СОФИЙСКИ УНИВЕРСИТЕТ
„СВ. КЛИМЕНТ ОХРИДСКИ“

ФИЗИЧЕСКИ ФАКУЛТЕТ

SOFIA UNIVERSITY
ST. KLIMENT OHRIDSKI

FACULTY OF PHYSICS

Primary measurement methods in radionuclide metrology

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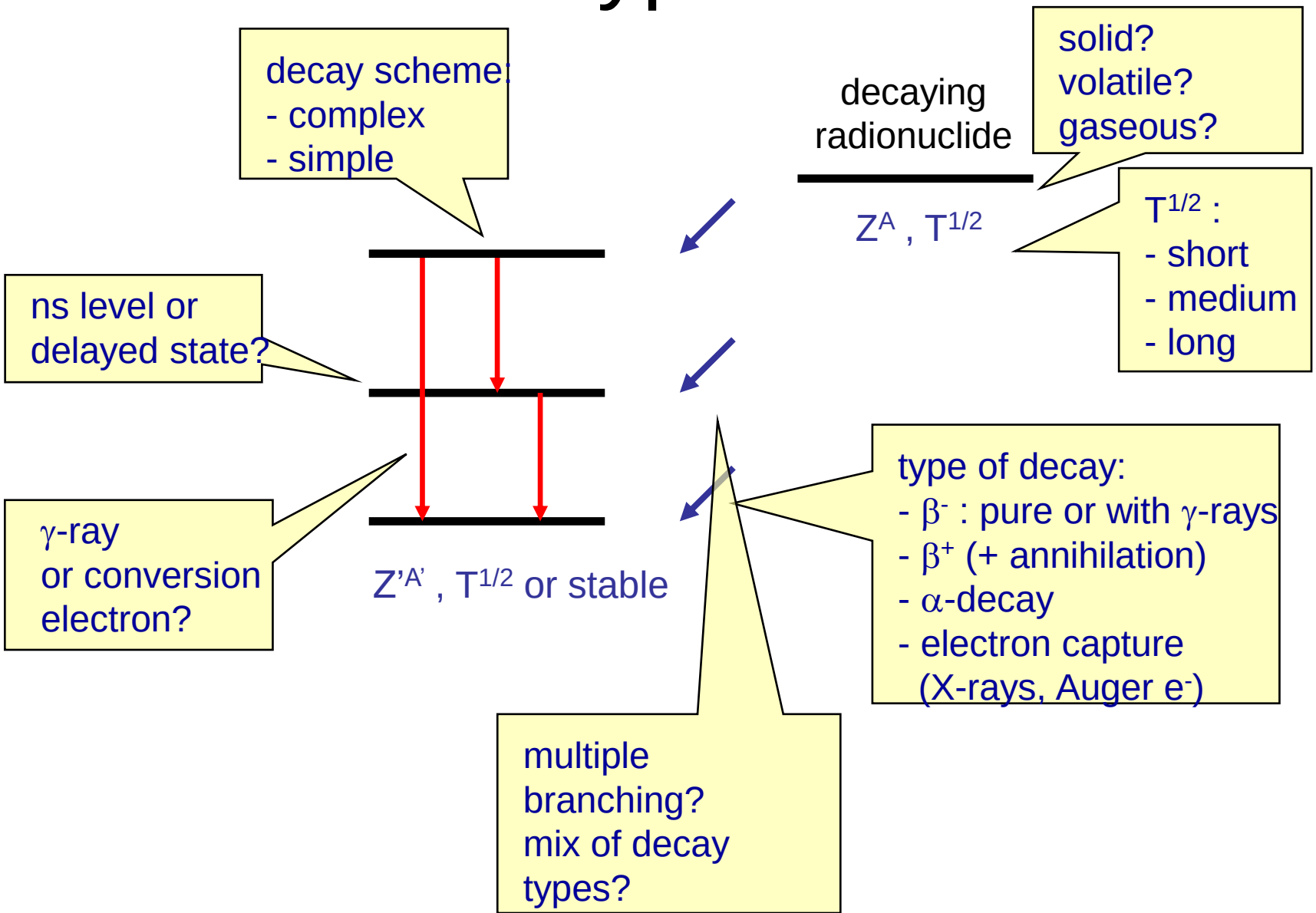
Realization of the SI-unit Becquerel

- *"Direct"* measurement of the number of *spontaneous transitions* of a radionuclide in a time interval
  = primary standardisation
  = realization of the Becquerel [s^{-1}]

- Primary standard
  = directly measured source
  + Activity + Uncertainty

a multitude of decay types



Primary measurement methods

- "Transitions" are measured through the emitted radiation (X, γ, α, e^-, e^+)
- Different physical detection principles and devices are used, depending on radionuclide.
- Counting efficiency should be
  - ≈100% with small corrections
    - high-geometry (4π) methods
  - <100%, but calculated with low uncertainty
    - coincidence counting
    - defined solid angle counting
- The 'ideal' primary method is accurate, precise, under statistical control, independent of decay scheme parameters and not based on calibrations with other radioactivity standards

Primary Standardisation of activity

Counting at a defined small solid angle

Defined Solid Angle = DSA

Particles should

- not easily scatter

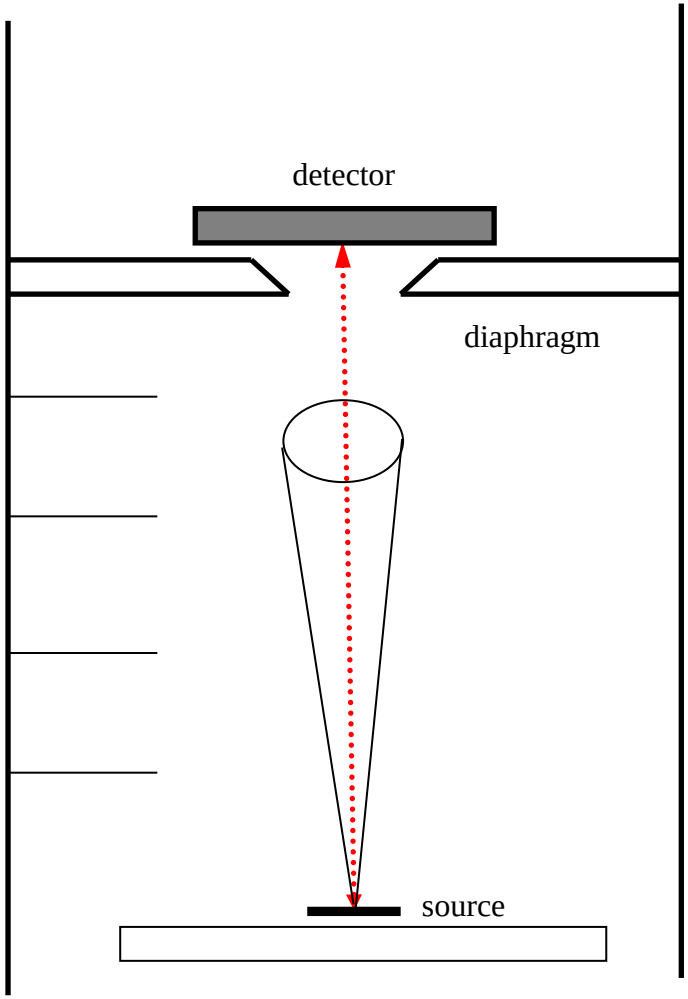
- not pass through diaphragm edge

- be detected with known efficiency

Method works with:

1. **α-particles** (MeV)
2. **X-rays** <10 keV
3. not β^- or γ-rays

Counting @ Defined Solid Angle



<u>Principle assumptions</u> :

- one particle is emitted per transition

- moves in random direction, along straight line

- is counted when reaching detector

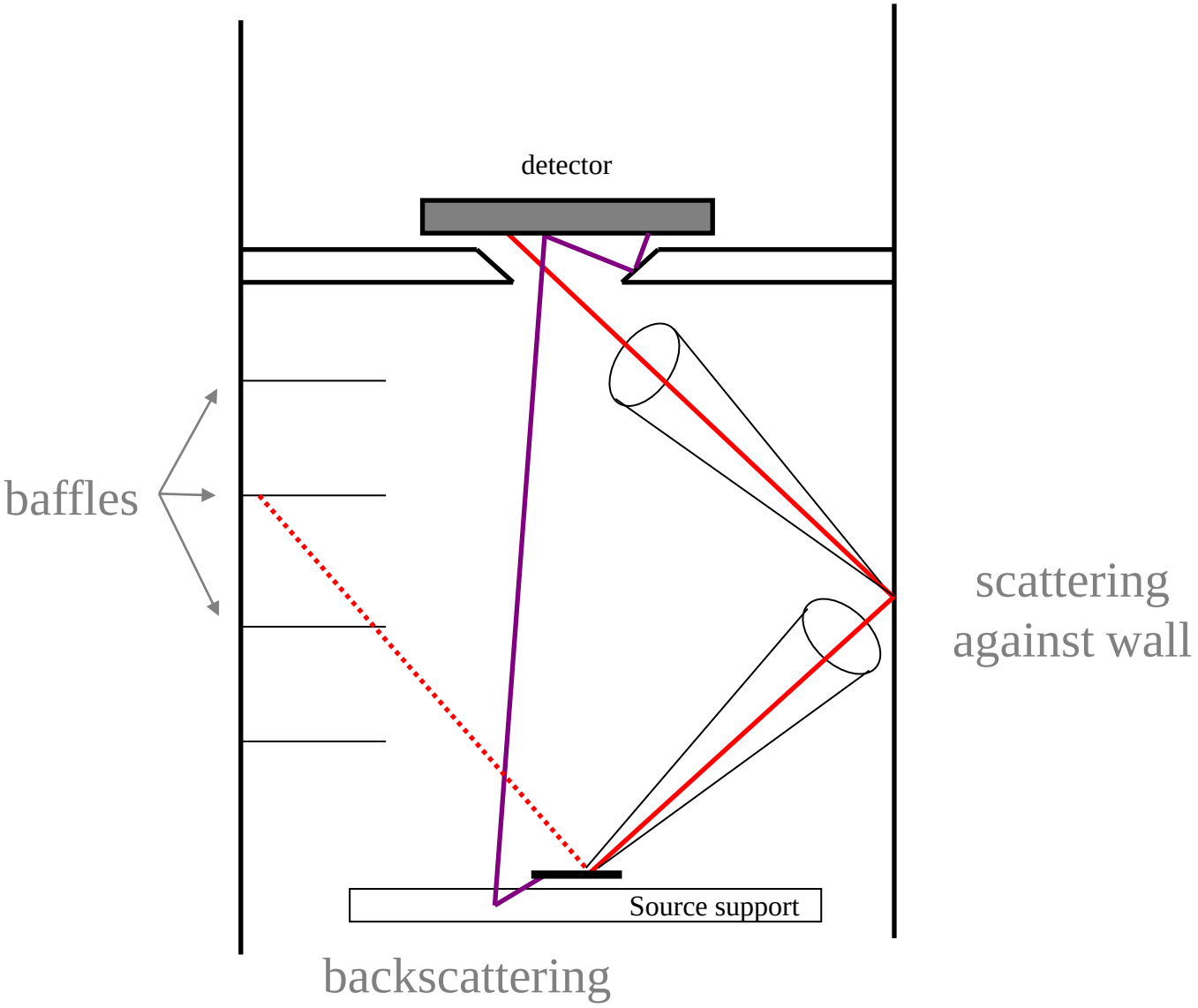
- geometry is extremely well defined

<u>Counting efficiency</u> :

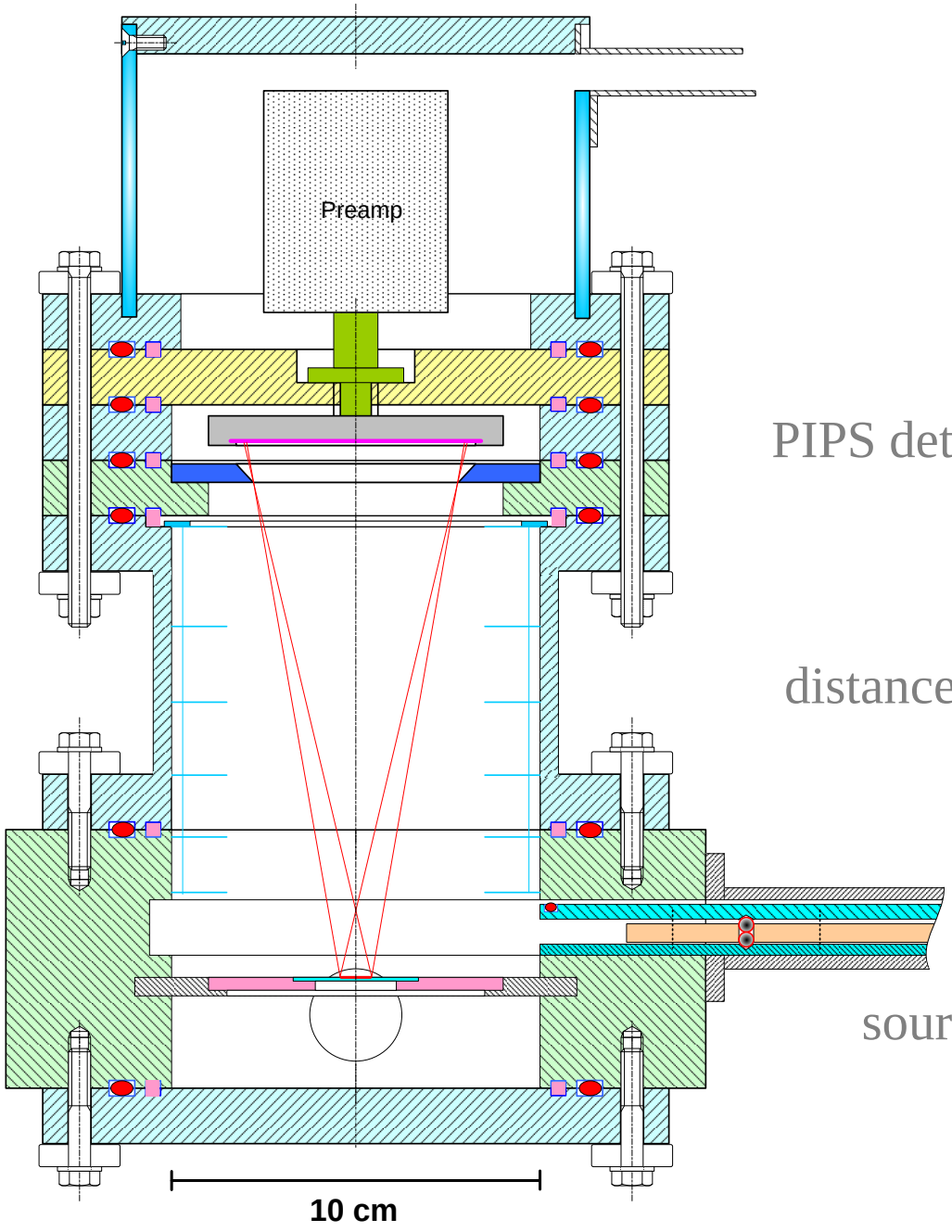
- detector efficiency = unity

- geometrical efficiency = solid angle / 4π

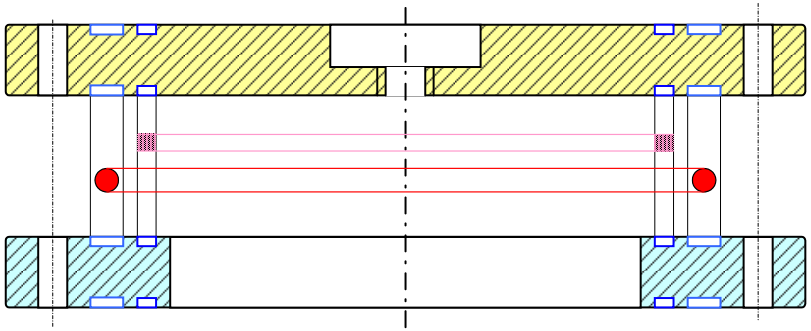
Scattering effects should be small

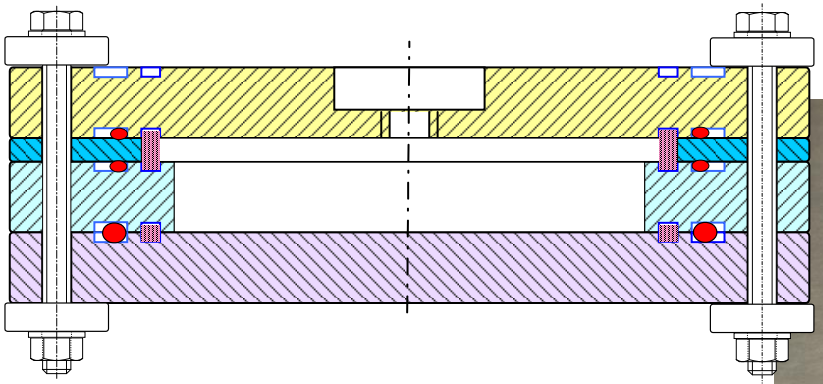


Alpha-particle counter with well-defined geometry



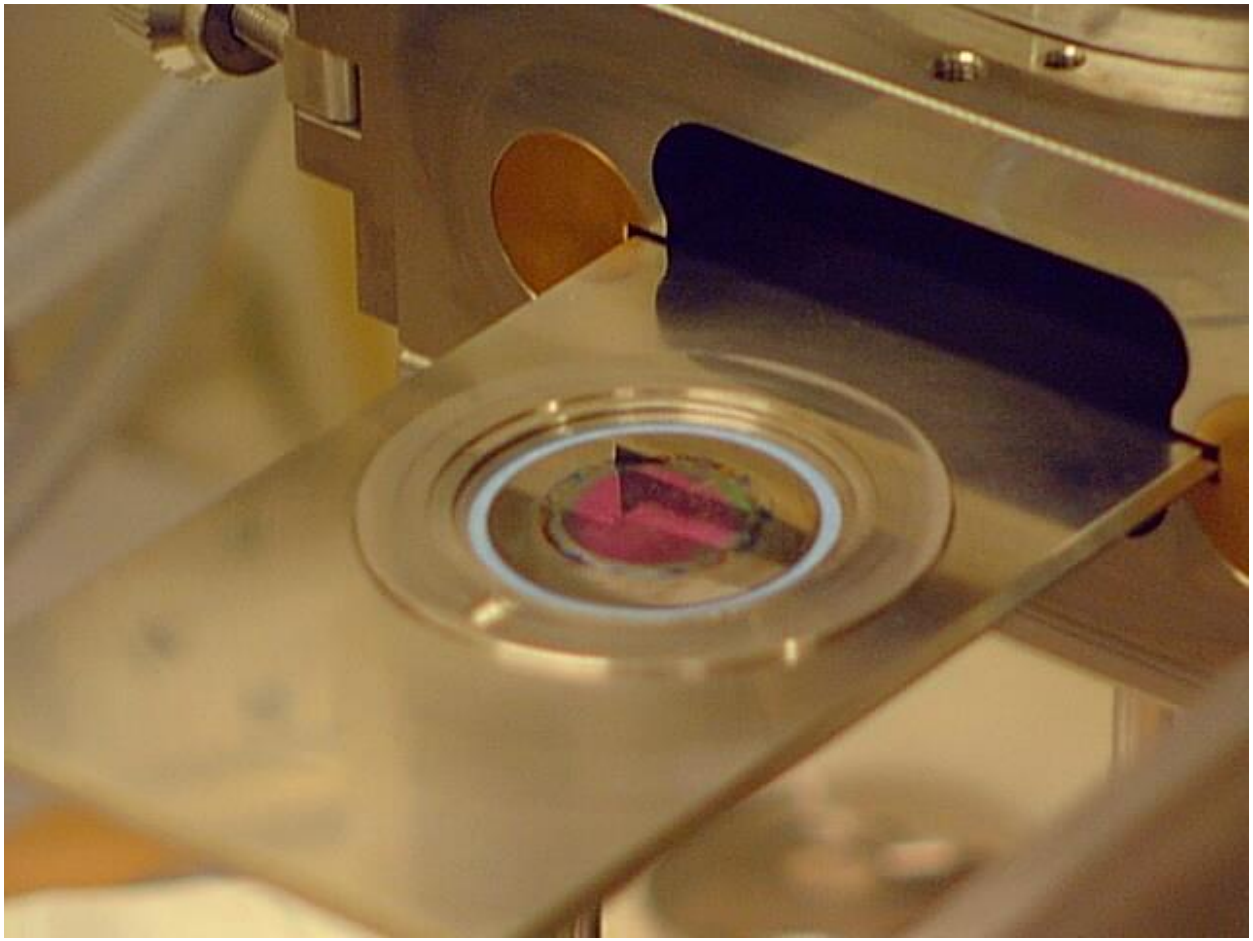
Reproducible geometric assembly



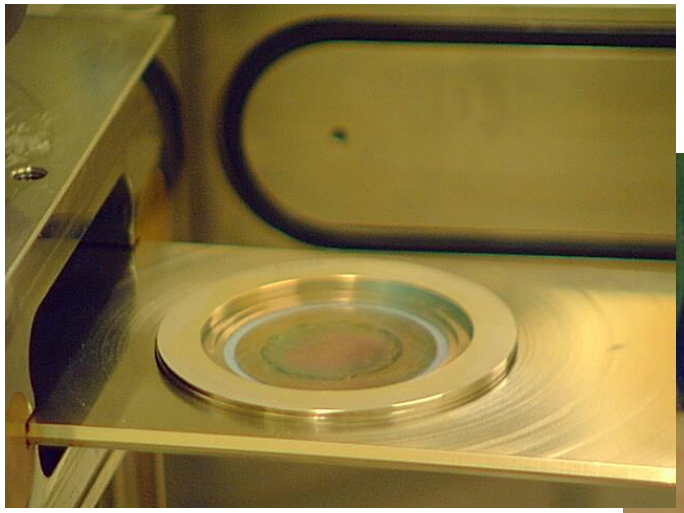


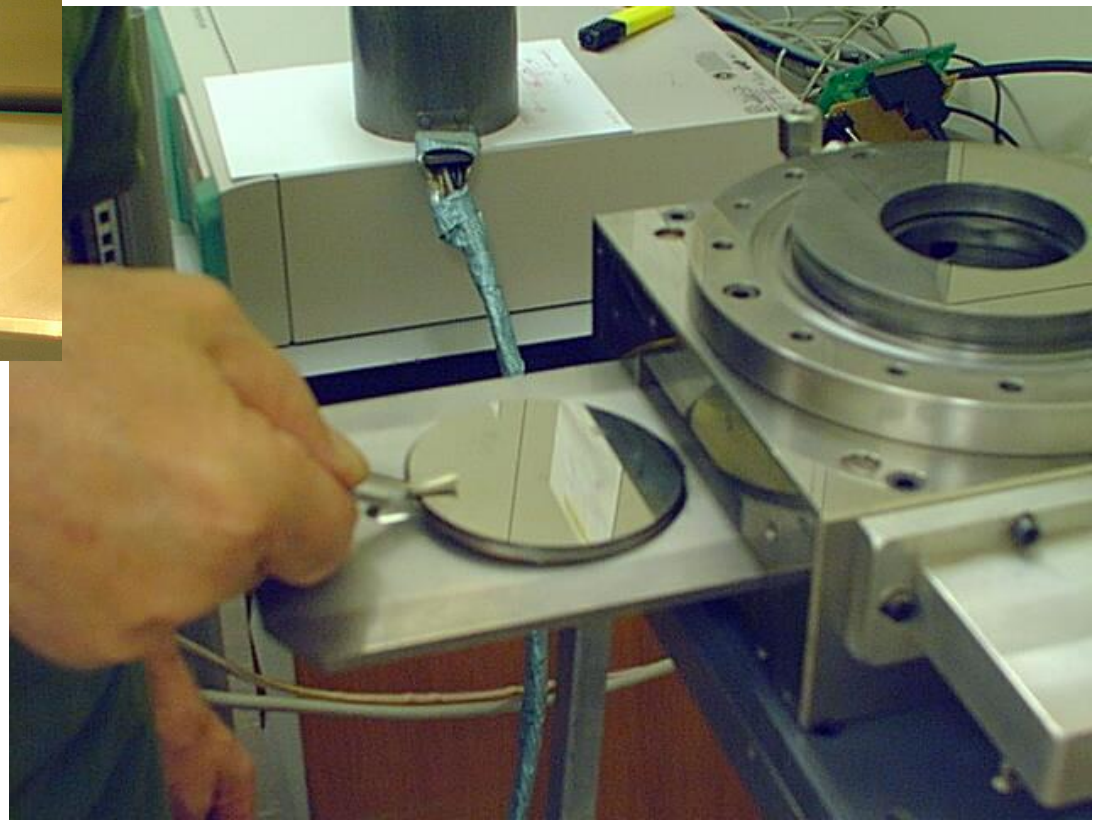
- **Coaxial flange system**
- **Planparallel faces**
- **Mechanical stability**

Example of how to measure source-detector distance without touching?



