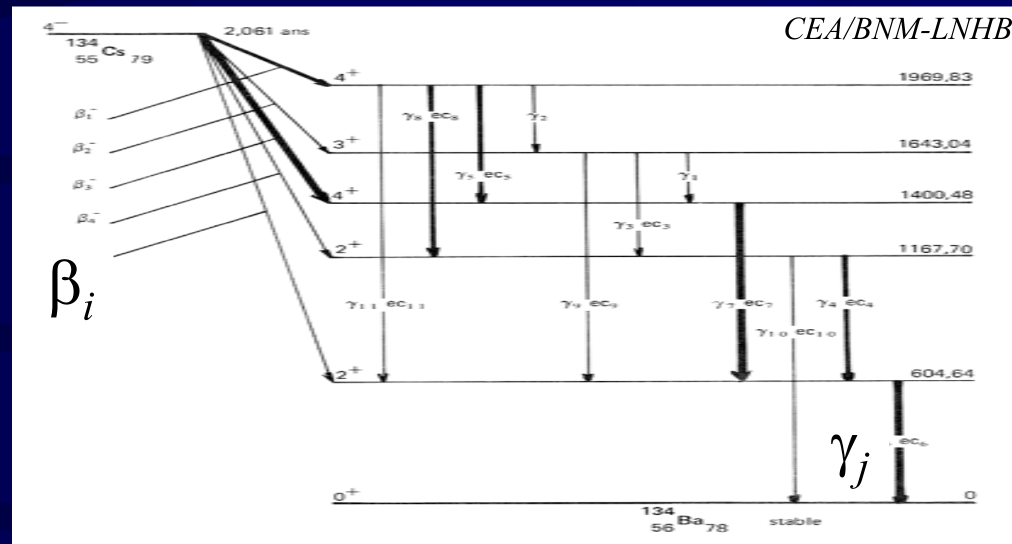


ε_β is estimated by $R_c\,/\,R_\gamma$

Vary ε_β with negligible change in b :

- absorber on the source (solid)
- change of detector/electronics parameters (electronic threshold, HV, gas pressure,...)
- chemical quenching of liquid scintillator

For complex decay scheme :



$$R_\beta = A \sum P_{\beta,\,i}\,[\varepsilon_{\beta,\,i} + (1-\varepsilon_{\beta,\,i})\,\varepsilon_{k,\,i}\,]$$

$$R_\gamma = A \sum P_{\beta,\,i}\,P_{\gamma,\,i}\,\varepsilon_{\gamma,\,i}$$

$$R_c = A \sum P_{\beta,\,i}\,[\varepsilon_{\beta,\,i}\,P_{\gamma,\,i}\,\varepsilon_{\gamma,\,i} + (1-\varepsilon_{\beta,\,i})\,\varepsilon_{c,\,i}\,]$$

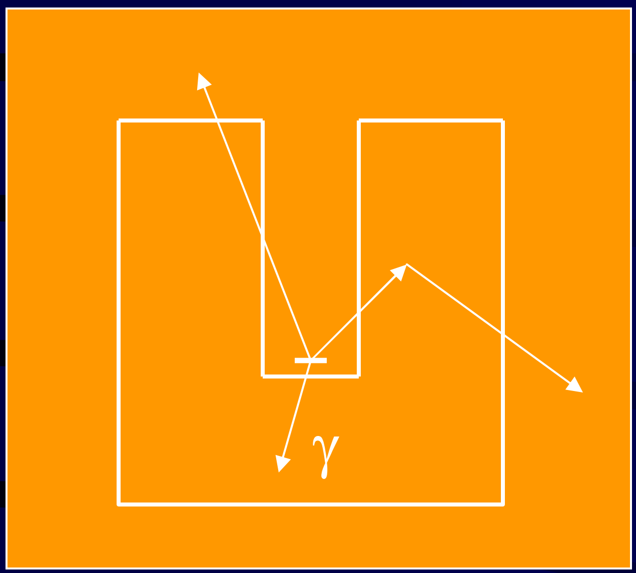
$$\frac{R_\beta R_\gamma}{R_c} = ??$$

- selection of some γ rays (energy window)
- experienced assumptions for parameters $\varepsilon_{k,\,i}$ and $\varepsilon_{c,\,i}$
- extrapolation not always linear