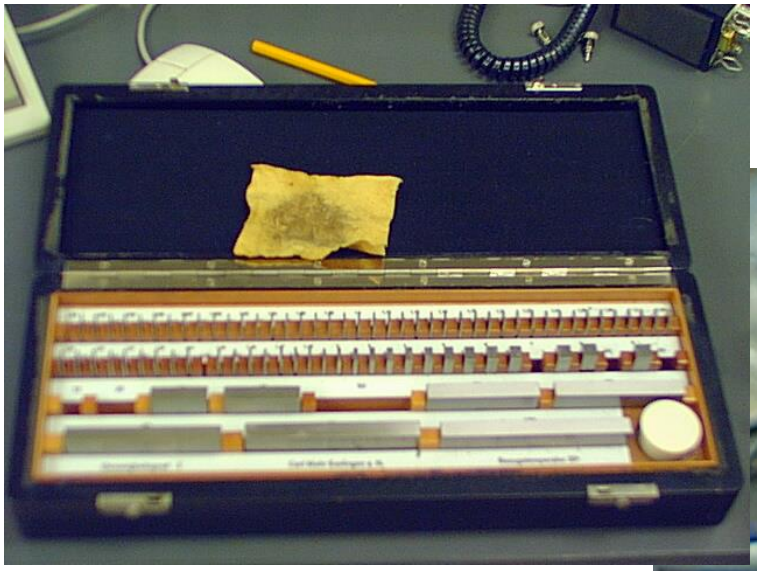
First : replace source by reference plate

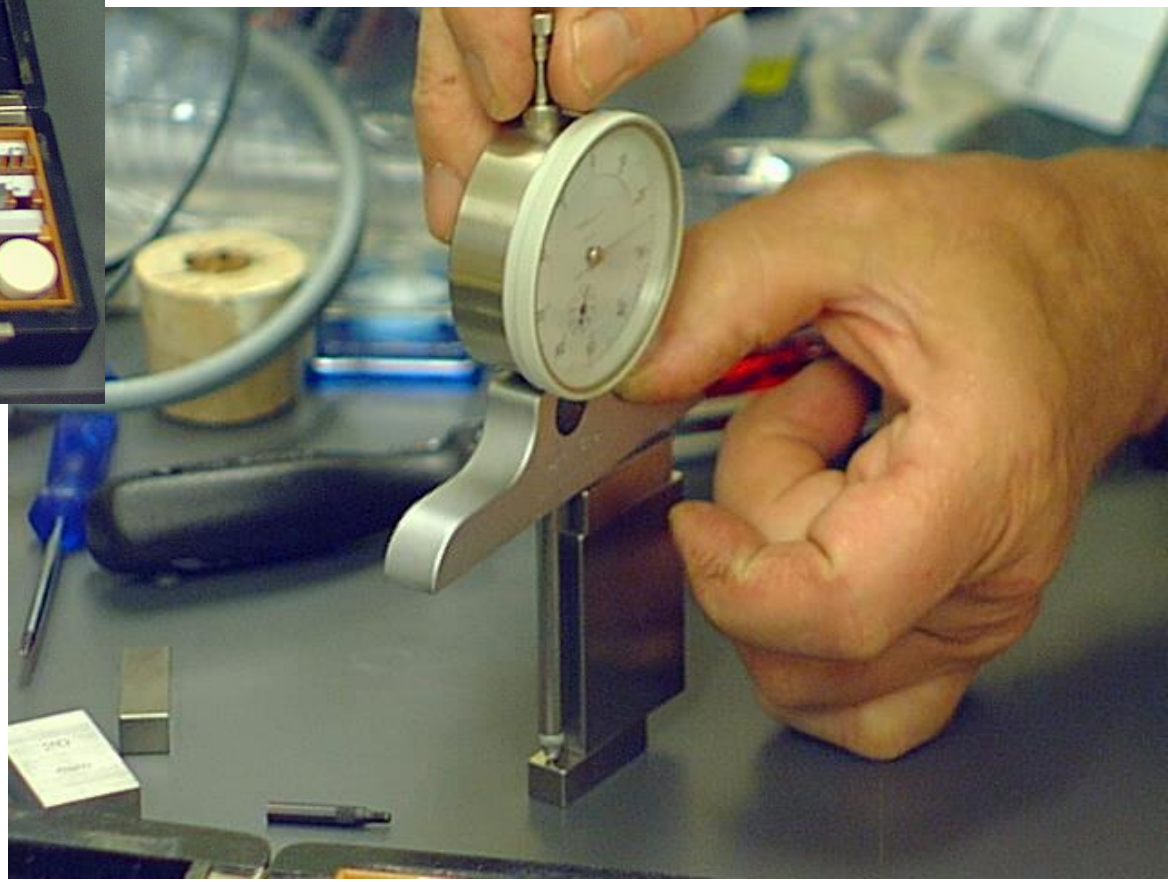




Then: measure distance reference plate - diaphragm

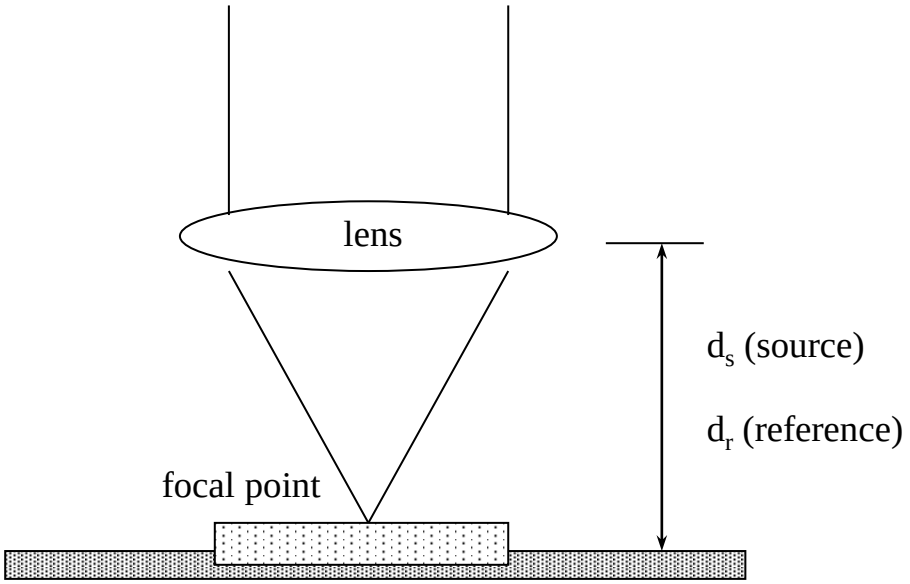
Compare with Gauge blocks as SI-traceable reference





Last : compare thickness source and reference plate

No touching,
just optical focussing on surface



tray with reference plate ‘R’ or source

Geometry factor

- **= the fraction of alpha particles emitted in the 'right' direction**

- **= the counting efficiency, if the detection efficiency is 100%**

- **= the ratio of the solid angle to 4π steradians**

Mathematical representations

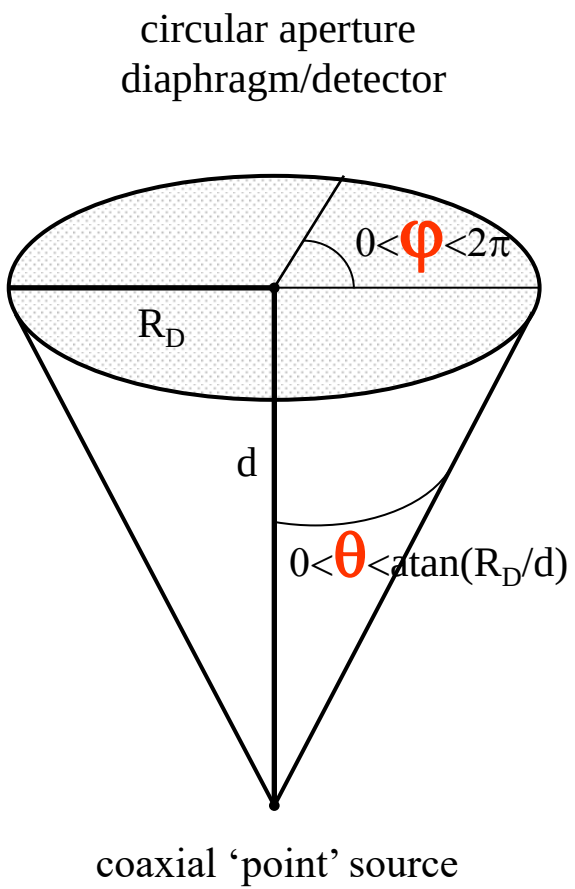
- Simplifications：
  - axial symmetry
  - point source, circular source
  - circular diaphragm (detector)
  - parallel planes
  - homogeneous activity distribution
- Reality：
  - inhomogeneous, off-axis, non-circular source
  - diaphragm edge has certain thickness

Point source on symmetry axis

- The only geometry with a simple solution for Ω

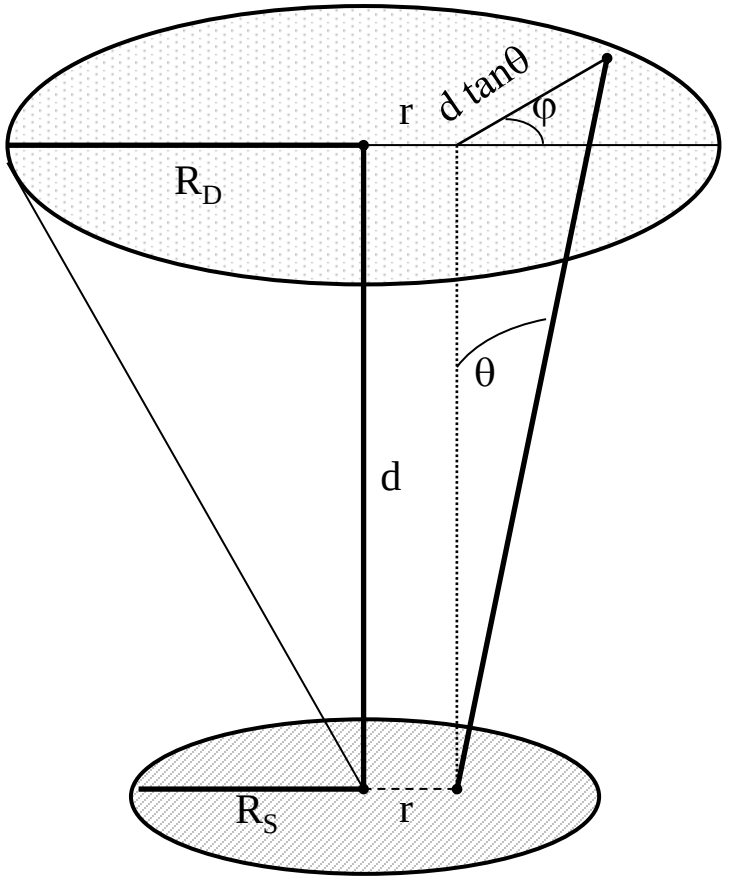
$$\Omega = 2\pi(1-\cos\theta)$$

$$\theta = \mathrm{a}\tan(\mathrm{R_D}/\mathrm{d})$$

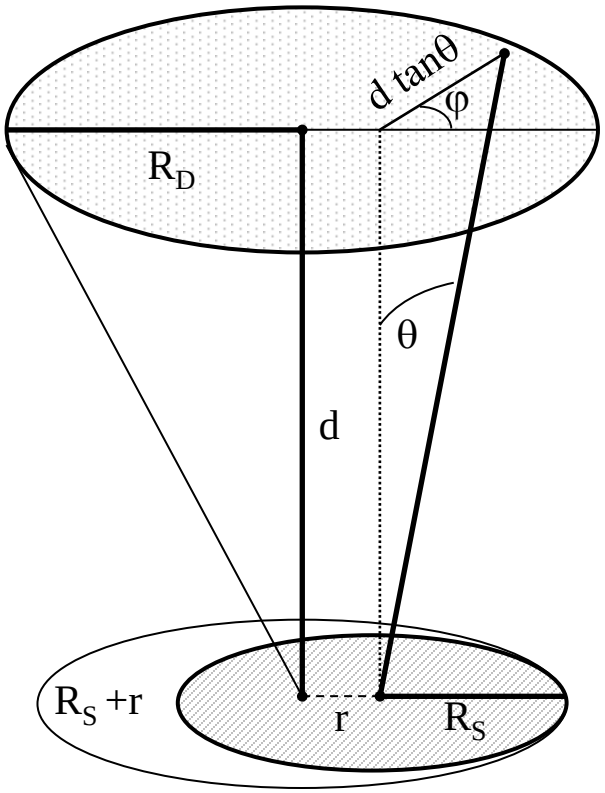


Coaxial, homogeneous, disk source

- Tables and Approximations
  Masket, Jaffey, etc.
- Numerical Integration
  Pommé et al., NIM A505
- Elliptic functions
  Tryka, Optics Com. 137
- Bessel functions
  Ruby, NIM A337
  Pommé, NIM A531

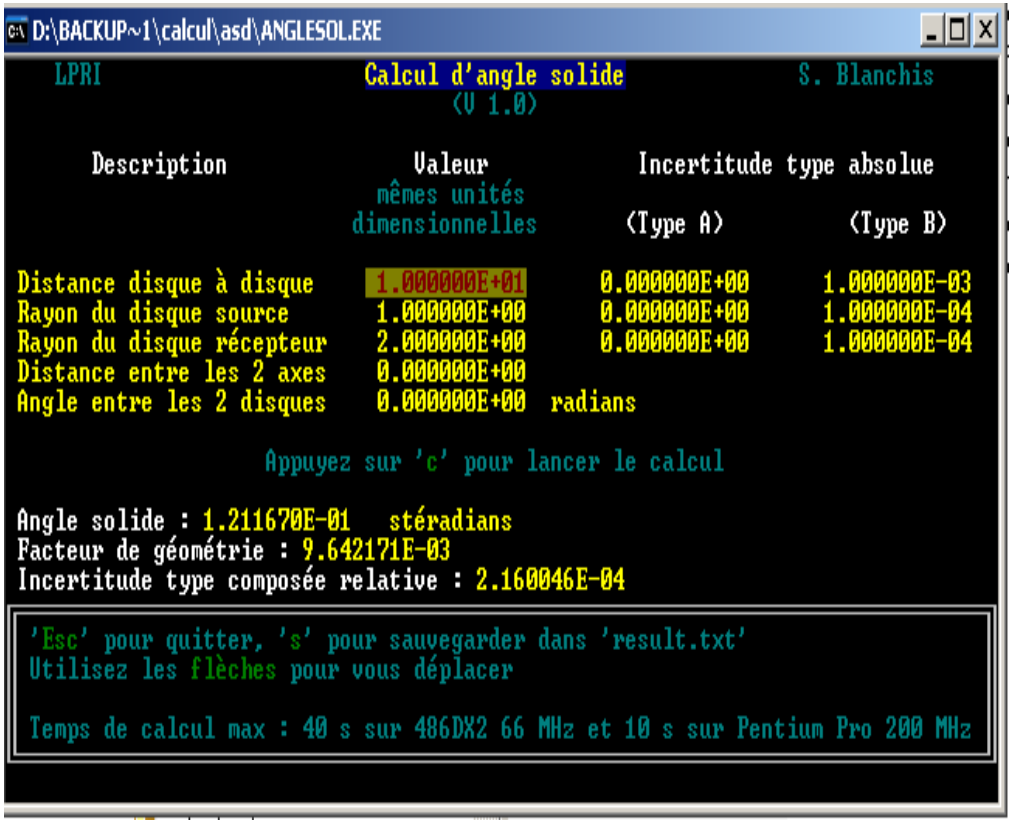


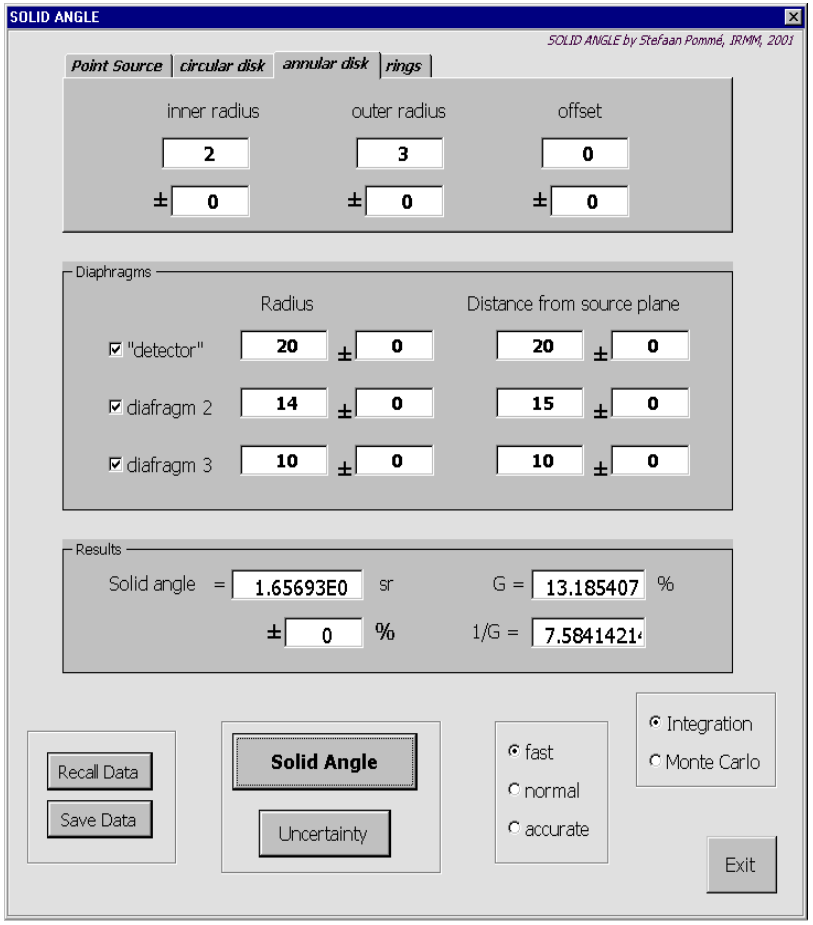
Disk or point source out-of-centre



- Numerical Integration
  Pommé et al., NIM A505
- Bessel functions
  Conway, NIM A562, A583
  Pommé et al., NIM A579

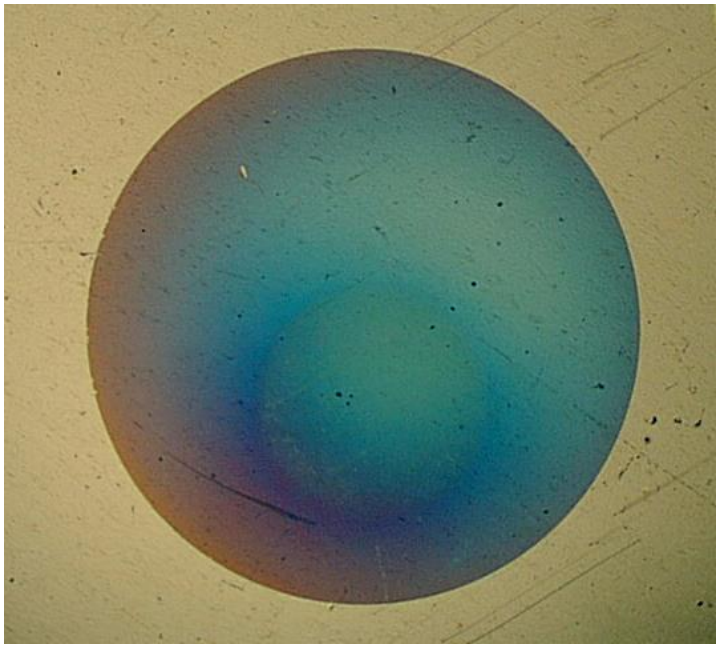
Software: ANGLESOL (LNHB) and SOLIDANGLE (JRC)



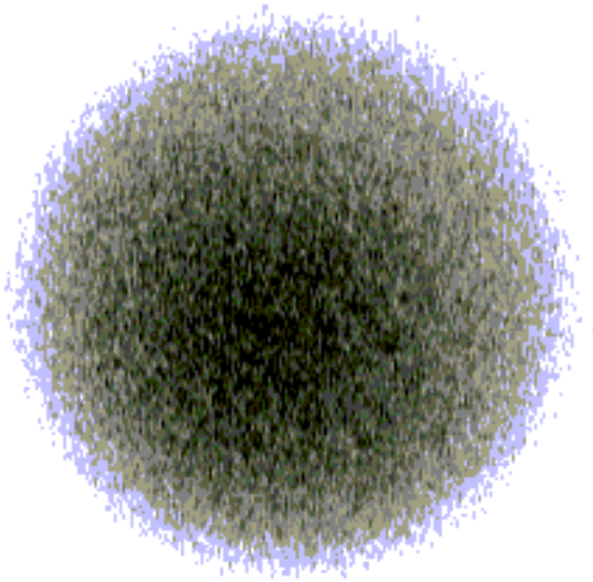


Vacuum evaporated source

- Visual



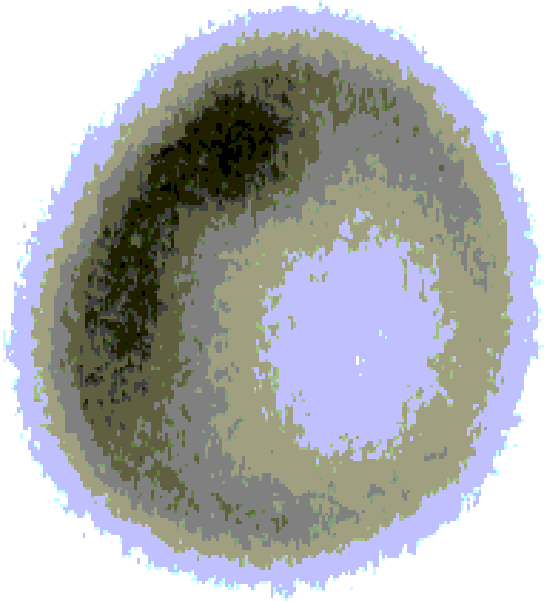
Autoradiograph



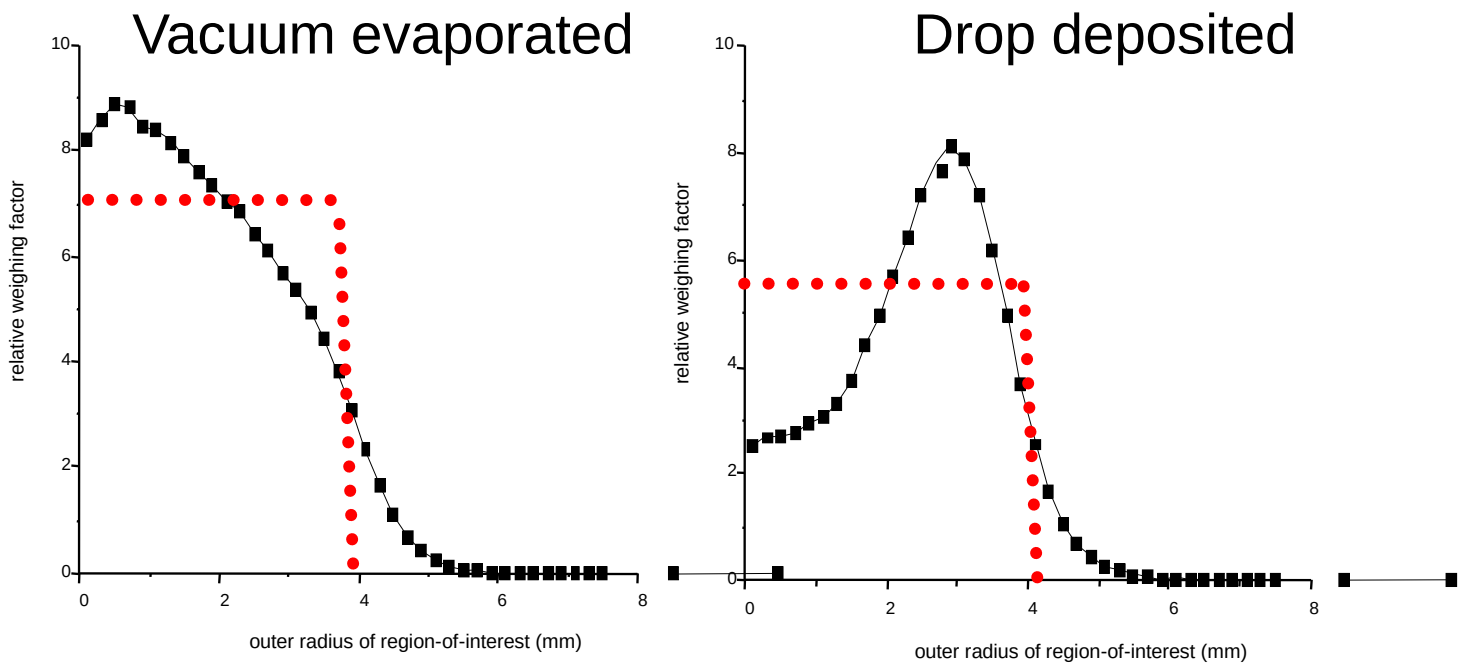
Drop deposited source

- Visual

Autoradiograph

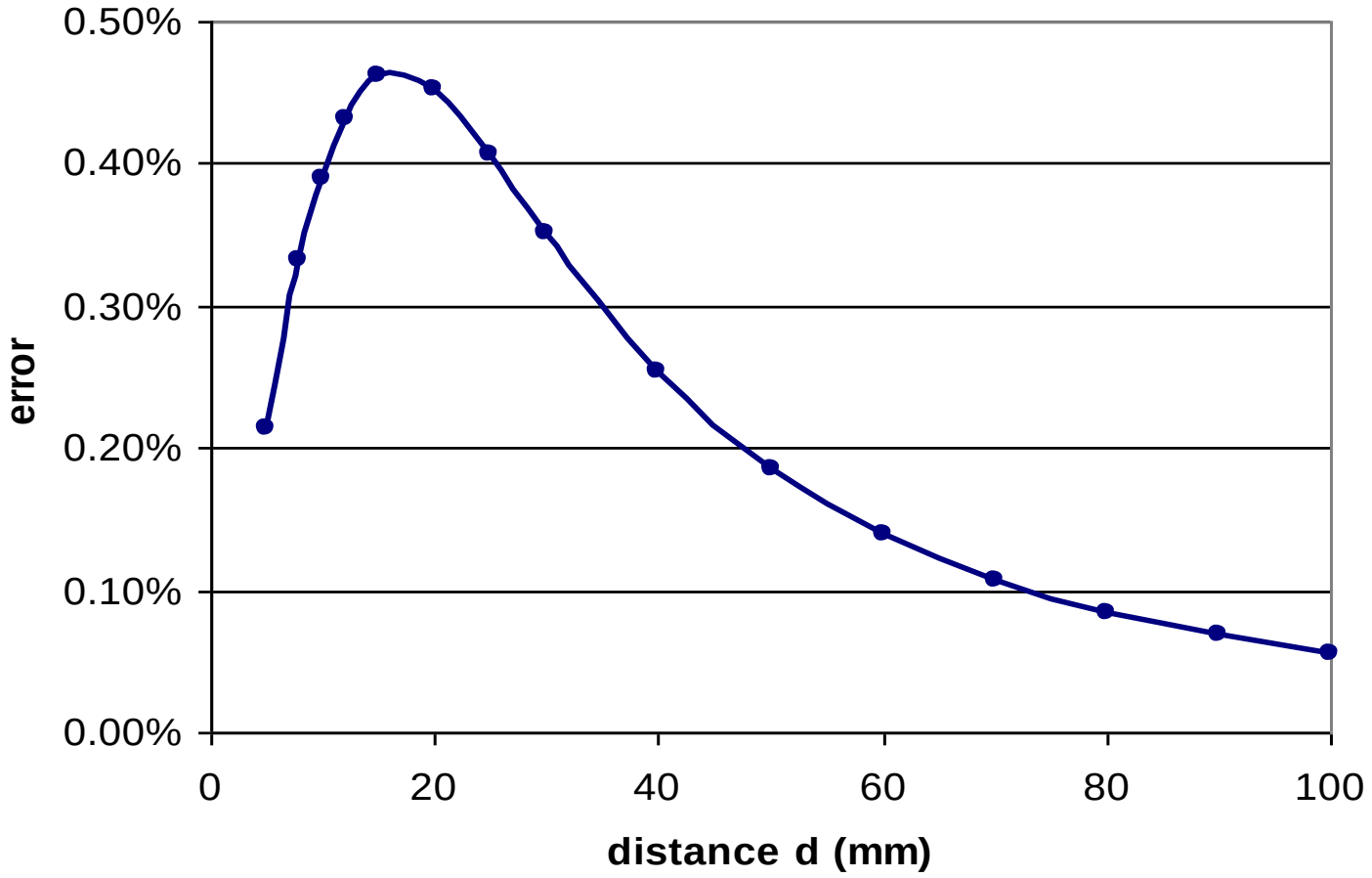


Activity distribution in sources

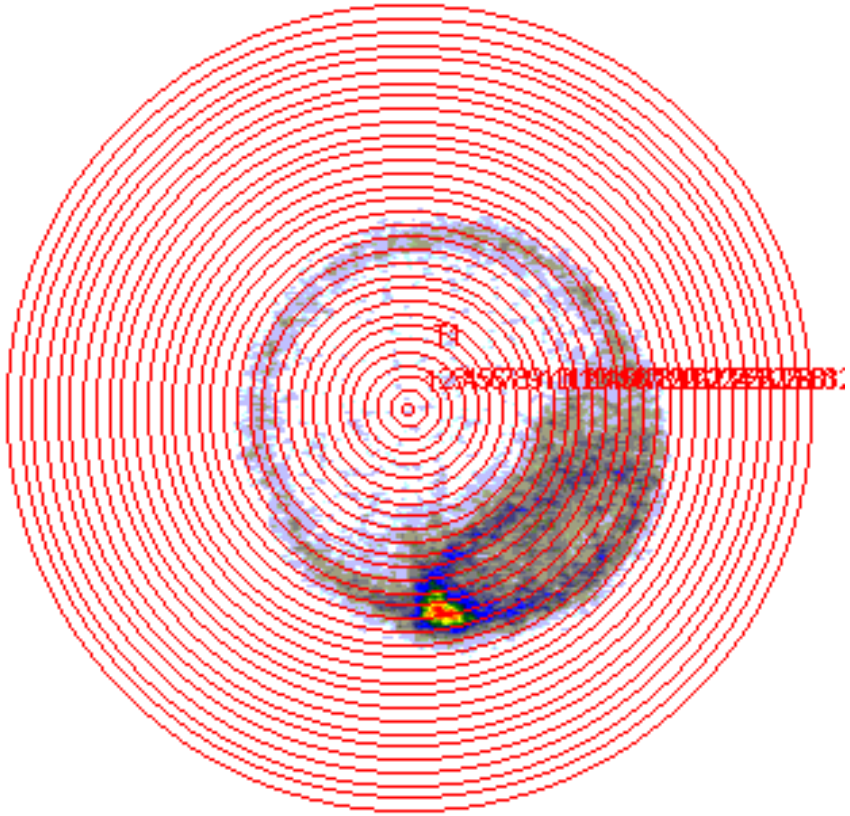


- ...not as homogeneous as you would like!

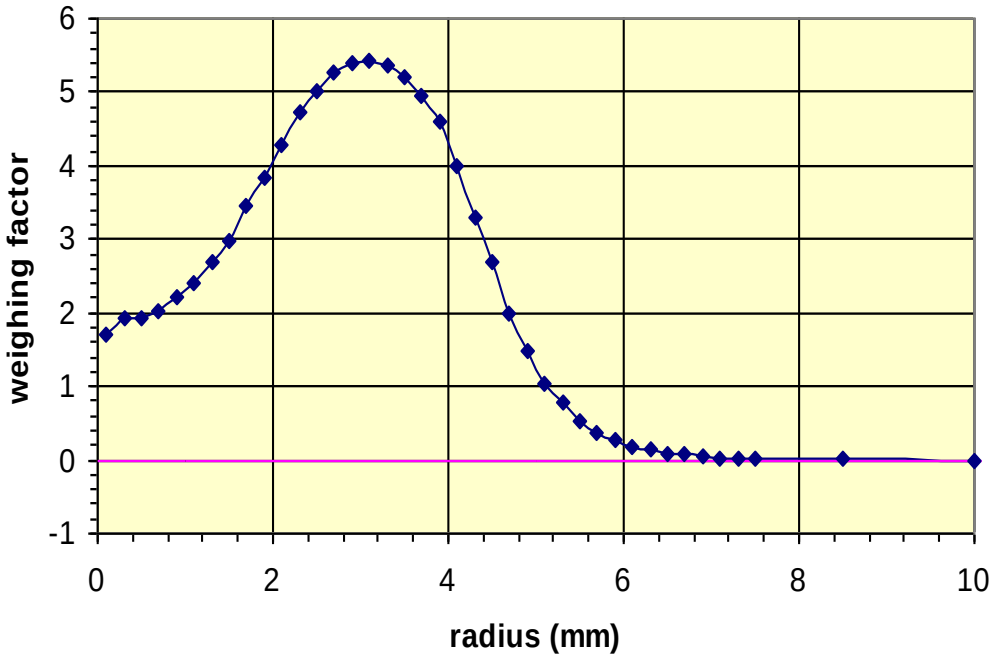
Error by misrepresenting source as a centred, homogeneous disk

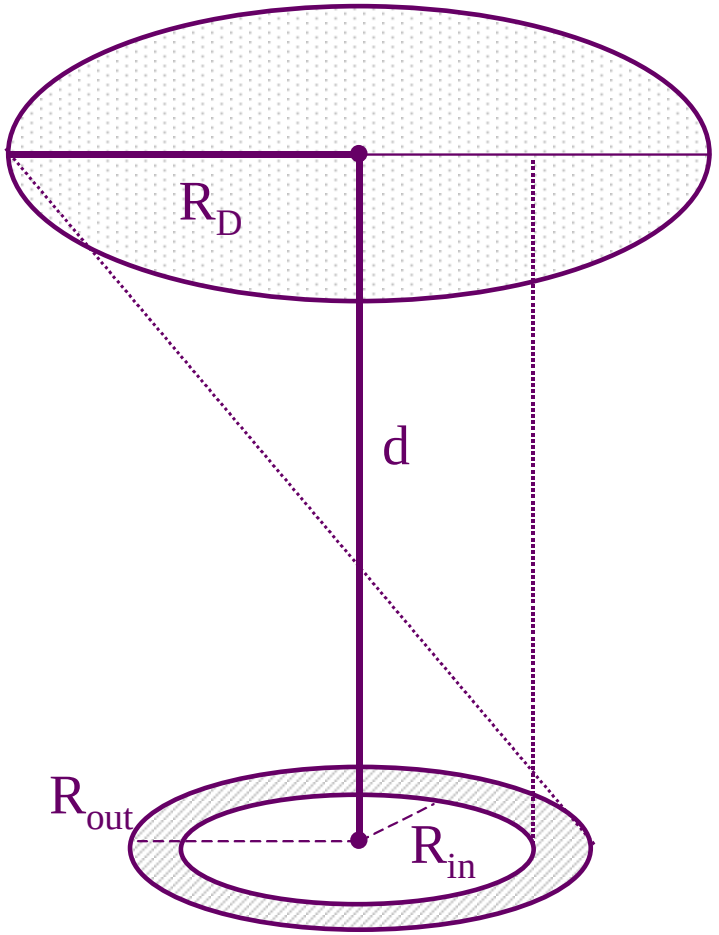


Subdivide source in concentric rings



Weighted sum of each ring's contribution to Ω





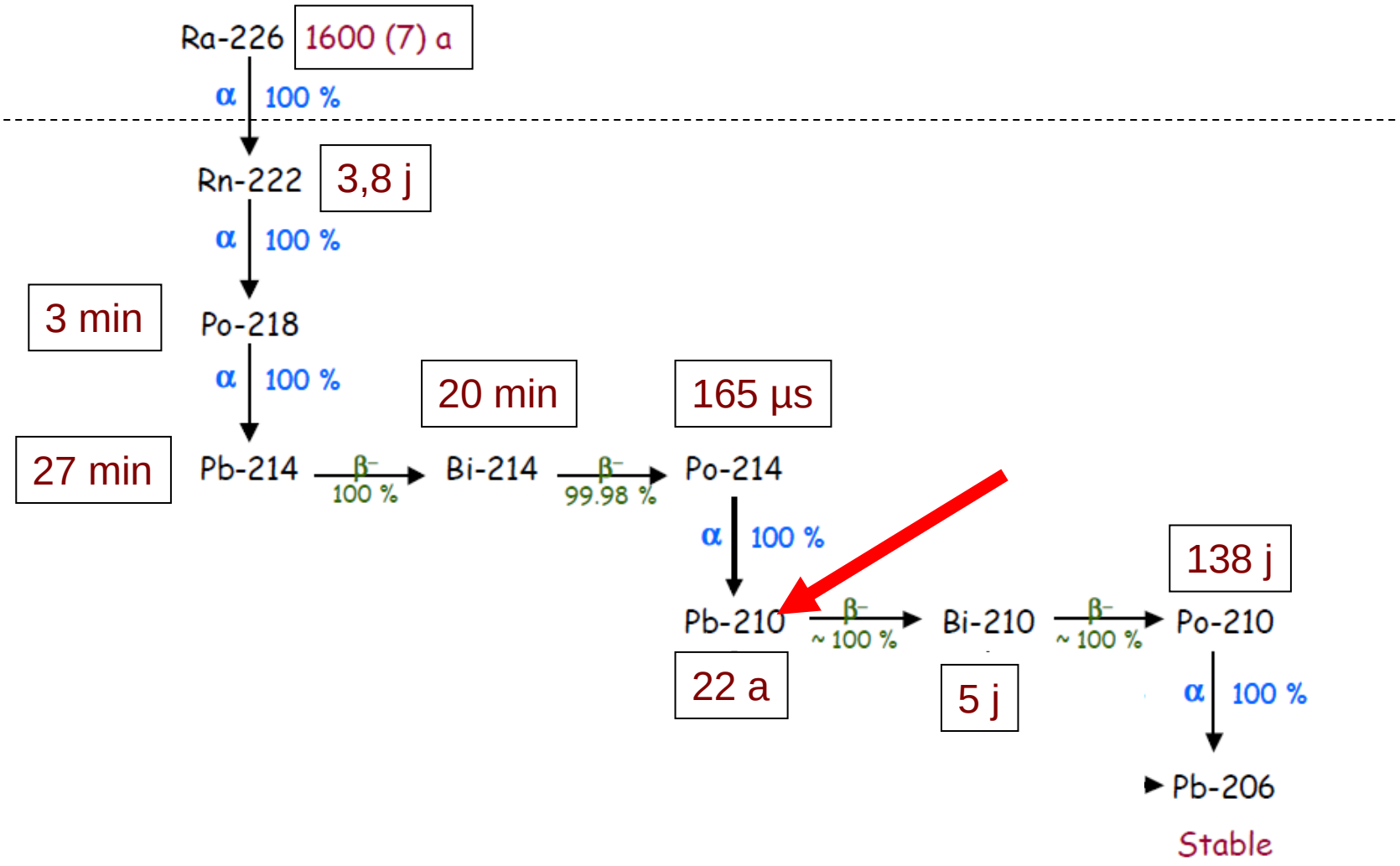
Primary Standardisation of activity

^{222}Rn standard

Various radon

- « **radon** » (^{222}Rn), half-life 3,8 d
  from ^{226}Ra (^{238}U natural decay chain)
- « **thoron** » (^{220}Rn), half-life 56 s
  from ^{224}Ra (^{232}Th natural decay chain)
- « **actinon** » (^{219}Rn), half-life 4 s
  from ^{227}Ac (^{235}U natural decay chain)

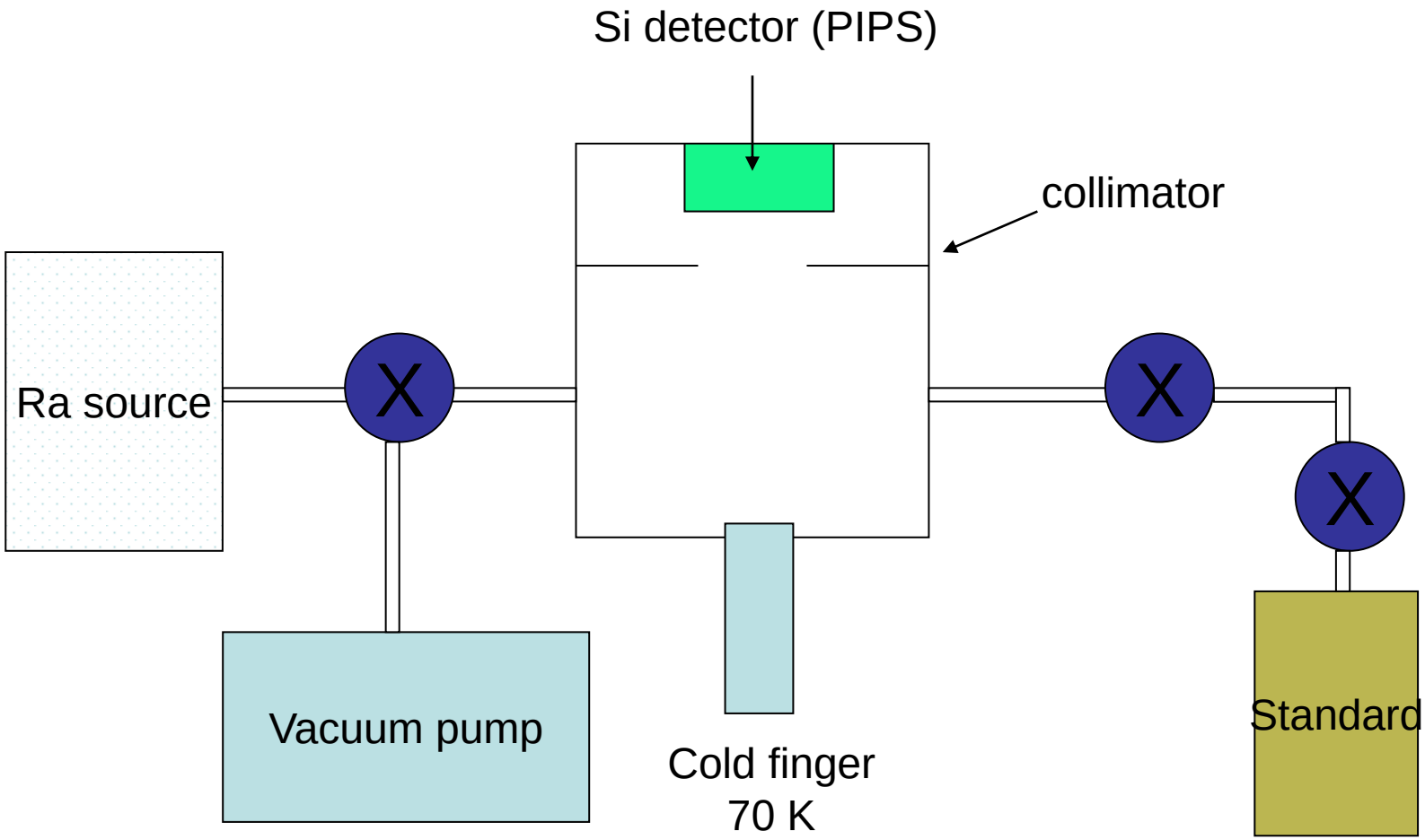
^{222}Rn simplified decay scheme



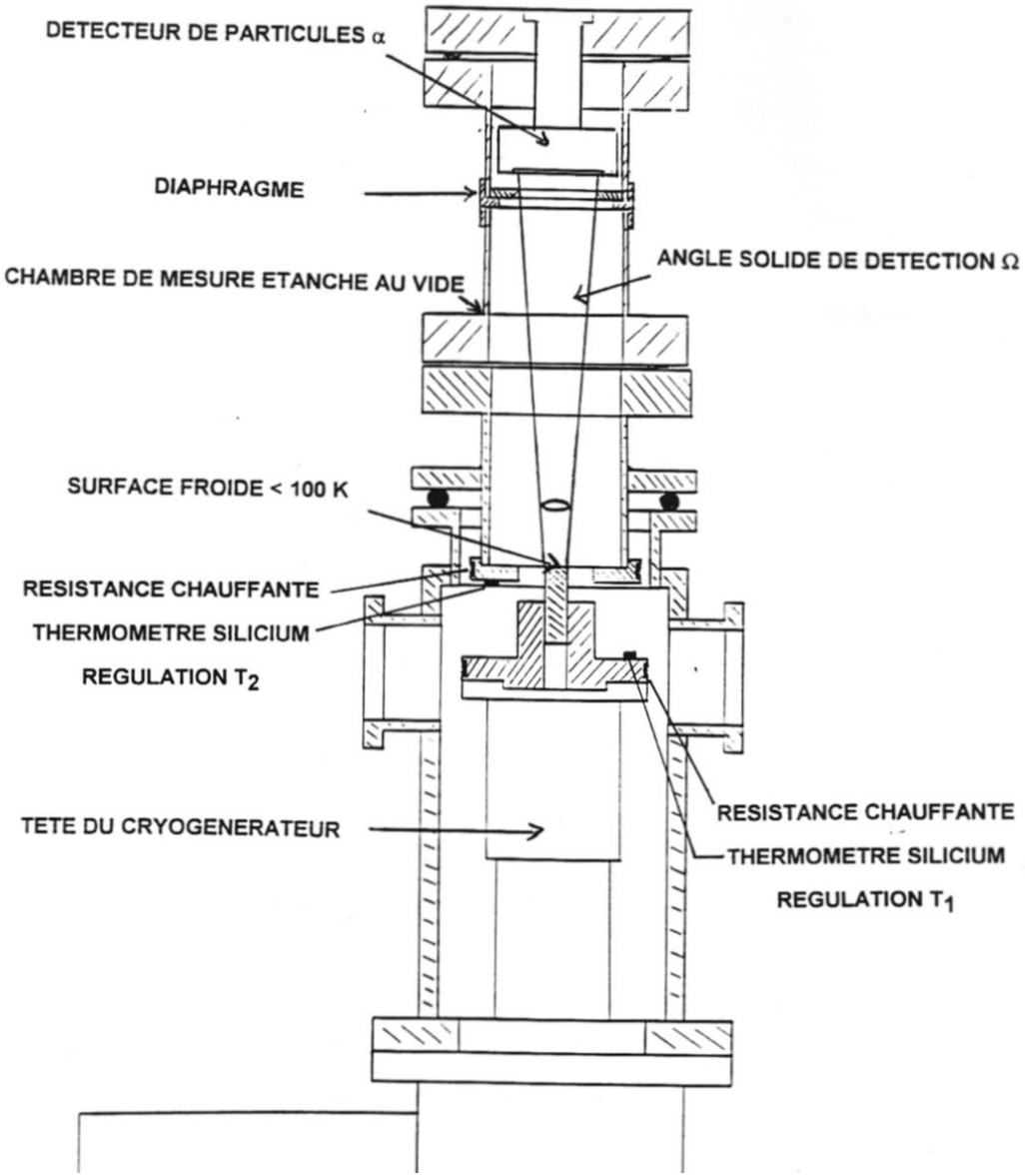
^{222}Rn standards

- Before 1995 : from ^{226}Ra with emanation coefficient
- In 1995, first cryogenic radon standard at LNHB (France)
- Development of the same instrument at IRA-METAS (Switzerland, PTB (Germany), KRIIS (Korea) and NIM (China)