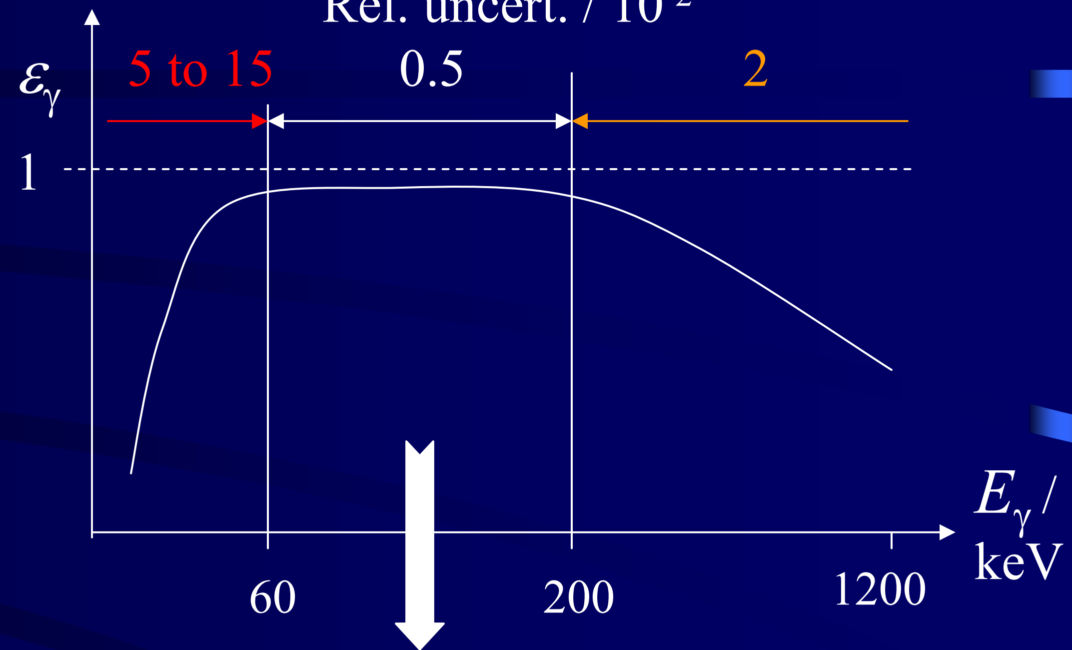
=> delicate and long exercise / rel. uncert. 10^{-3} to 10^{-2}

(5×10^{-4} at best)

EXAMPLE 2 :
4πγ counting with a well-type NaI(Tl) for multi-γ emitters



1980 's

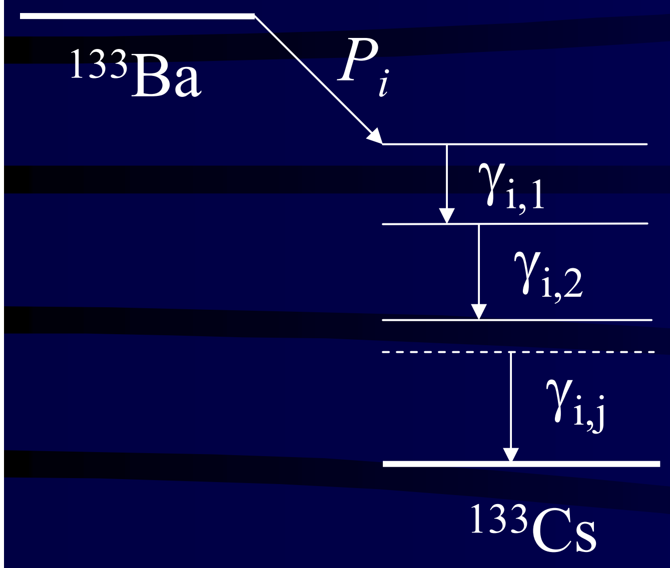


Best results for medium energy γ-rays

The efficiency curve is generally calculated
by analytical integration or by Monte Carlo simulations

Example 2 / continued

BASIC PRINCIPLE



$$\varepsilon\,(^{133}\mathrm{Ba}) = \sum_i P_i\, p(\text{detecting branch } i)$$