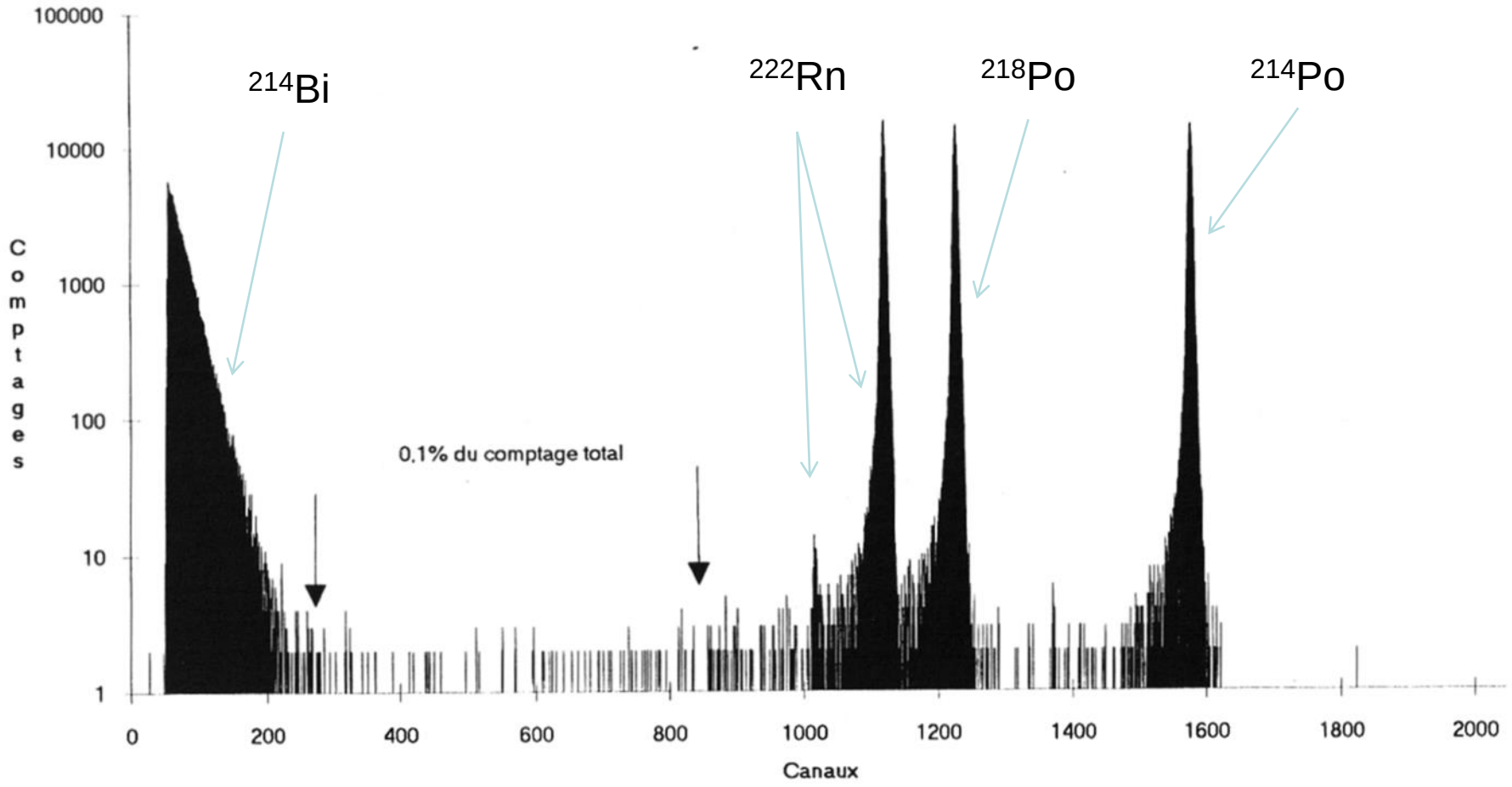
Measurement method



Measurement chamber



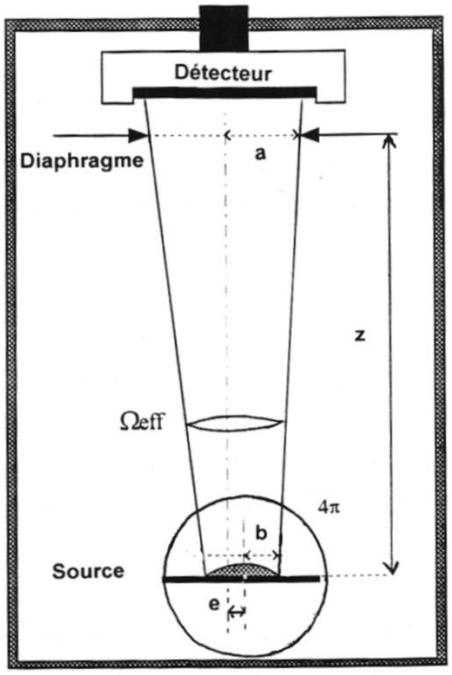
Alpha spectrum (at equilibrium)



Detection efficiency

- Intrinsic efficiency of the silicon detector = 1
  (1 α of 5 MeV creates 1,5 10^6 e^-/holes pairs)

- Geometry factor:



$$G = \frac{1}{2}\left[1 - \frac{z}{\sqrt{z^2 + a^2}}\right]$$

Activity $\longrightarrow A = \frac{n}{G}$ ← Counting rate

← Geometry factor

Uncertainty

$$\frac{u(A)}{A} = \left[\left(\frac{u(n)}{n} \right)^2 + \left(2(1-G)(1-2G) \sqrt{\left(\frac{u(a)}{a} \right)^2 + \left(\frac{u(z)}{z} \right)^2} \right)^2 \right]^{1/2}$$

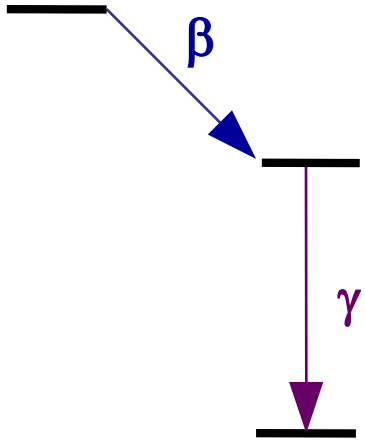
In practice 0.3 to 0.4 % relative standard uncertainty in the best conditions

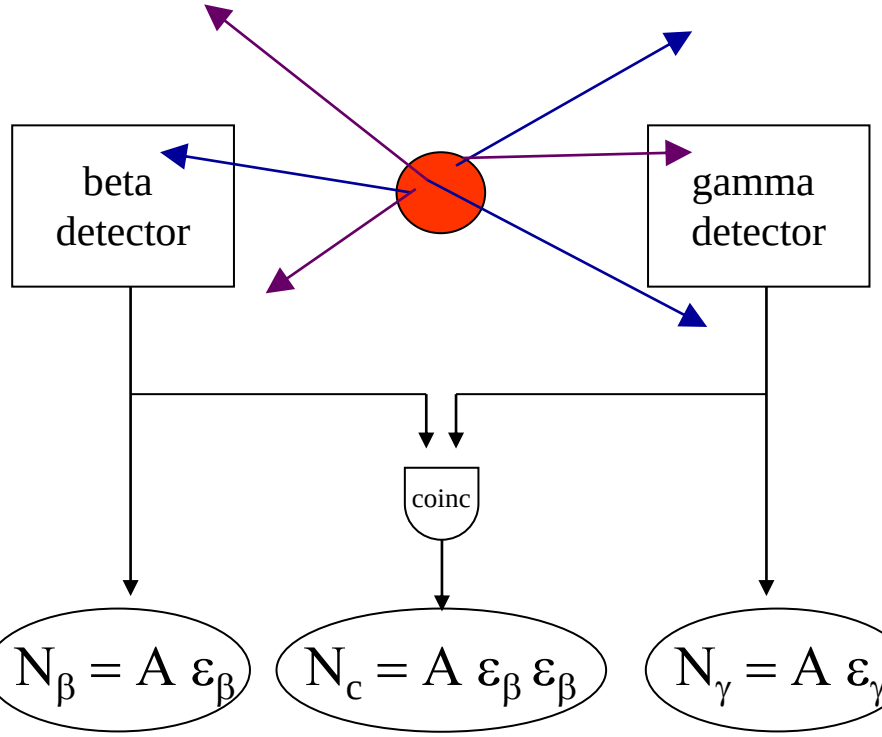
Primary Standardisation of activity

Coincidence Counting

Basics of Coincidence Counting

Consider simple decay scheme: one beta followed by one gamma.
Measure count rates in each detector along with "coincidence" rate.

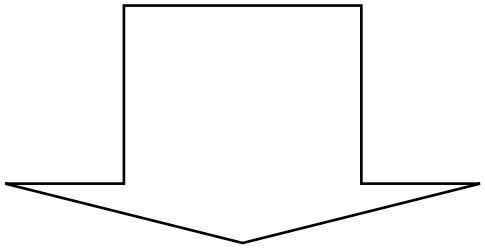




$$A = \frac{N_\beta N_\gamma}{N_c}$$

Three equations… three unknown

$N_\beta = A\,\varepsilon_\beta$ $N_c = A\,\varepsilon_\beta\,\varepsilon_\beta$ $N_\gamma = A\,\varepsilon_\gamma$



$$\varepsilon_\beta = \frac{N_c}{N_\gamma} \qquad A = \frac{N_\beta N_\gamma}{N_c} \qquad \varepsilon_\gamma = \frac{N_c}{N_\beta}$$

Only true under certain conditions…

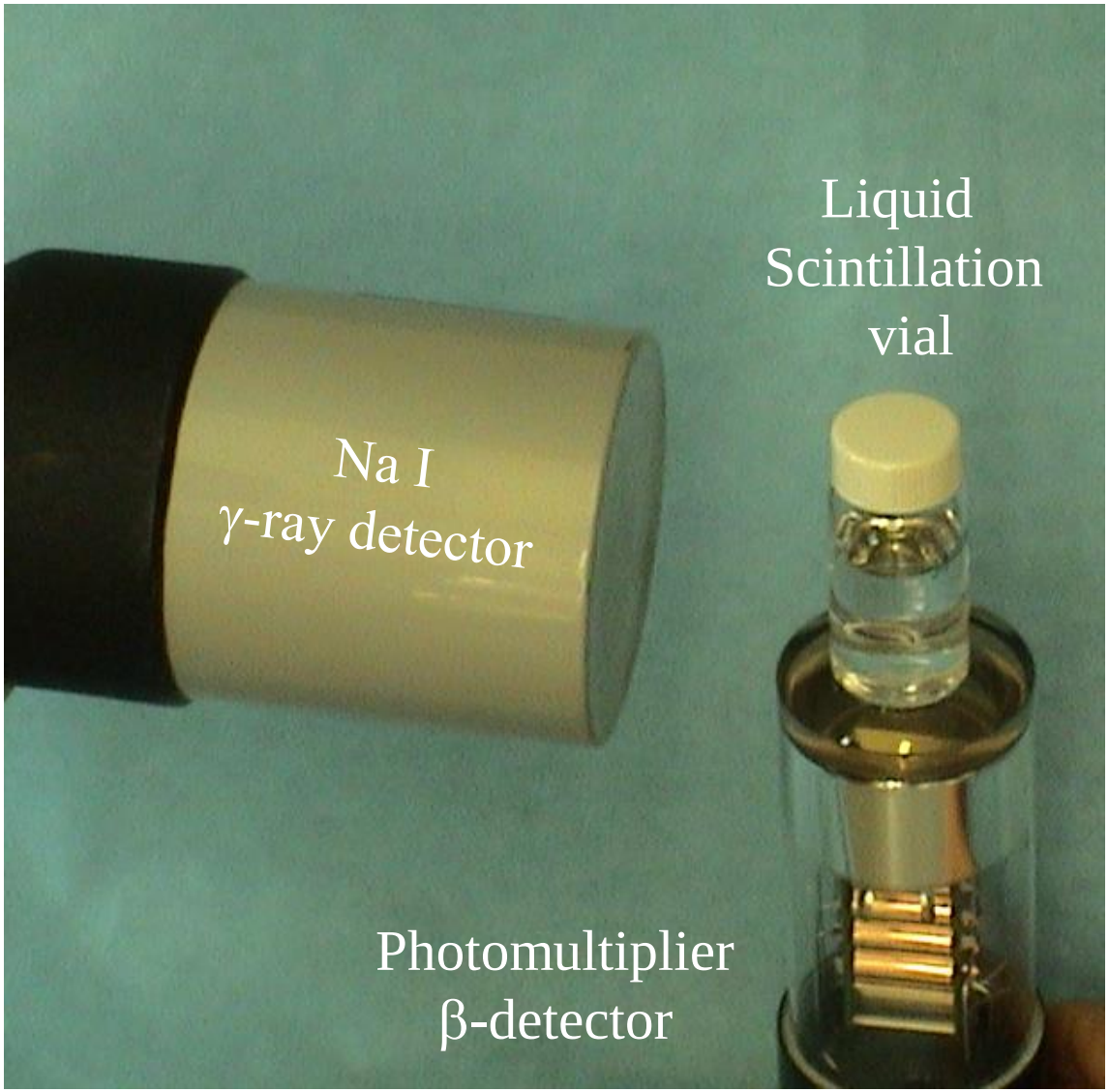
Some requirements

- Detectors sensitive to one type of radiation only
  - no gamma-ray detection in beta-counter
  - also no pickup of electronic noise !!
- ε_β and ε_γ must be independent and constant
  - no directional correlation between β and γ
    $\rightarrow$ use 4π beta detector
  - at least one of the efficiencies should be the same in all parts of the source
- No coincidences should be lost
  - coincidence window wide enough to avoid loss through time-jitter between β and γ signals
  - compensate for accidental coincidences and dead time

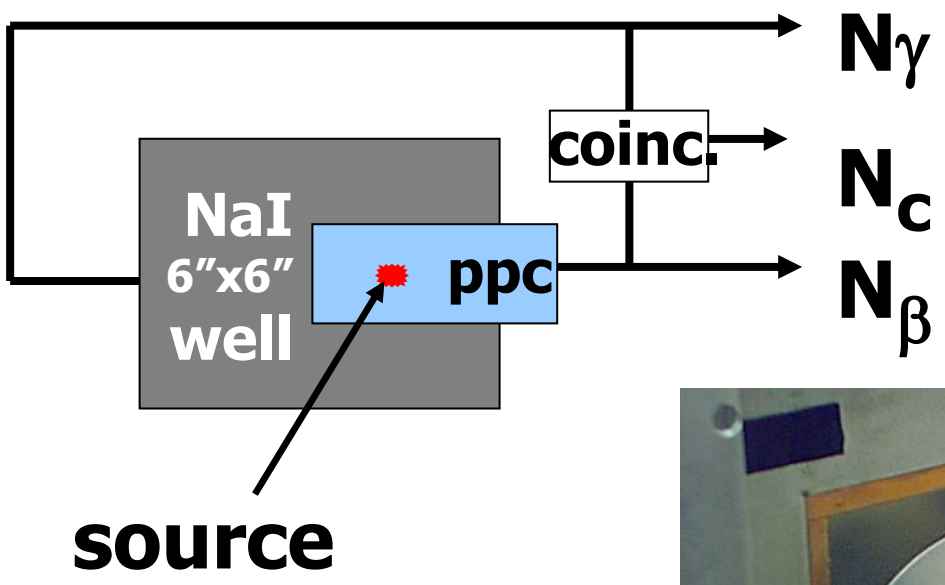
Suitable beta detectors

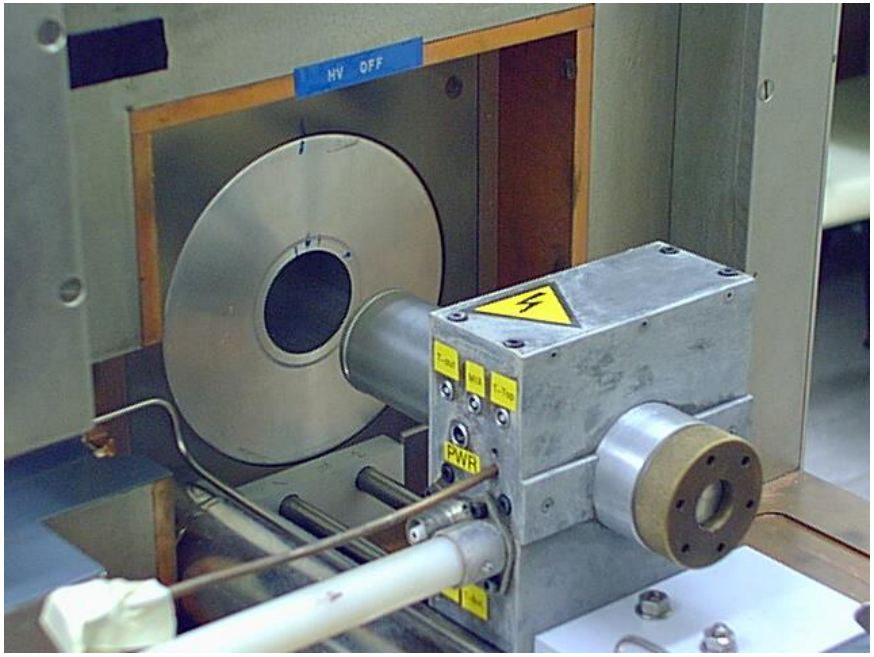
- Need 4π geometry
  - high ε reduces uncertainty of extrapolation
  - if $< 4\pi$, will be error in activity if > 1 beta branch
  - no directional correlation
  - no scattering/absorption correction outside source(cf. 2π or defined solid angle)
- Proportional counters with thin sources
  - high gas gains & low dead times
  - almost all charged particles escaping source are counted
  - requires suitable chemical form for thin stable solid source
- Liquid scintillation counters
  - dead times usually larger than PC (after-pulses …)
  - higher $\varepsilon_{\beta\gamma}$ interaction than PC (by ~ 10x)
  - requires suitable chemical form to be stable, compatible with scintillant

A rudimentary set-up



A performant set-up



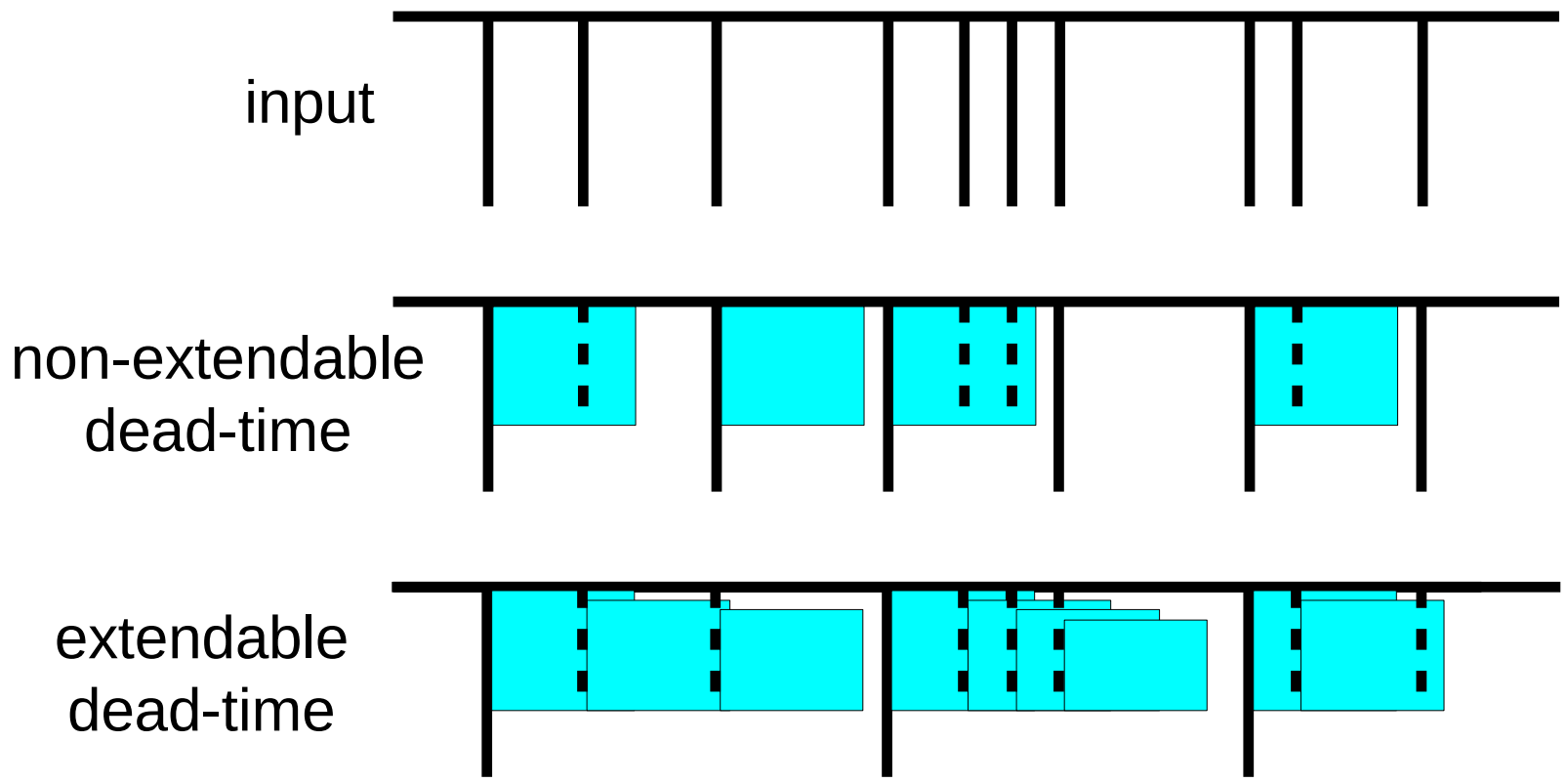


PPC = Pressurised
Proportional Counter

So far the good news, but how about…

- dead time in both counters and their combined effect in the coincidences?
- extended, non-uniform sources?
- sensitivity of beta detector for photons and conversion electrons?
- sensitivity of γ-ray detector for annihilation photons created after β^+ decay?
- complex decay schemes with multiple branches?
- pure beta emitters?