$$= \sum_i P_i\,[1 - p(\text{not detect. } \gamma\text{ 's})]$$

$$= \sum_i P_i\,[1 - \prod_j p(\text{not detect. } \gamma_{i,j})]$$

$$= \sum_i P_i\,[1 - \prod_j (1 - \varepsilon\,(\gamma_{i,j}))]$$

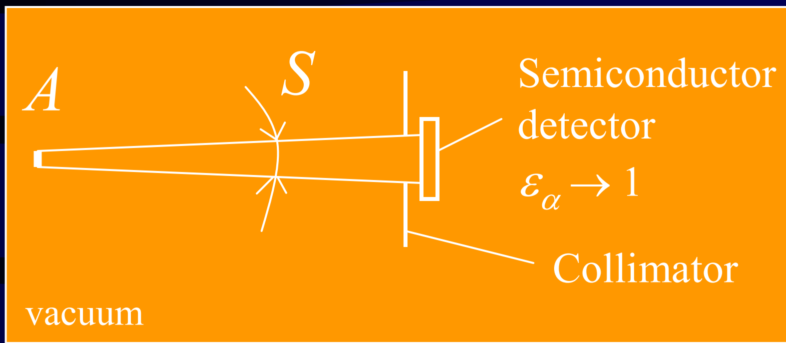
Effic. curve

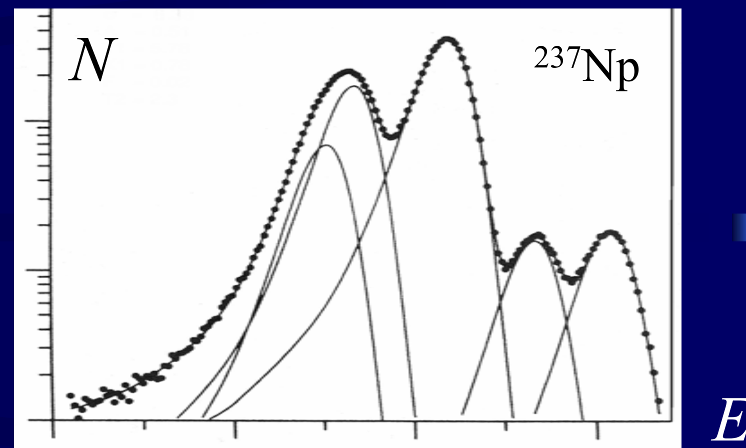
Example

ε_11	u	ε_12	u	ε_13	u	P_1
0.70	0.07	0.90	0.04	0.85	0.05	1
	0.03 & 0.06		0.03 & 0.03		0.03 & 0.04	
ε	0.9956	u_c	0.0034	no correlation		
			0.0038	correlated uncertainty = 3×10^{-2}		

EXAMPLE 3 : definite solid angle counting for α emitters

E_α : (4 to 6) MeV (stopped by 20 μm Al)

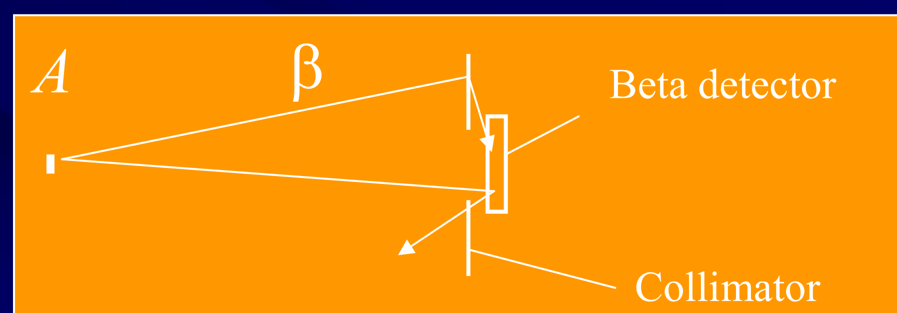




$$R_\alpha = A\,(\underline{\Sigma\, P_{\alpha i}})\ \varepsilon_\alpha\, S\,/\,4\pi$$

↳ = 1

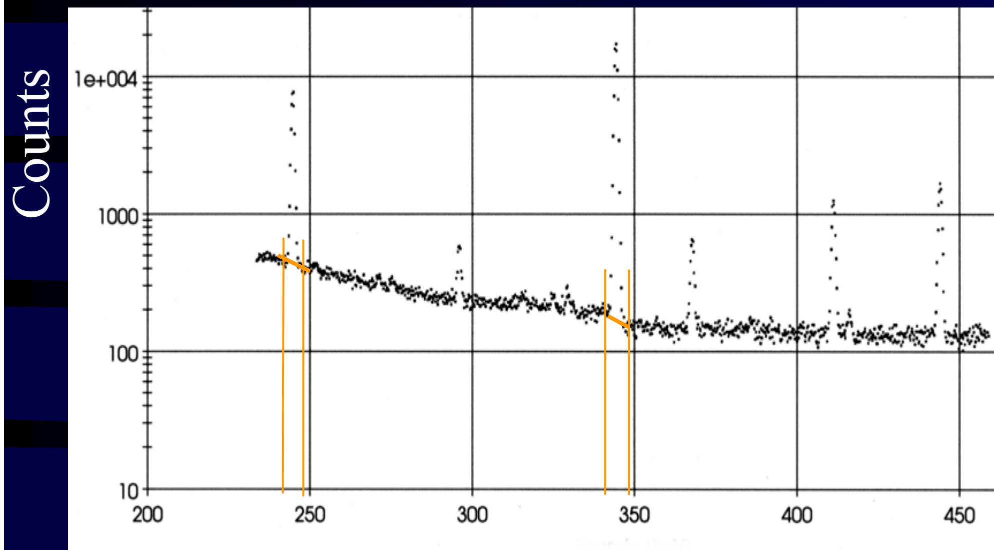
- relative uncertainty ~ 10^{-3}
- difficulties may come from
  - source preparation : thin and uniform to minimize self-absorption
  - a mixture of radionuclides, as for environmental samples (complex α spectrum to deconvolute)
- Not applicable to β emitters due to scattering :