Add artificial dead time



advantage: counters now have a known **type** and **length** of dead time
=> one can apply dedicated dead time corrections formulas

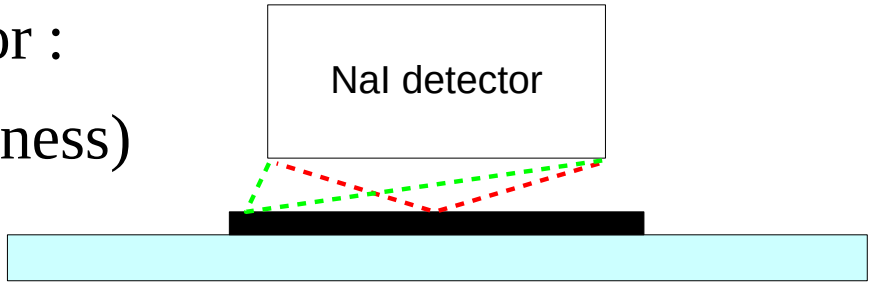
Extended sources

Uniformity requirements

Different parts of source may give rise to different efficiencies in a detector :

a) non-uniform source (e.g. thickness)

b) extended source



Let f_i = fraction with efficiencies $\varepsilon_{\beta i}$ and $\varepsilon_{\gamma i}$, where $\sum f_i = 1$.

$$\left.\begin{aligned} N_\beta &= A \sum f_i\, \varepsilon_{\beta i} \\ N_\gamma &= A \sum f_i\, \varepsilon_{\gamma i} \\ N_c &= A \sum f_i\, \varepsilon_{\beta i}\, \varepsilon_{\gamma i} \end{aligned}\right\}$$

$$\frac{N_\beta\, N_\gamma}{N_c} = A \cdot \frac{\sum f_i\, \varepsilon_{\beta i} \cdot \sum f_i\, \varepsilon_{\gamma i}}{\sum f_i\, \varepsilon_{\beta i}\, \varepsilon_{\gamma i}} \quad \neq \quad A$$

Possible solutions

Possibility 1: make sure that the **beta** efficiency is **constant**

$$\text{all } \varepsilon_{\beta i} = \varepsilon_{\beta} \quad \Longrightarrow \quad \frac{N_\beta N_\gamma}{N_c} = A \cdot \frac{\varepsilon_\beta \sum f_i \cdot \sum f_i \varepsilon_{\gamma i}}{\varepsilon_\beta \sum f_i \varepsilon_{\gamma i}} \equiv A$$

e.g. 4π geometry

Possibility 2: make sure that the **gamma** efficiency is **constant**

$$\text{all } \varepsilon_{\gamma i} = \varepsilon_{\gamma} \quad \Longrightarrow \quad \frac{N_\beta N_\gamma}{N_c} = A \cdot \frac{\sum f_i \varepsilon_{\beta i} \cdot \varepsilon_\gamma \sum f_i}{\varepsilon_\gamma \sum f_i \varepsilon_{\beta i}} \equiv A$$

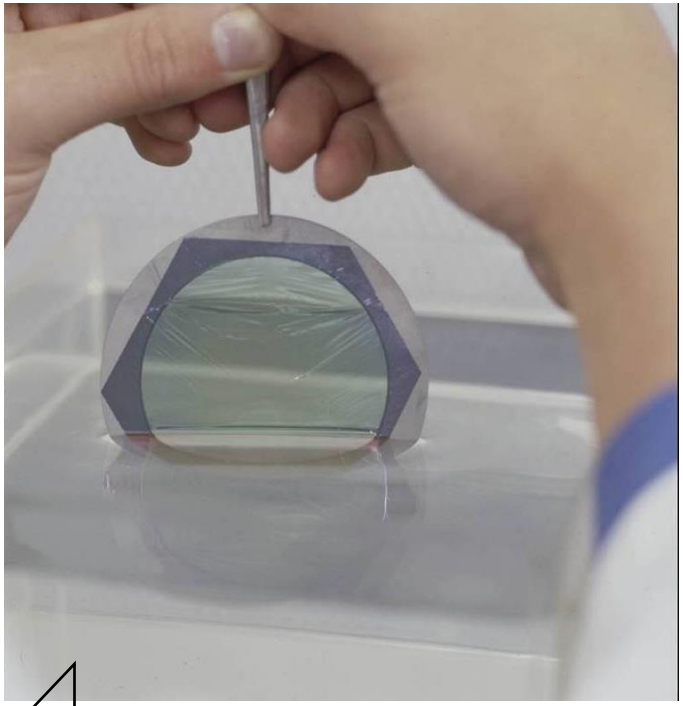
e.g. well detector

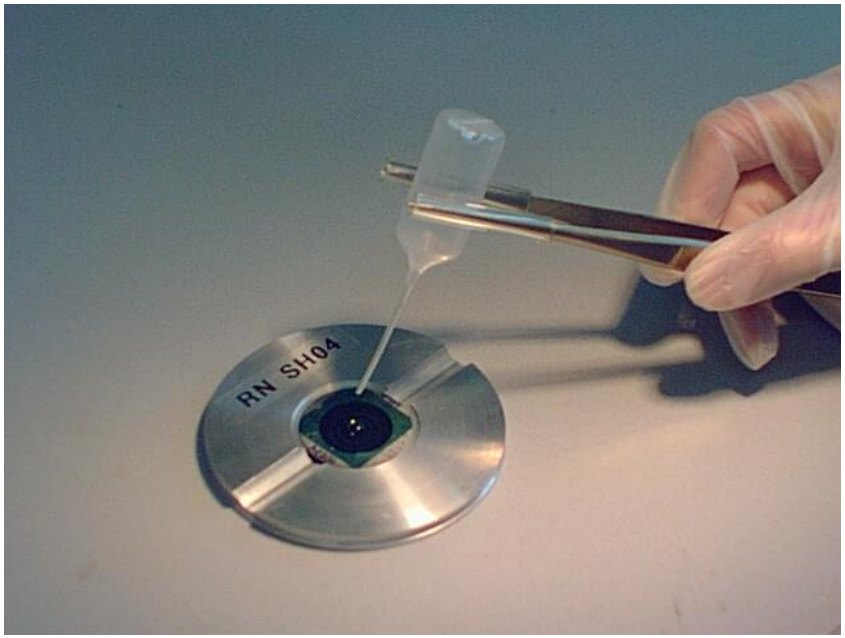
Possibility 3: use **thin** and **small** sources

… and compare several sources of different mass

Typical sources

substrate of ultra-thin
gold-coated VYNS polymer





source drop deposition
quantified by weighing

beta detector sensitive to γ-rays and conversion electrons

$$N_\beta = A\left[\varepsilon_\beta + (1-\varepsilon_\beta)\left(\frac{\varepsilon_{\beta\gamma} + \alpha\varepsilon_{ce}}{1+\alpha}\right)\right]$$

$$N_\gamma = A\frac{\varepsilon_\gamma}{1+\alpha}$$

$$N_c = A\left[\frac{\varepsilon_\beta\varepsilon_\gamma}{1+\alpha} - (1-\varepsilon_\beta)\varepsilon_c\right]$$

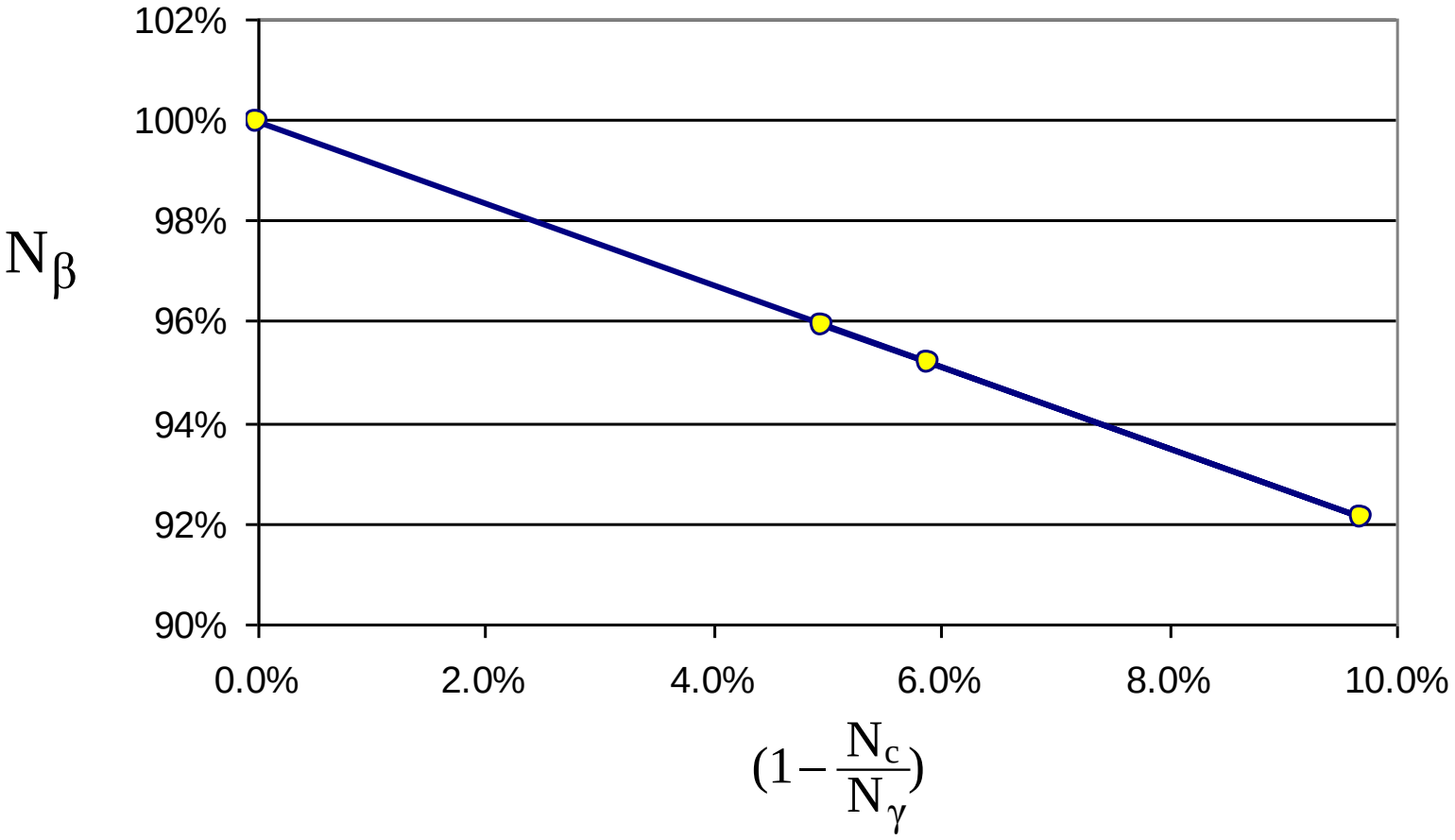
α= conversion factor

ε_β = interaction probability for γ-ray in β-detector

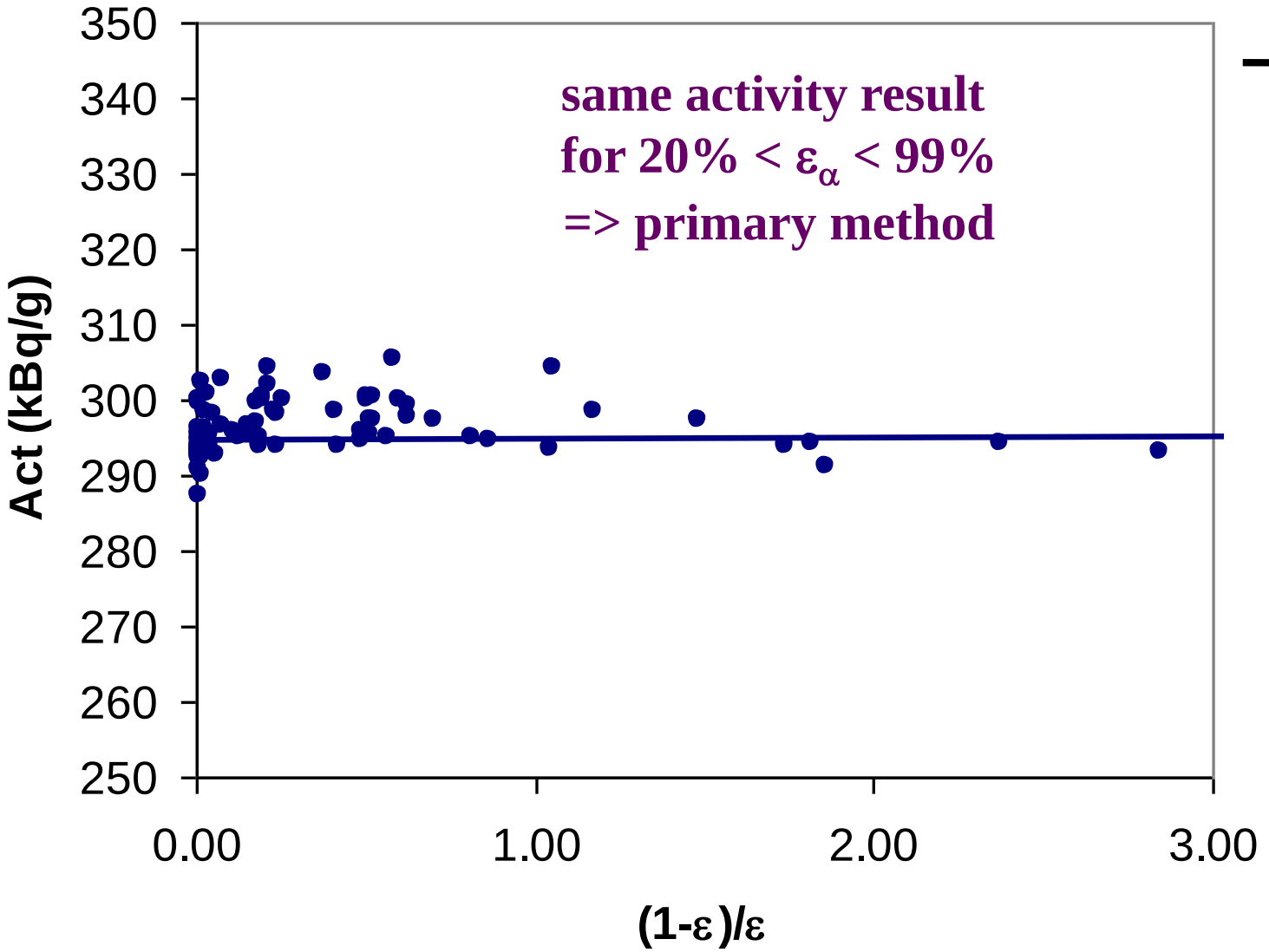
corrections cancel out for $\varepsilon_\beta \rightarrow 1$

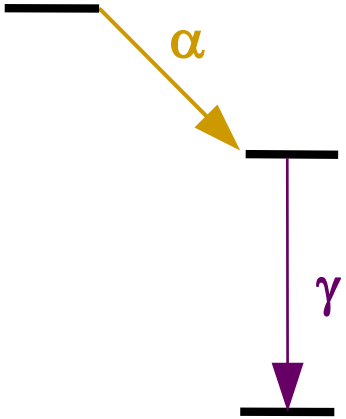
extrapolation $\varepsilon_\beta \rightarrow 1$, $N_\beta \rightarrow A$

$$N_\beta = A\left[1 - \frac{C}{D}\left(1 - \frac{N_c}{N_\gamma}\right)\right]$$



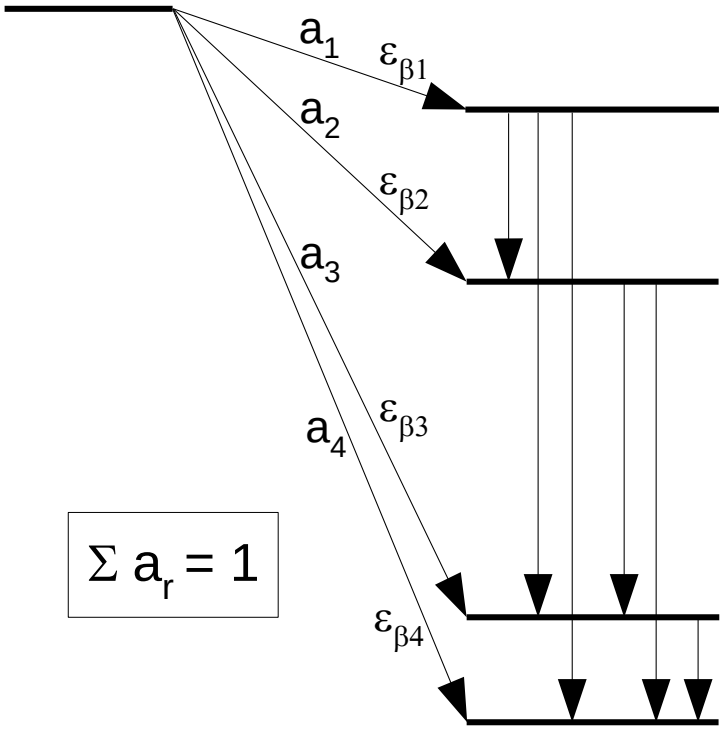
Idem for α, example : ^{241}Am by α–γ coincidences





$$\varepsilon_\alpha = \frac{N_c}{N_\gamma}$$

Multiple branches



$\Sigma a_r = 1$

$$N_\beta = A\left[1 - \frac{\Sigma C_r (1-\varepsilon_{\beta r})}{\Sigma D_r (1-\varepsilon_{\beta r})}\left(1-\frac{N_c}{N_\gamma}\right)\right]$$

$$= \text{Linear if } \frac{\Sigma C_r (1-\varepsilon_{\beta r})}{\Sigma D_r (1-\varepsilon_{\beta r})} \text{ is constant.}$$

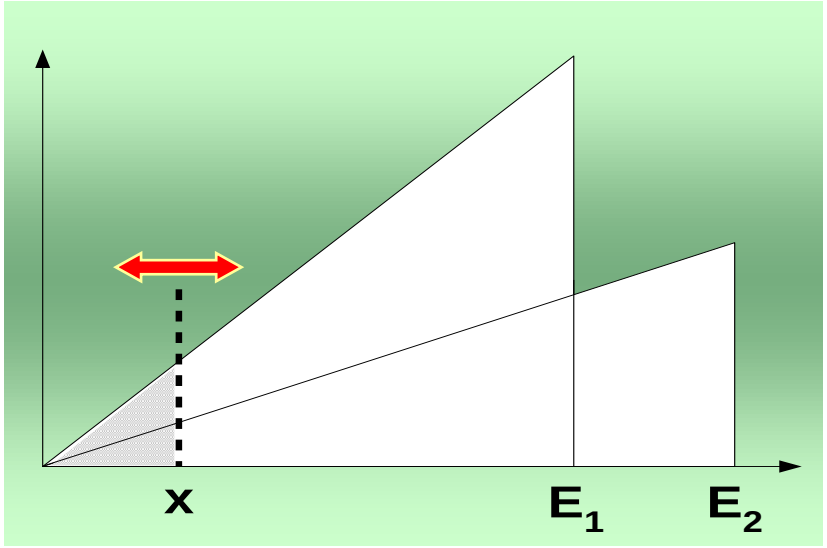
N_β extrapolates to A only in **4π beta detector** !

vary inefficiency (1- ε_β) proportionally for all branches
→ **linear** extrapolation

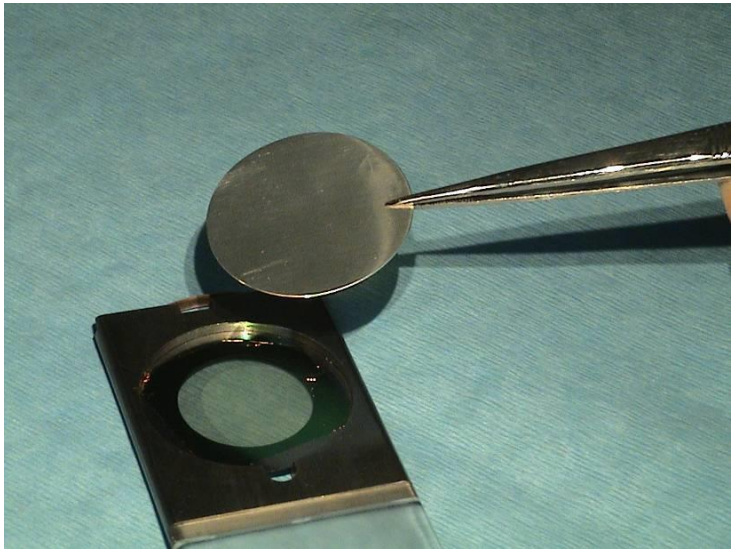
Energy discrimination

sufficient condition for correct extrapolation => vary counts in the beta detector in a manner that provides energy discrimination