Secondary measurement methods

EXAMPLE 1 : Ge γ-spectrometers

^{152}Eu



Good energy resolution => selection of peaks of interest => quantitative identification of radionuclides

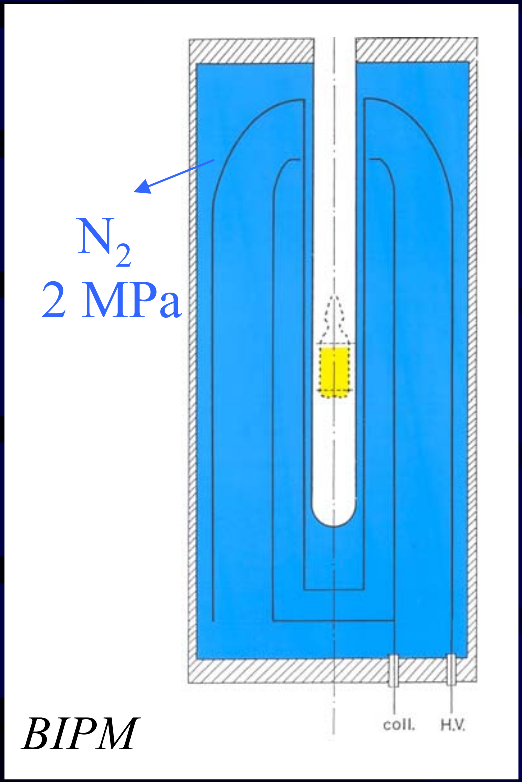
$$R_{\text{peak area}} = A\, P_\gamma\, \varepsilon_{\text{peak}}$$

efficiency calibration (vs energy), using a set of standards

Identical procedures for the calibration and the subsequent analysis

Widely used in nuclear research, environmental measurements, nuclear survey,…(commercial software)

EXAMPLE 2 : well-type ionization chamber (IC)



- Measurement of the ionization current (1 pA to 400 pA) produced by the emitted γ-rays (β-rays)
- Need for a calibration factor for each radionuclide and container (ampoule, syringe,…)
- Highly stable ($< 10^{-3}$ over 25 years)
  => often used - for half-life measurements
  - as secondary standards in NMIs
  => transfer instrument at the BIPM (*SIR*)
- Rapid and easy => widely used in hospitals

Uncertainty : - primary measurement used for the calibration
- IC measurement : 3×10^{-4} to 5×10^{-3}

Monte Carlo simulations

Monte-Carlo code

= set of subroutines containing the physics and tools for the calculation step by step of all the possible trajectories of a particle and its interaction with matter

KEY PARAMETER

Mean free path λ of a particle in given material at energy E :
$\lambda(E) = (N\sigma_T(E))^{-1}$ / m where $\sigma_T(E) = \sum \sigma_i(E)$ / m^2

N → atoms /m^3

$\sigma_i(E)$ → Calc. or meas. cross section σ_i for scattering, fluorescence,… ($u \sim$ a few 10^{-2})

MECHANISM

Random sampling of :

0. initial position and $\vec{v}$ of the particle
1. distance d to next interaction : $p\,(d = x) = \exp(- x / \lambda)$
2. type of interaction : p (interaction i) $= \sigma_i / \sigma_T$
   possible creation of secondary particle
   possible absorption of primary particle
3. angular and energy distribution
   for the outgoing particles

LOOP

=> NEW (x,y,z) and $\vec{v}$
+ record of e.g. the energy deposited in the detector

APPLICATIONS

- optimization of the geometry of a detection system
- calculation of detection efficiencies
  - need to know the geometry in detail
  - experimental verification is essential

  => apply the simulation to other similar geometries or energies
  => enables a reduction in the number of measurements
- study the influence of some parameter of the geometry
  => deduce correction factor or uncertainty value
- study the influence of a particular physical process