threshold variation in beta channel

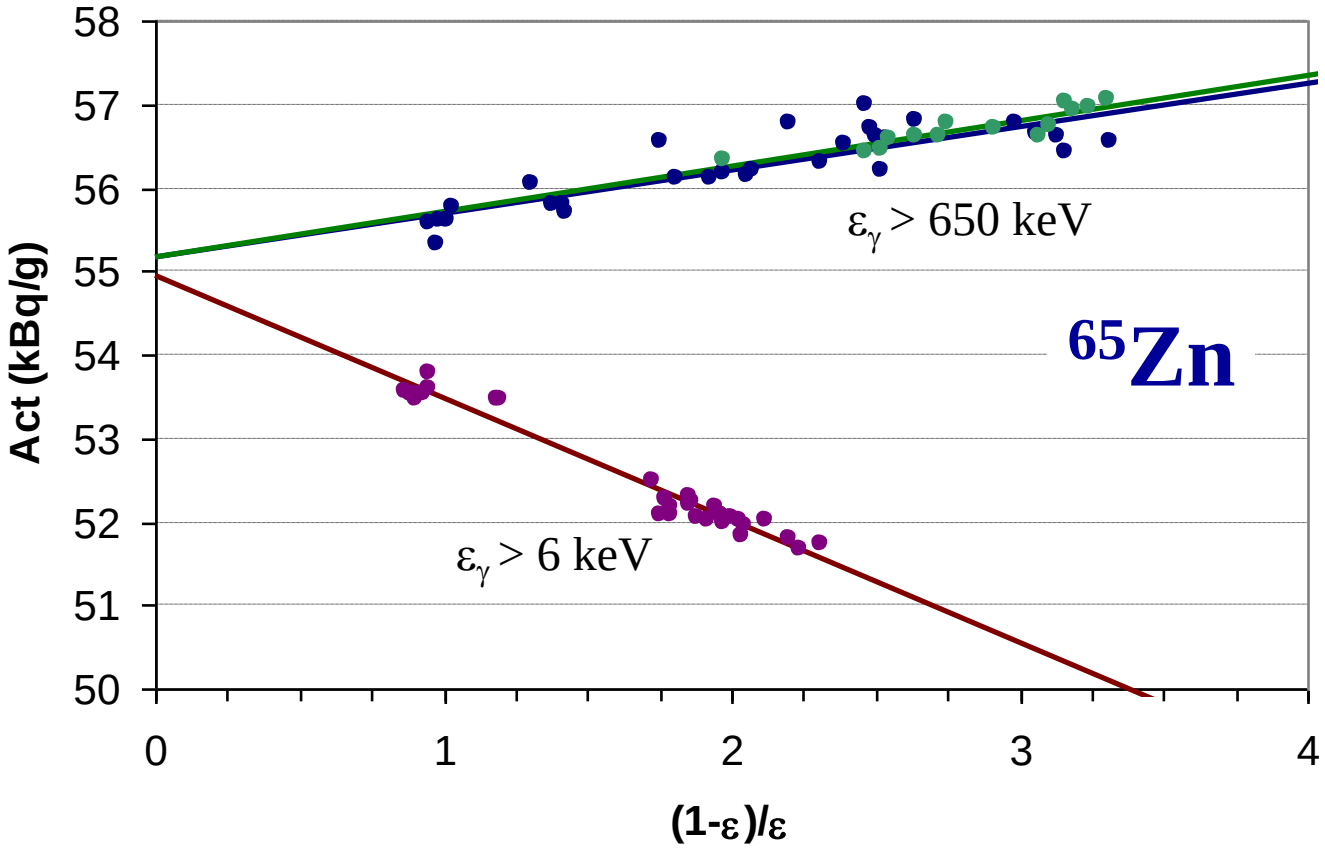


foil absorption of low-energy beta rays



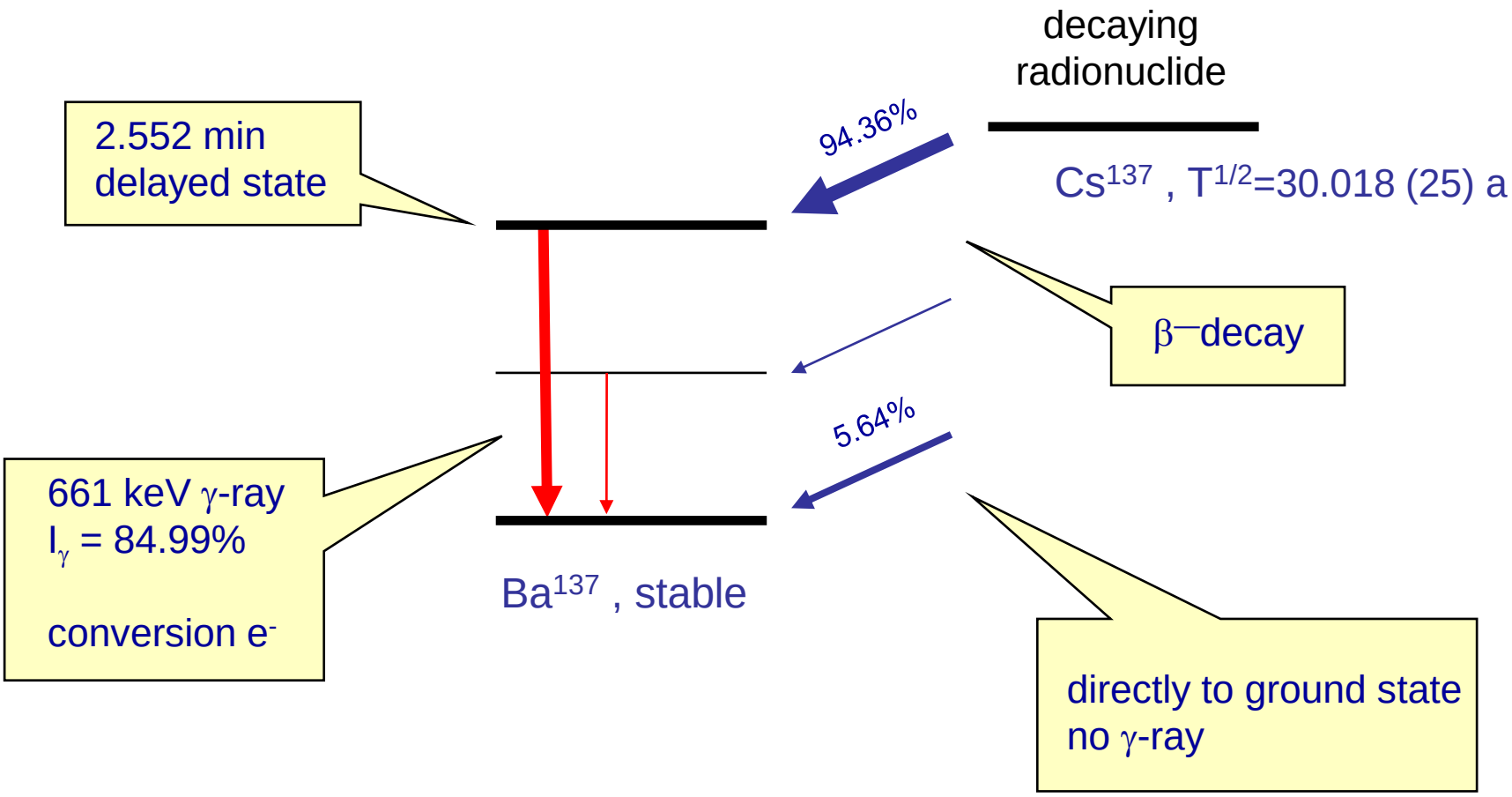
Example: β^+ decay

slope depends on inclusion of 511 keV annihilation gammas in γ-ray detector

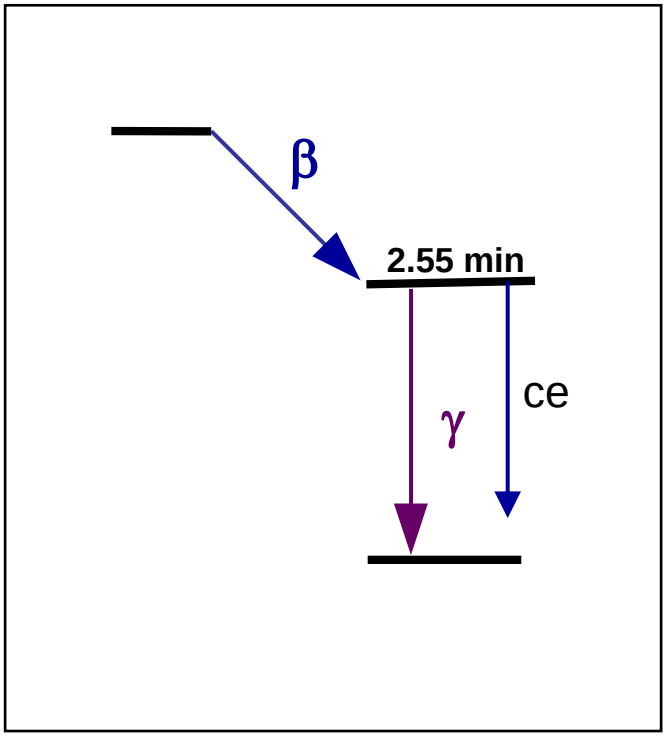


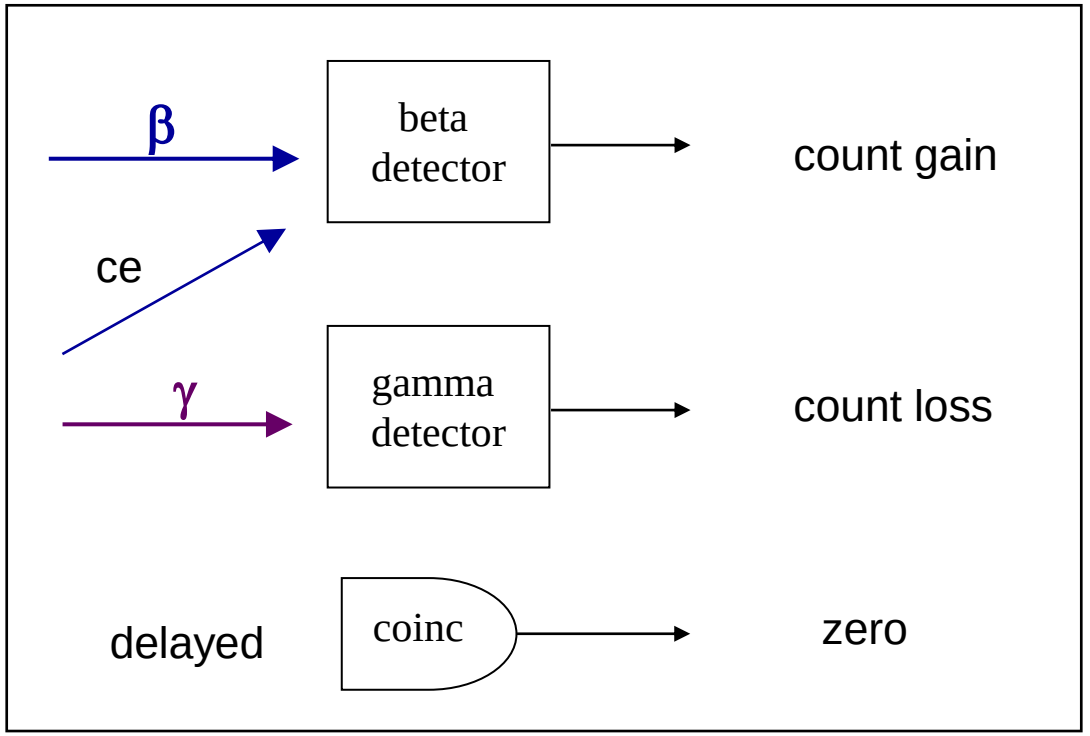
efficiency variation by cover foil absorption and threshold variation
(+ self-absorption in different sources)

^{137}Cs decay scheme



Problems with ^{137}Cs

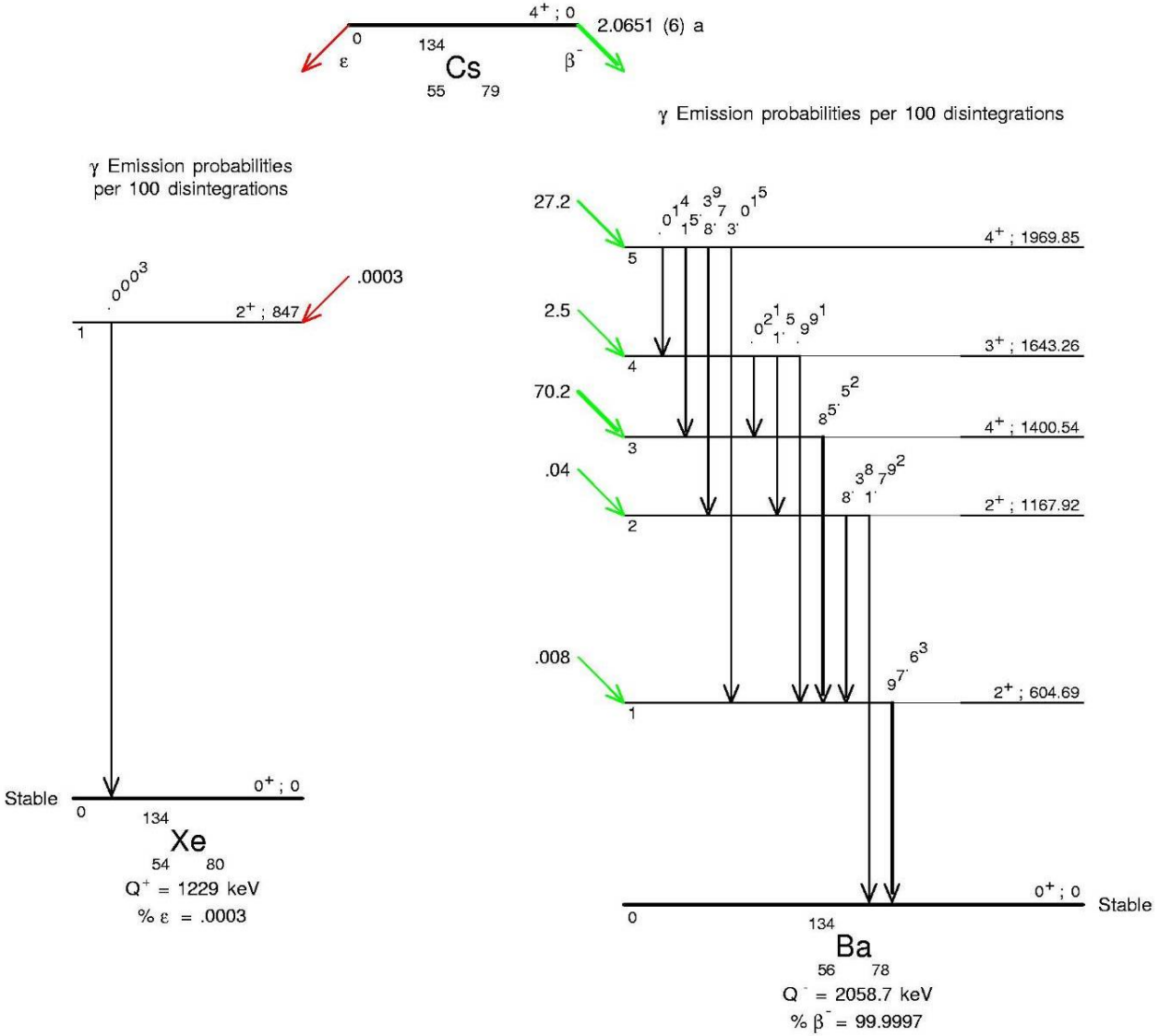




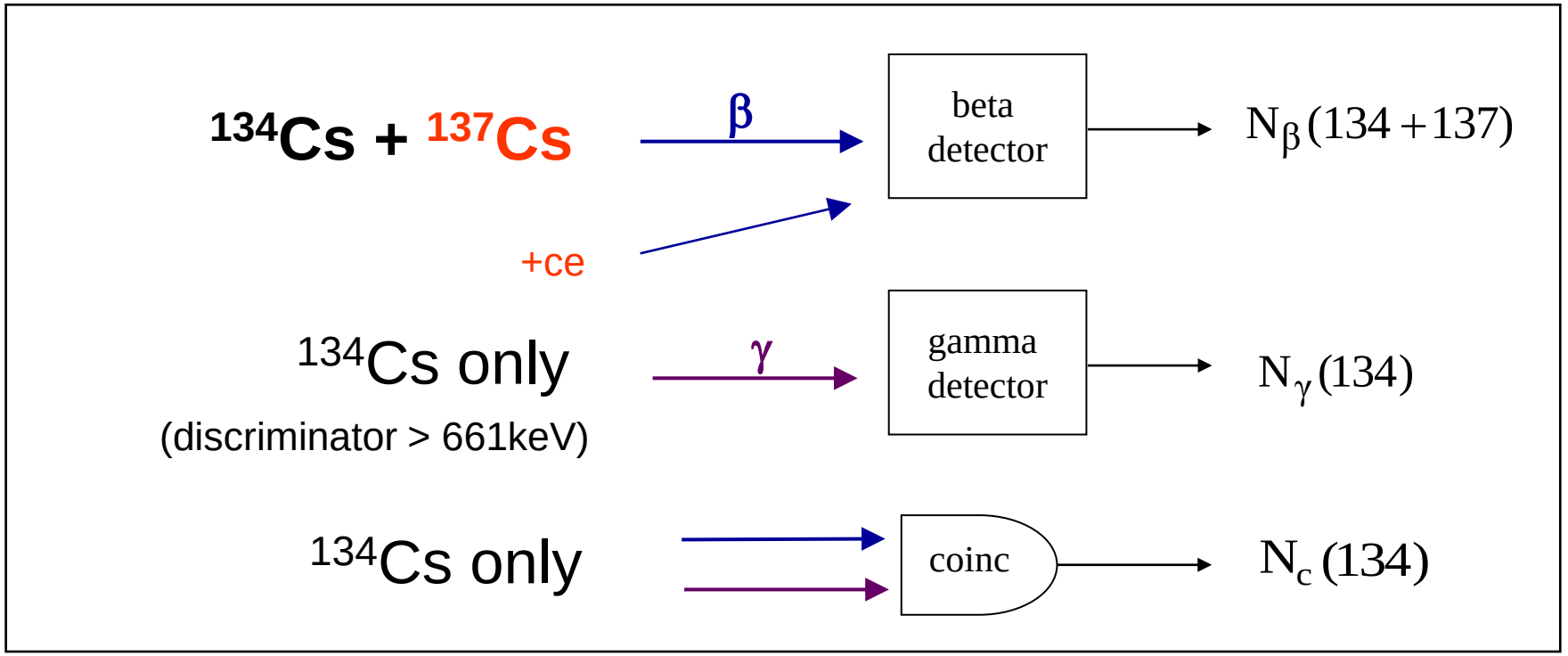
$$A = \frac{N_\beta N_\gamma}{N_c}$$

coincidence formula is no longer valid

Solution: add ^{134}Cs as tracer



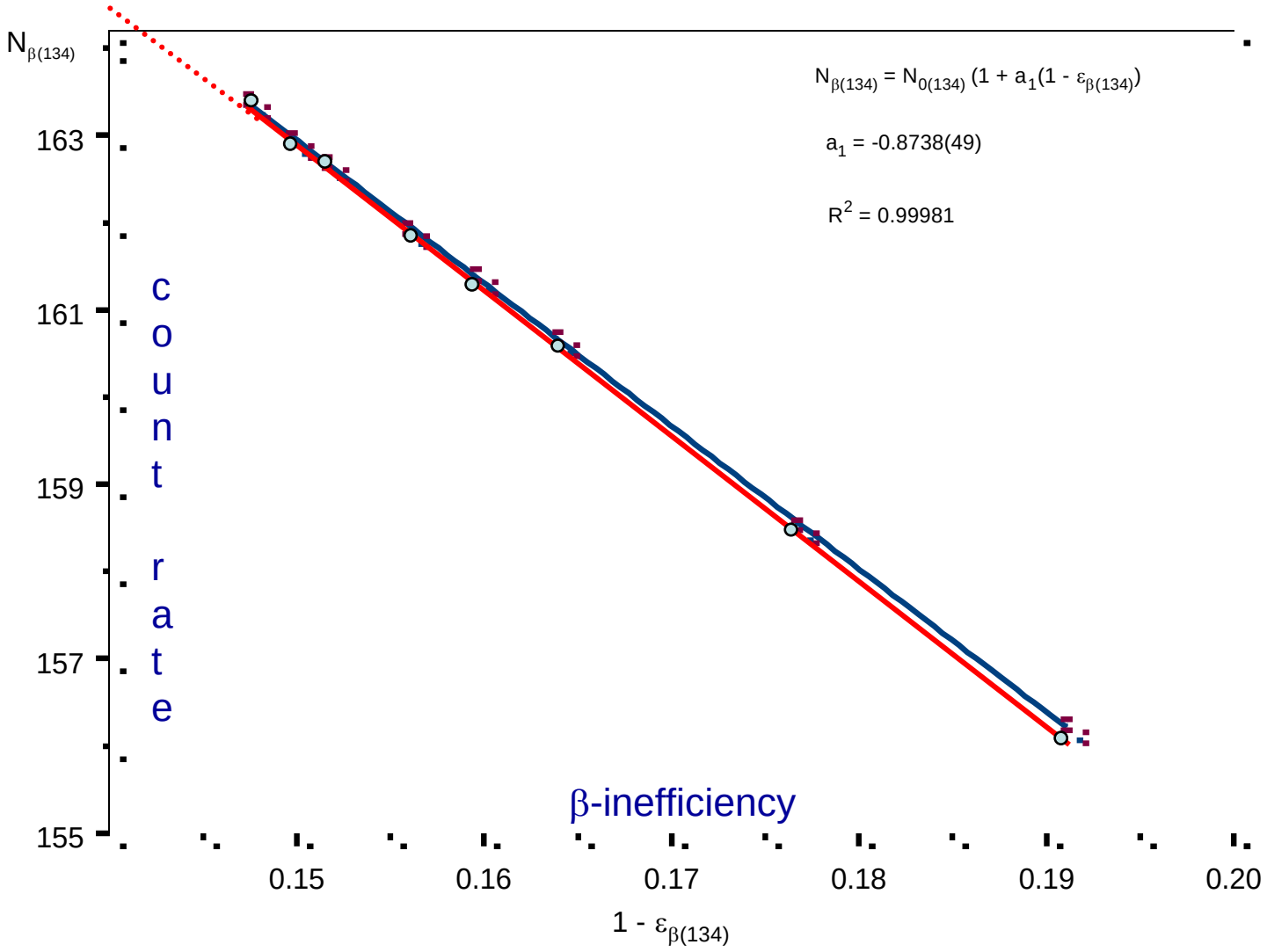
The tracer method



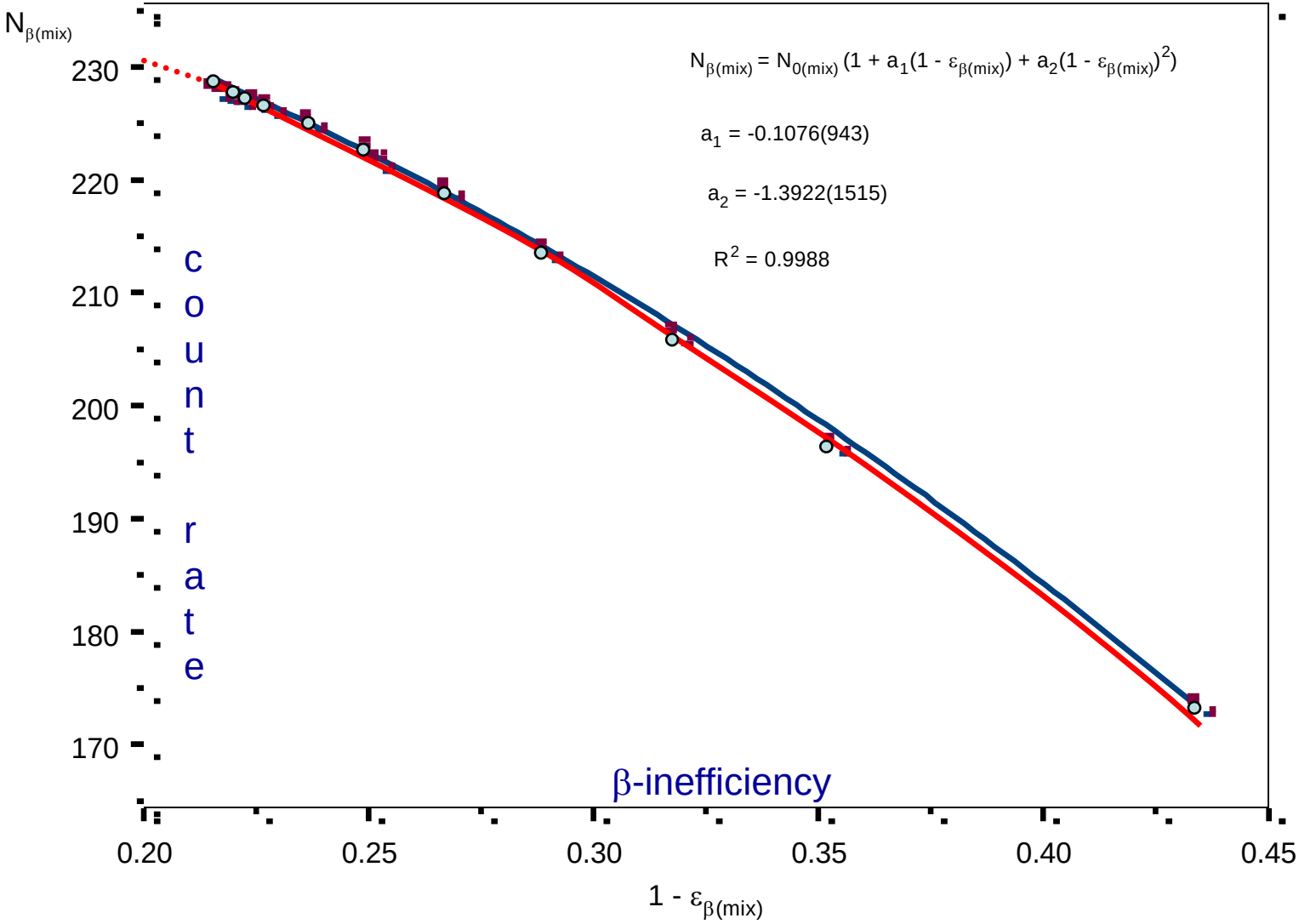
$$A(134+137) \sim \frac{N_\beta(134+137)\ N_\gamma(134)}{N_c(134)}$$

N_β=A +ce for $\varepsilon_\beta \rightarrow 100\%$

Extrapolation - pure ^{134}Cs



Extrapolation – $^{134}Cs+^{137}Cs$



References: coincidence counting

- **Metrologia 44 (2007) Special issue on Radionuclide Metrology**
- **ICRU Report 52 (1994): Particle Counting in Radioactivity Measurements**

Anticoincidence counting

Why anticoincidence counting ?