The role of the BIPM

- Organisation of (and participation in) international comparisons of activity measurements

Compare the activity concentration (Bq/g) of a given radioactive solution measured by each NMI at the same time

< 3 comparisons per year

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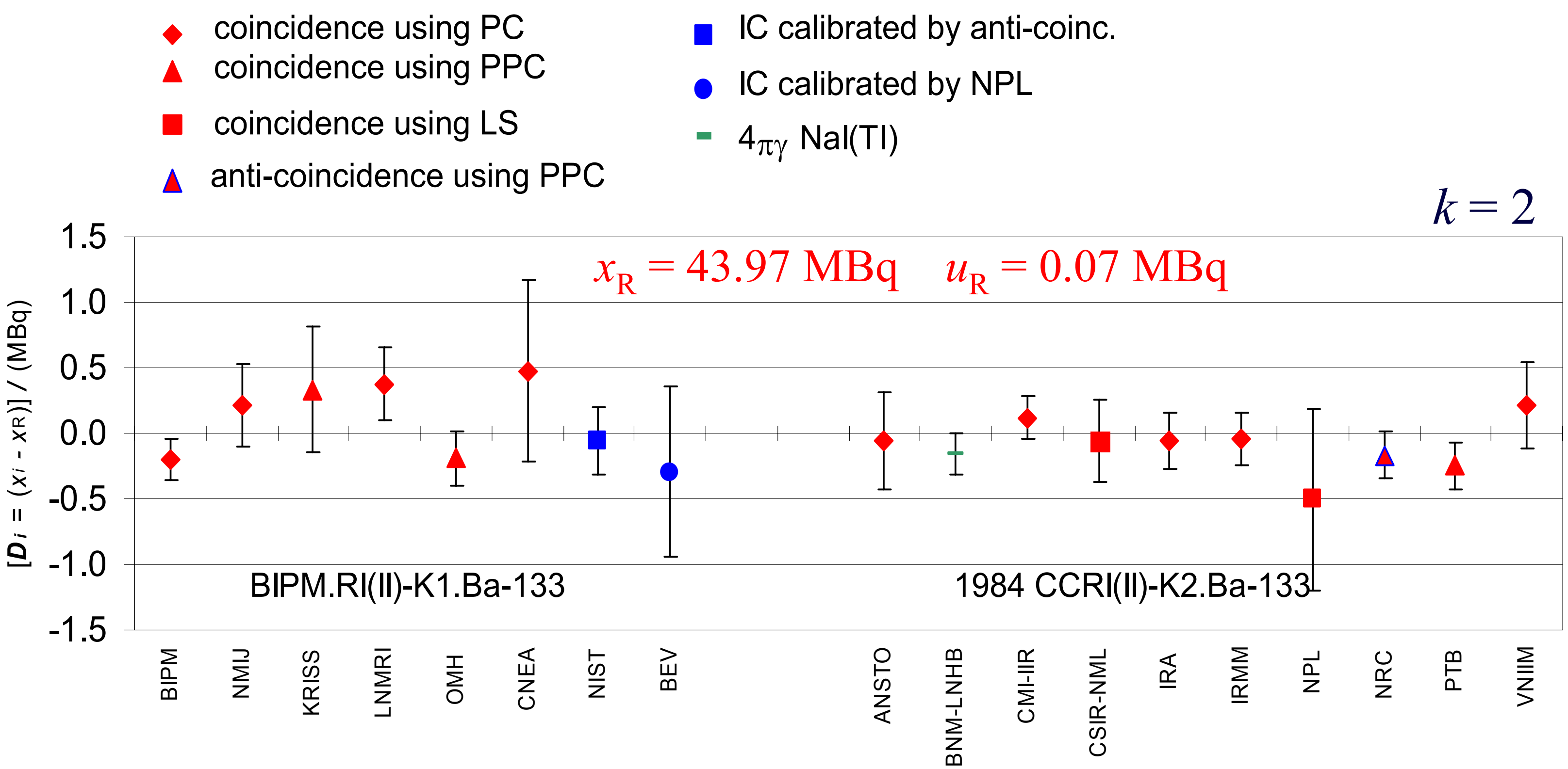
I
N
K

- « SIR » measurements at the BIPM (see visit)

On-going comparison. Participation of NMIs at their convenience

~ 20 measurements per year for some of 62 radionuclides

Degrees of equivalence for equivalent activity of ^{133}Ba



Conclusion

- Many radionuclides and nuclear data
- Many measurement methods

Huge amount of work …!

References

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