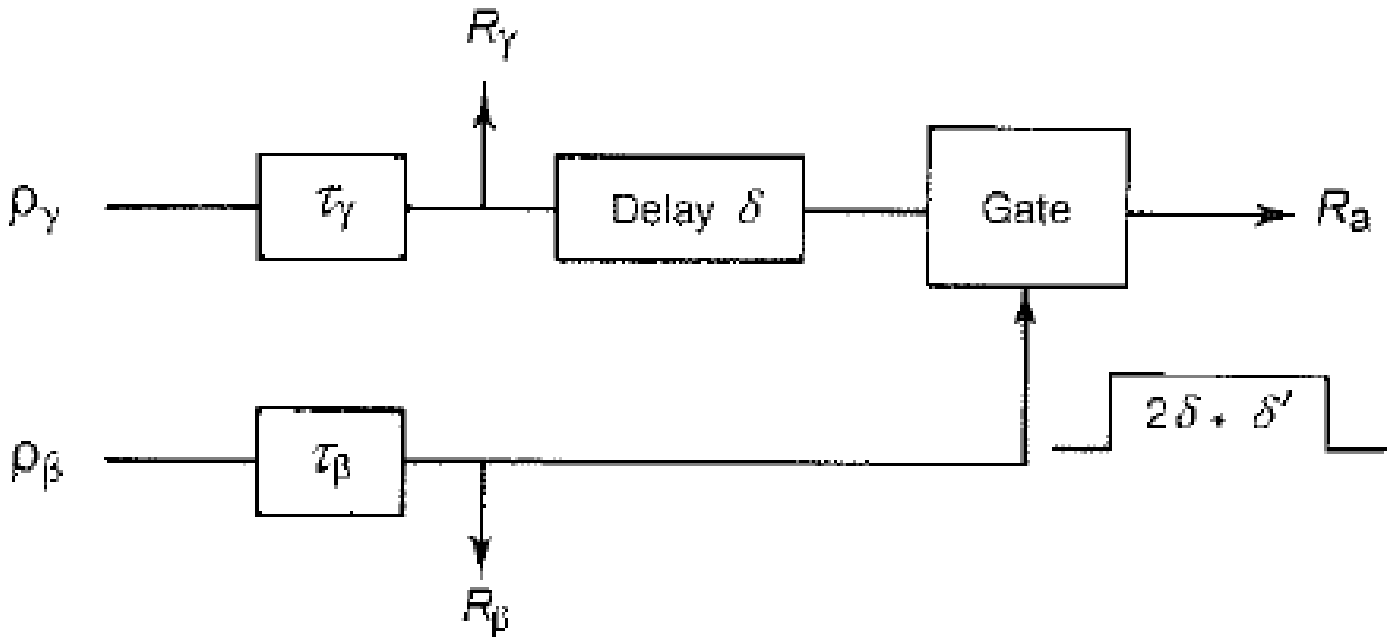
- No coincidence resolving time, no accidental coincidence correction
- Works with radionuclides with delayed excited states

Why extending-time dead-time?

- Optimum dead-time for saturated pulses and afterpulses
- Because a fixed dead-time is never really fixed

Why live-time counting?

- No dead-time correction needed
- Very simple implementation with live-time clock



$$\rho_{\beta\gamma} = \rho_\gamma - \rho_a.$$

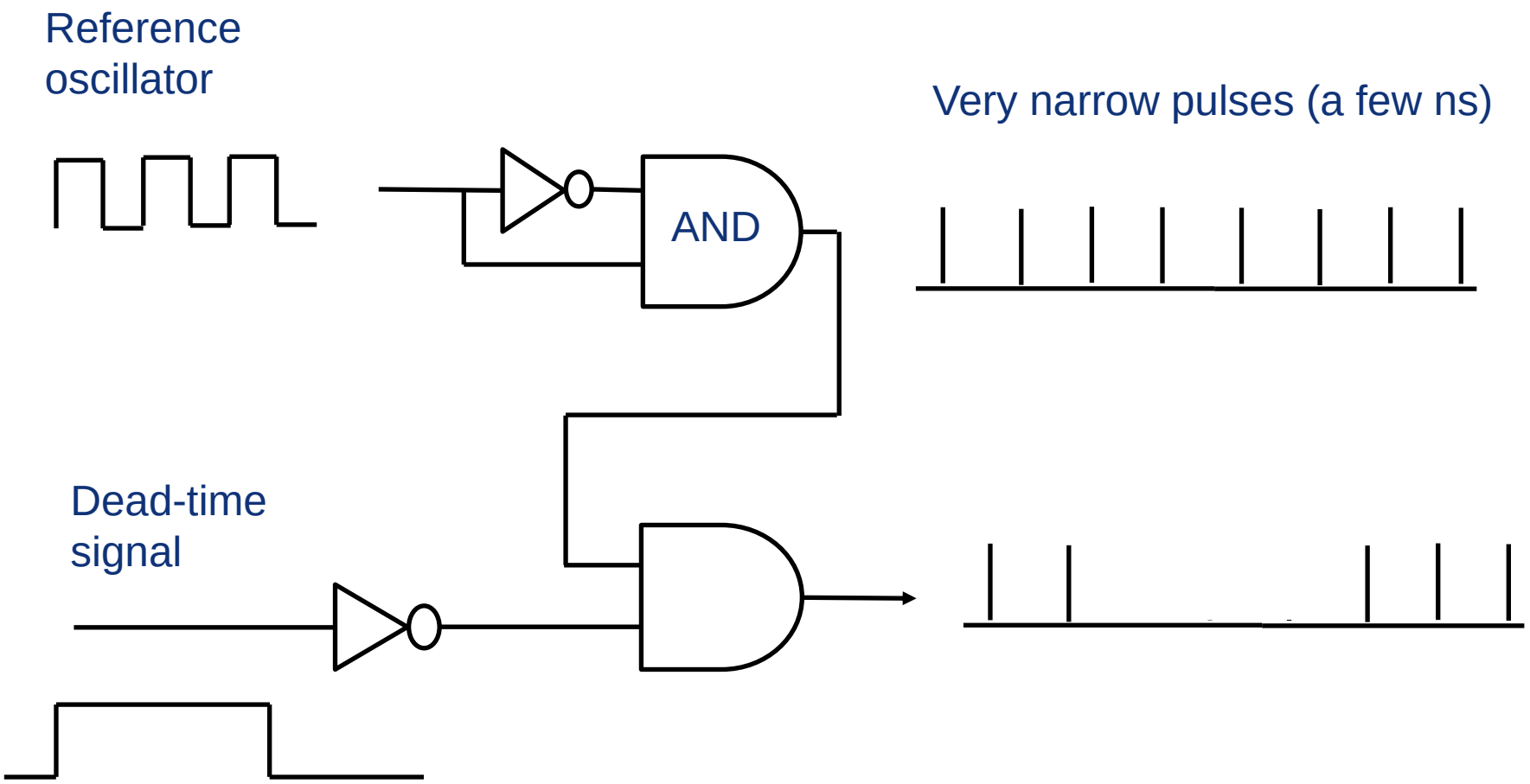
$$\rho_a \approx \frac{R_a}{1 - R_\beta (\tau_\beta + 2$$

B

$$A = \frac{\rho_\beta \rho_\gamma}{\rho_{\beta\gamma}}$$

Background corrected count rates

Principle of live-time clock



Counting the number of surviving pulses directly gives the live-time

ELSEVIER

Nuclear Instruments and Methods in Physics Research A 422 (1999) 395–399

NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A

A simple, powerful $4\pi\beta/\gamma$ coincidence system based on the pulse-mixing method

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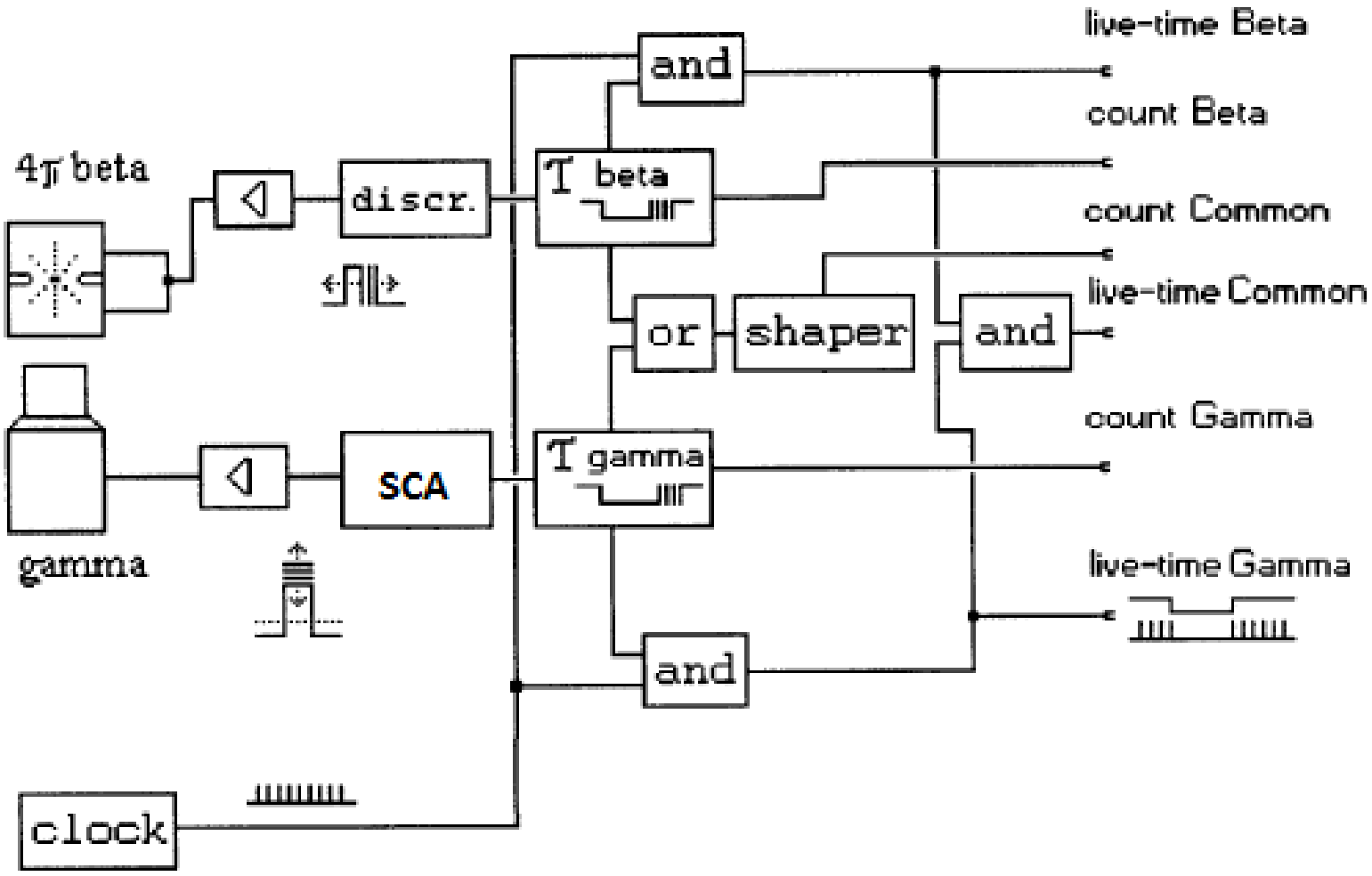
BNM-LPRI (Laboratoire Primaire des Rayonnements Ionisants) CEA/Saclay, F 91191 Gif-Sur-Yvette Cedex, France



Pulse mixing principle (single gamma channel)

A: source activity

B: uncorrelated beta pulses

G: uncorrelated gamma pulses

C: correlated pulses

Beta channel: B+C

Gamma channel: G+C

t_b: live time beta channel

t_g: live time gamma channel

t_c: live time common channel

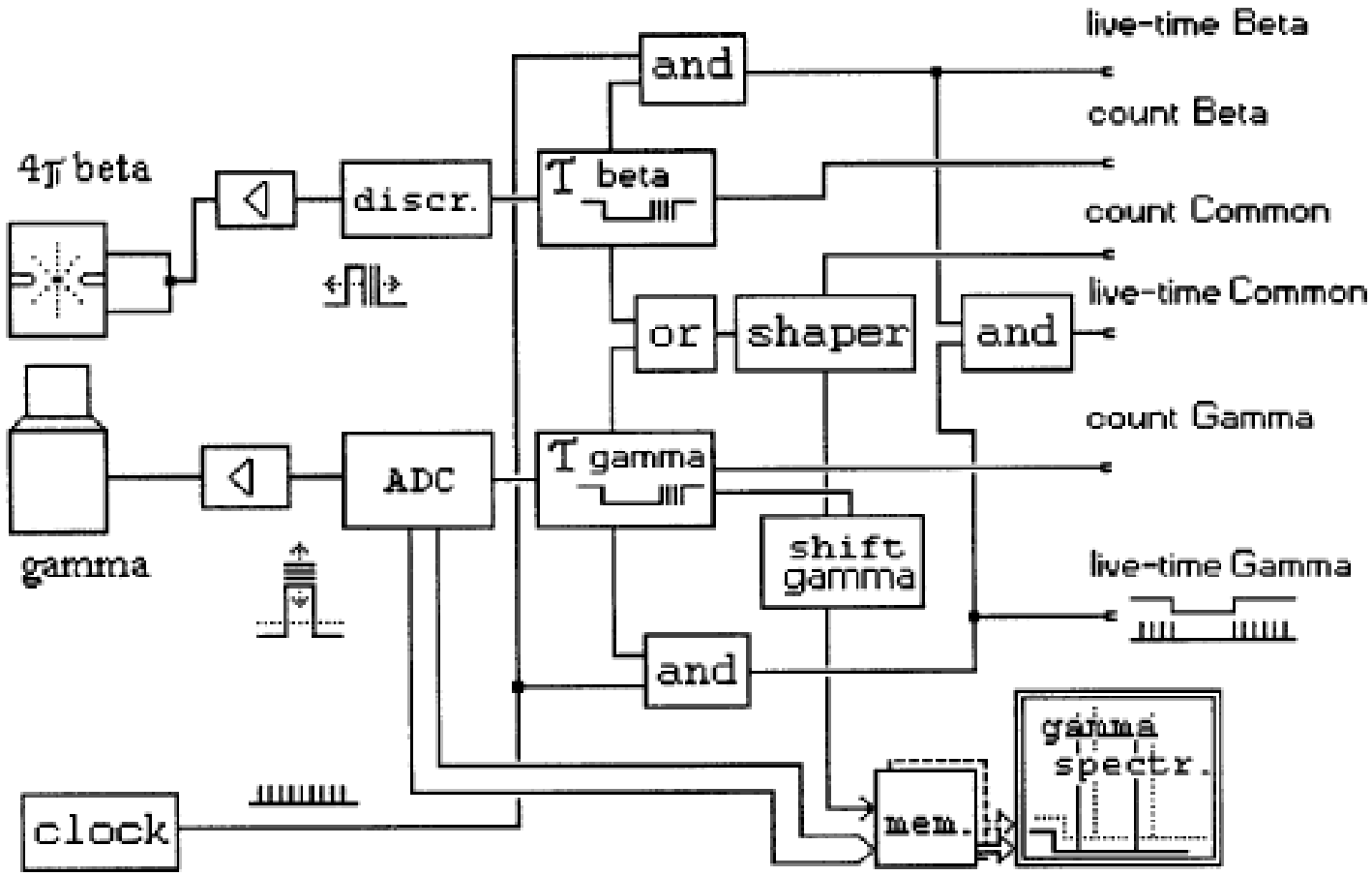
N_b, N_g, N_c counts in channels b, g and c

$$N_b/t_b = B + C$$

$$N_g/t_g = G + C$$

$$N_c/t_c = B + G + C$$

$$A = \frac{N_b/t_b * N_g/t_g}{N_c/t_c} = \frac{(B+C)+(G+C)}{(B+C)+(G+C)-(B+G+C)}$$



Pulse mixing principle (multiple gamma channels)

Advantages

- Counting statistics similar to traditional coincidence counting
- No correction needed: dead-time, accidental coincidences, time jitter…
- Can be used for radionuclide with metastable states
- Very simple implementation!

But

All requirements for coincidence counting must be fulfilled

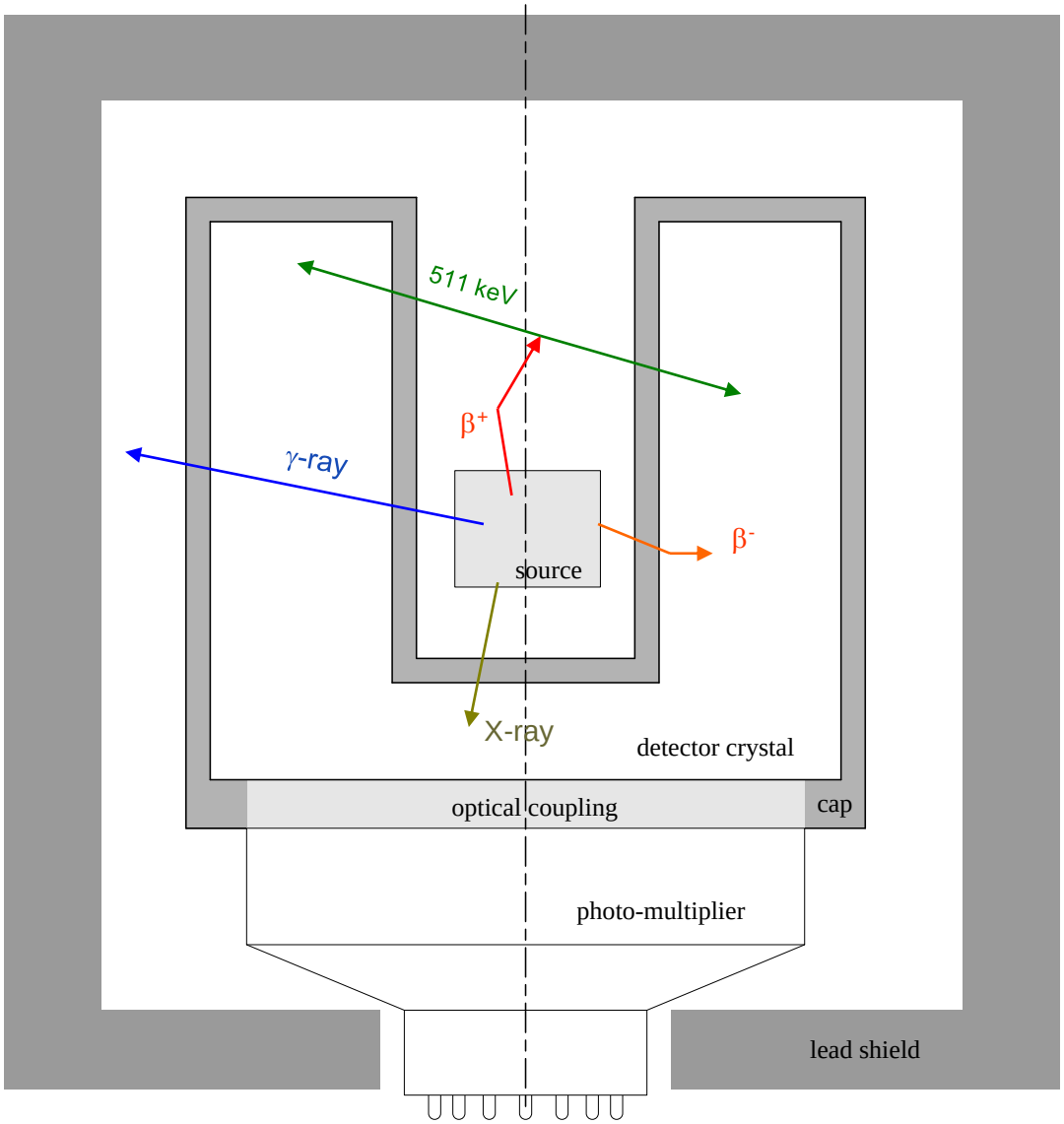
Extrapolation generally needed

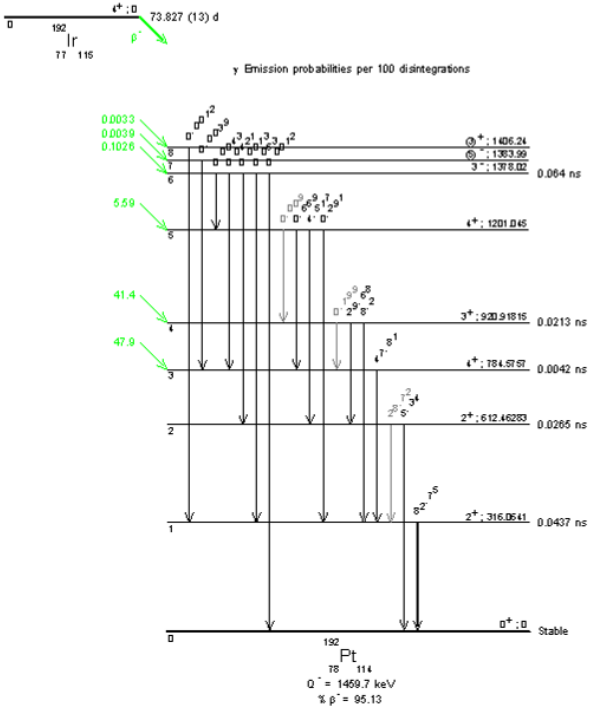
Primary Standardisation of activity

4π–γ Counting

“4πγ- counting” in a NaI well detector

4πγ- counting

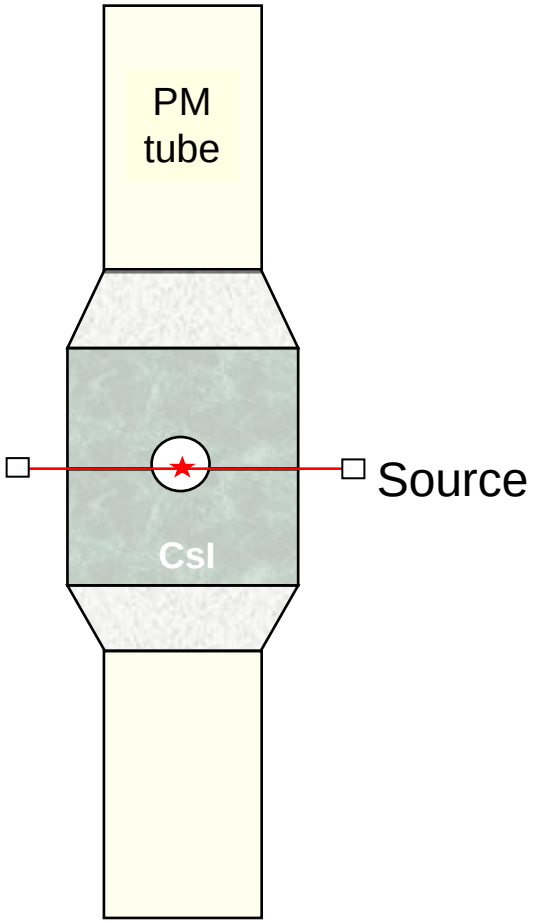




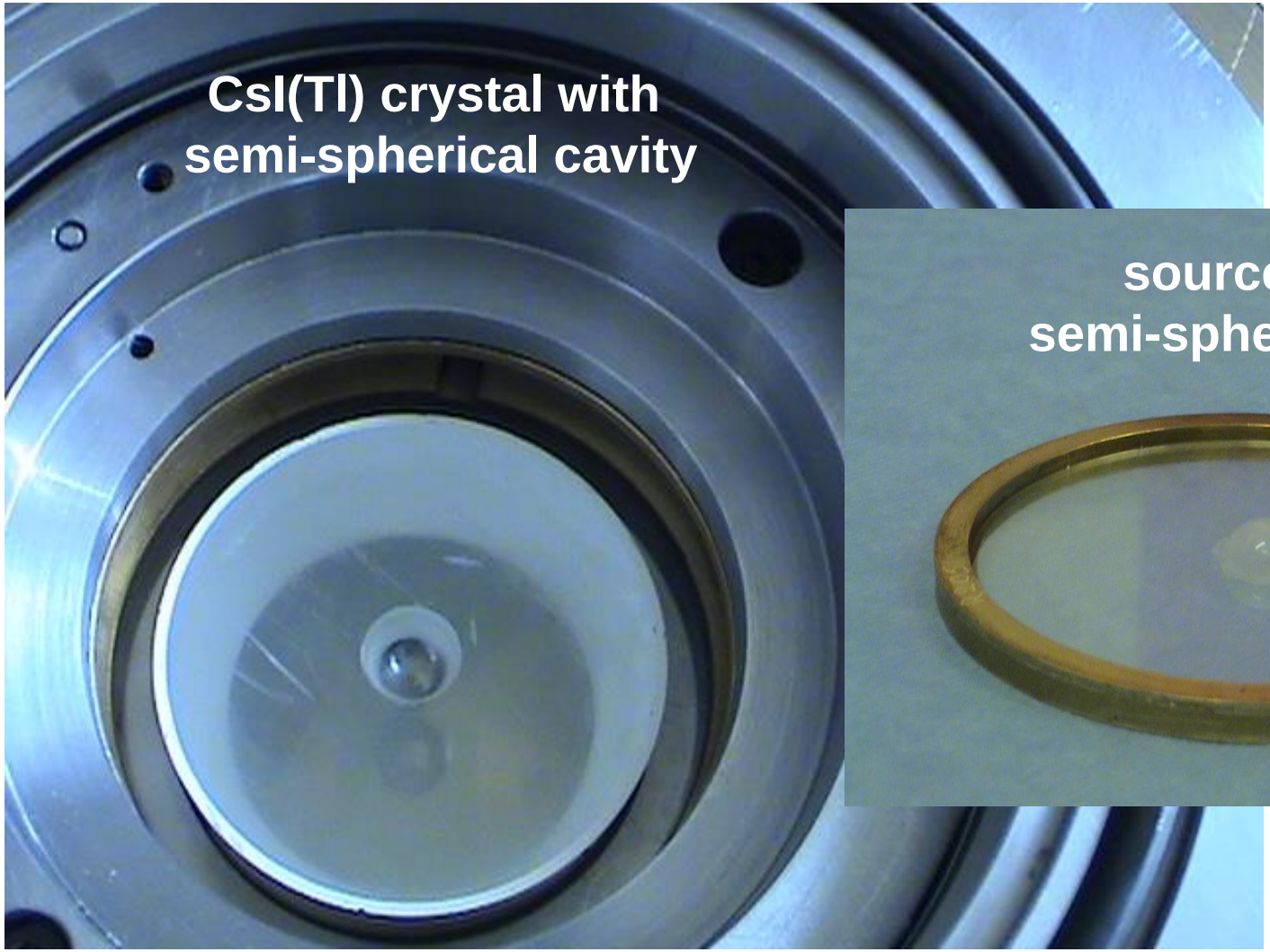
well-suited for complex decays

efficiency ≈ 100%

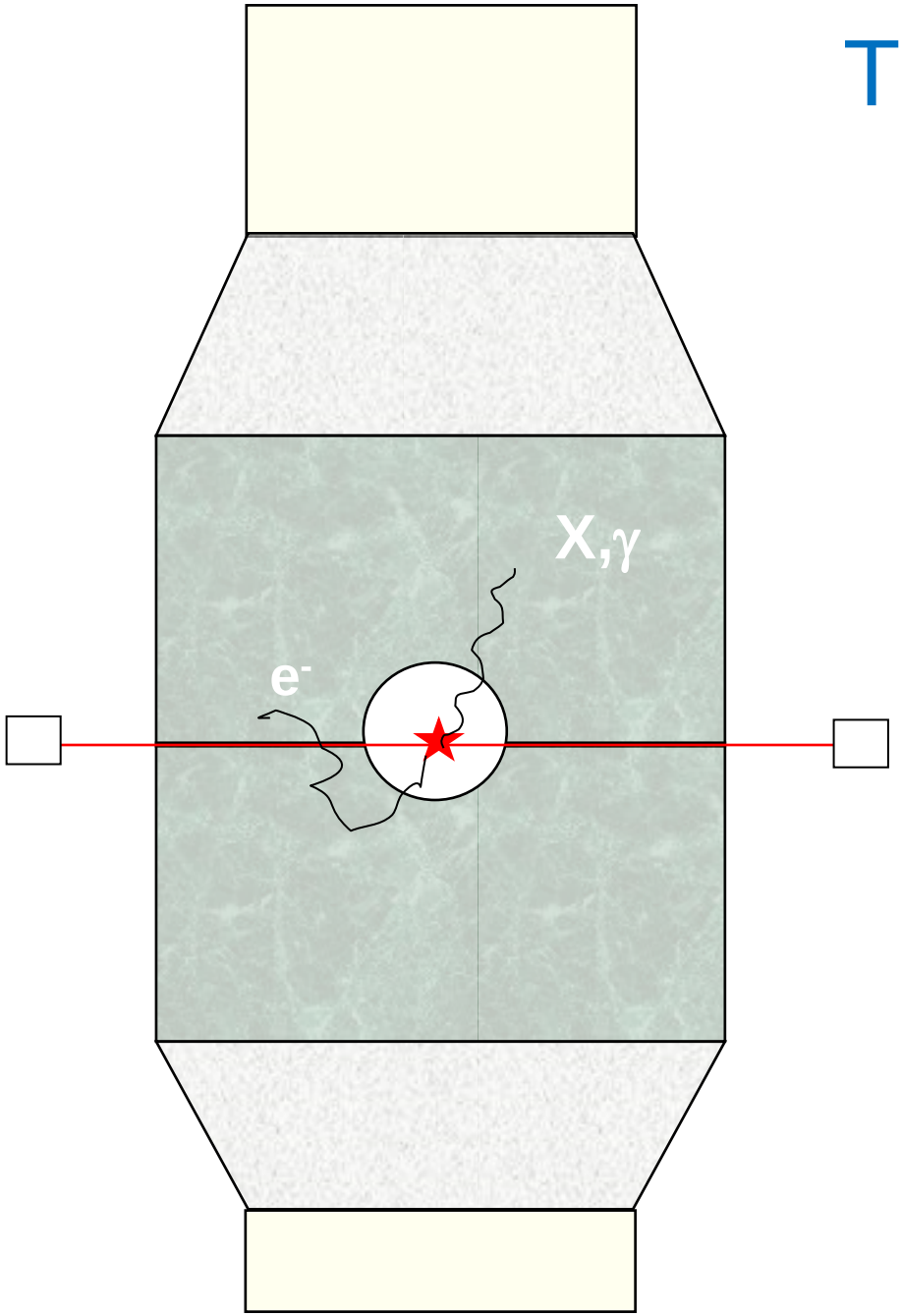
4π photon, electron, alpha counting by CsI(Tl) sandwich spectrometer



cover source by plastic caps
to stop electrons and alpha-particles



Total emission counting

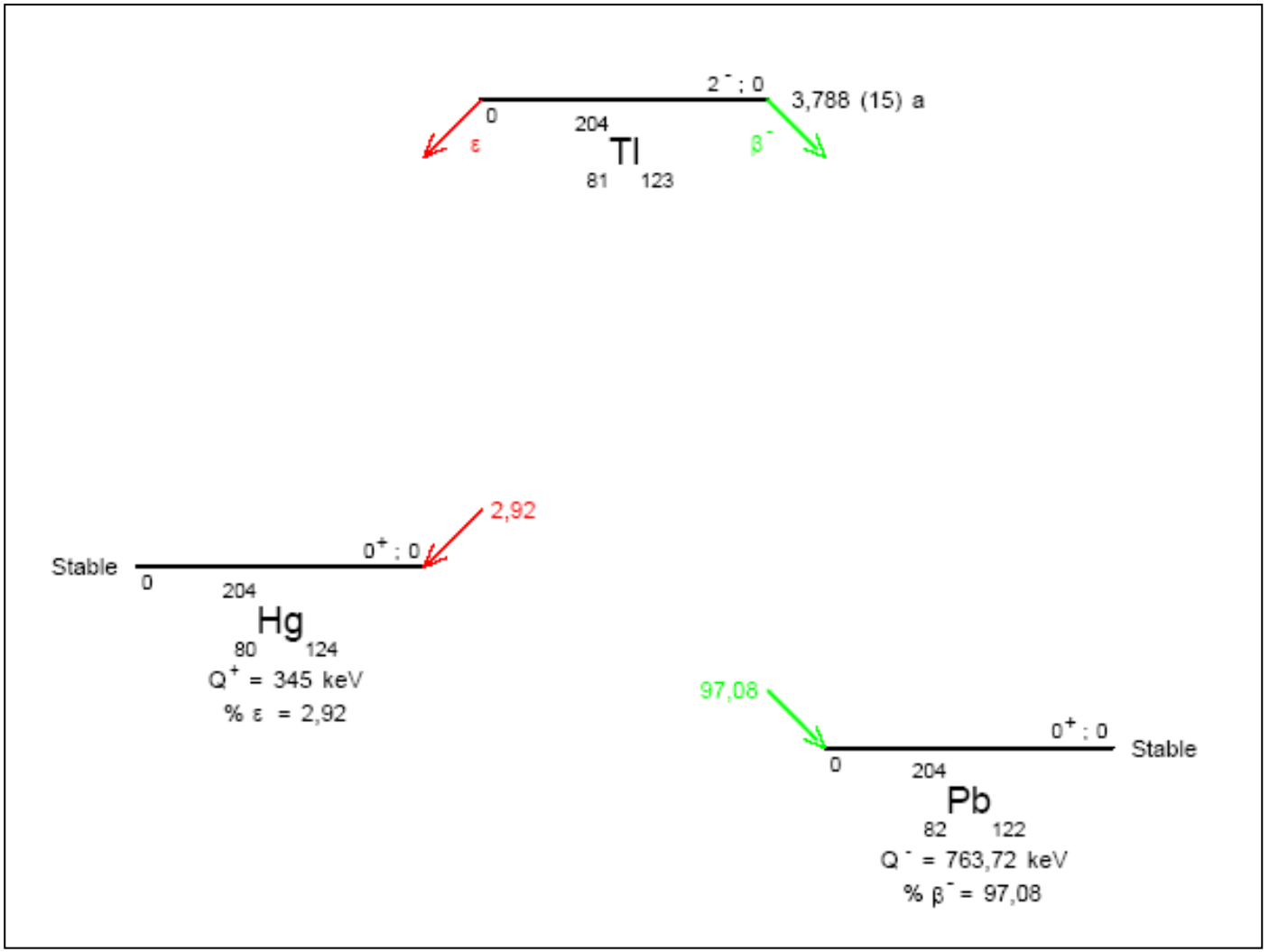


K X –rays and γ-rays and electrons are being measured

Primary Standardisation of activity

4π–β,α Counting by pressurised proportional counter

Example: standardization of ^{204}Tl



Pressurised proportional gas counter

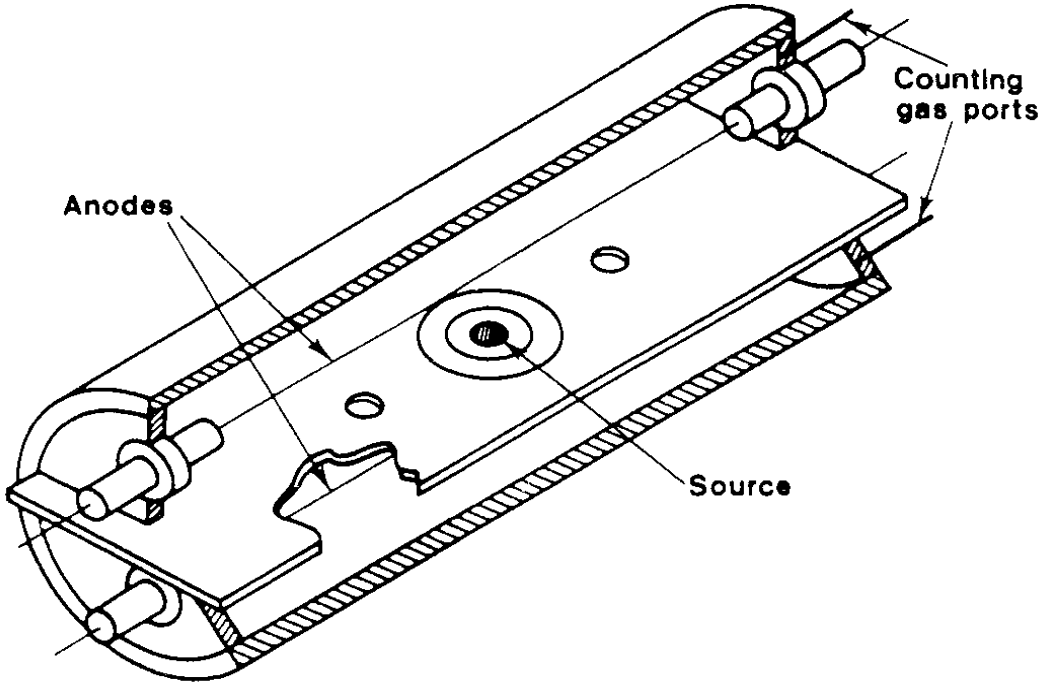
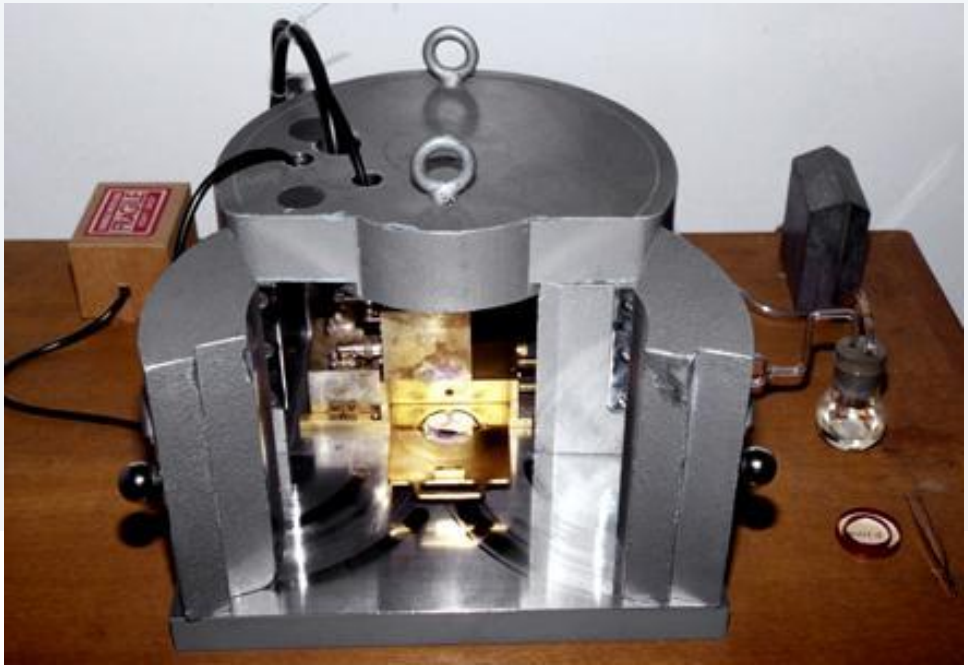
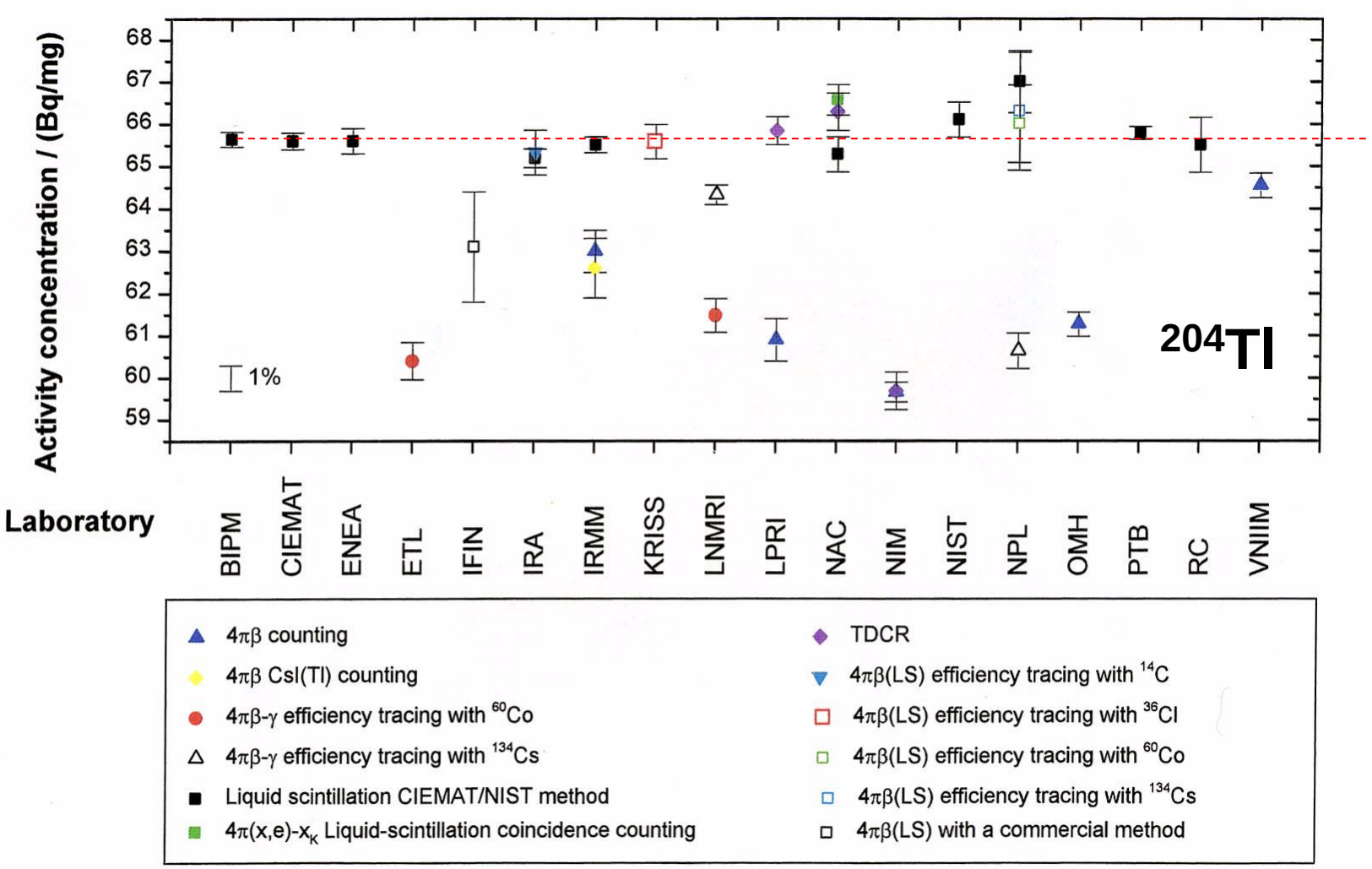


Fig. 5-11. Cut-away representation of a cylindrical 4π gas-flow proportional counter.

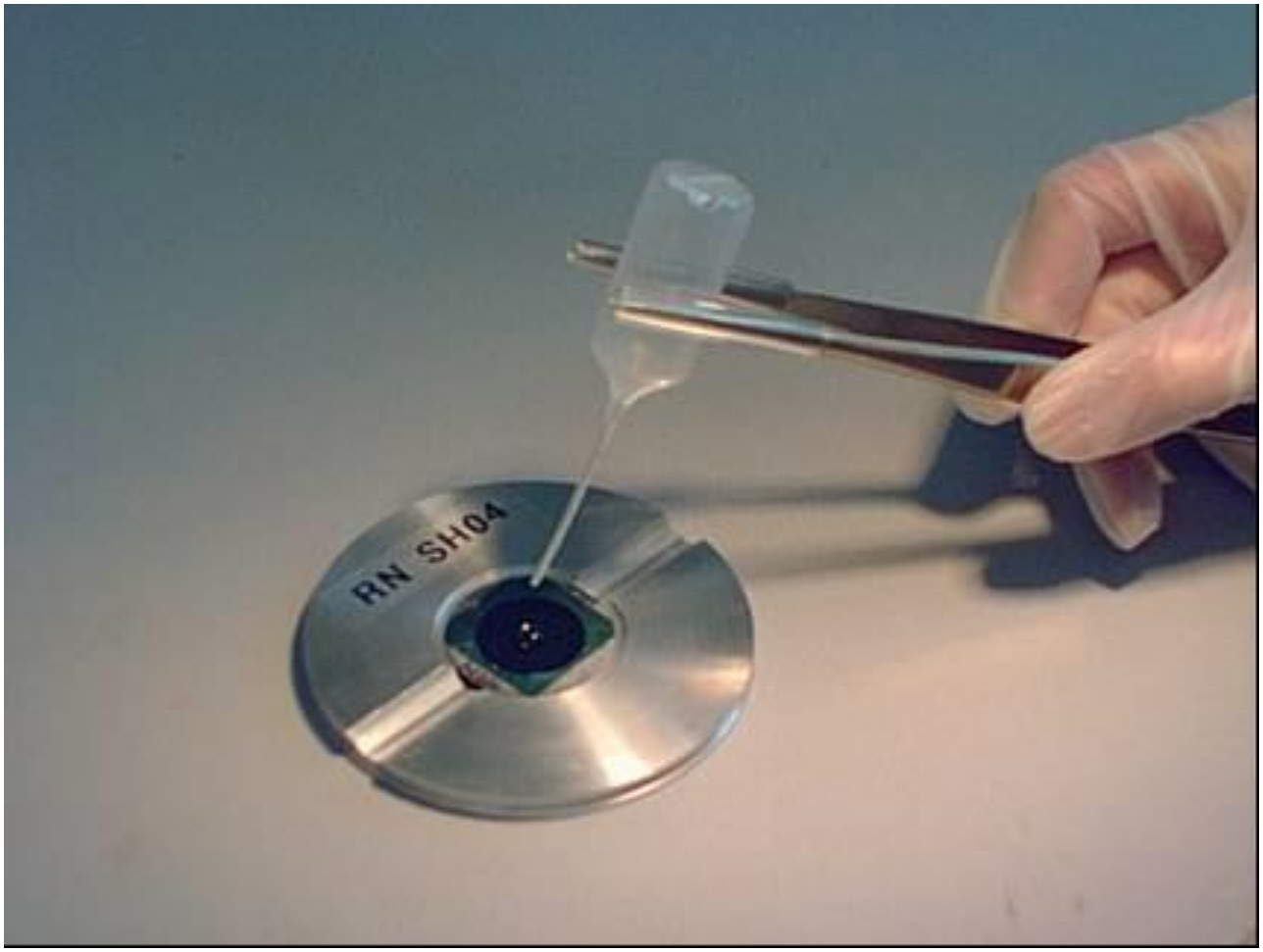
Example of 4π proportional counter (BIPM)



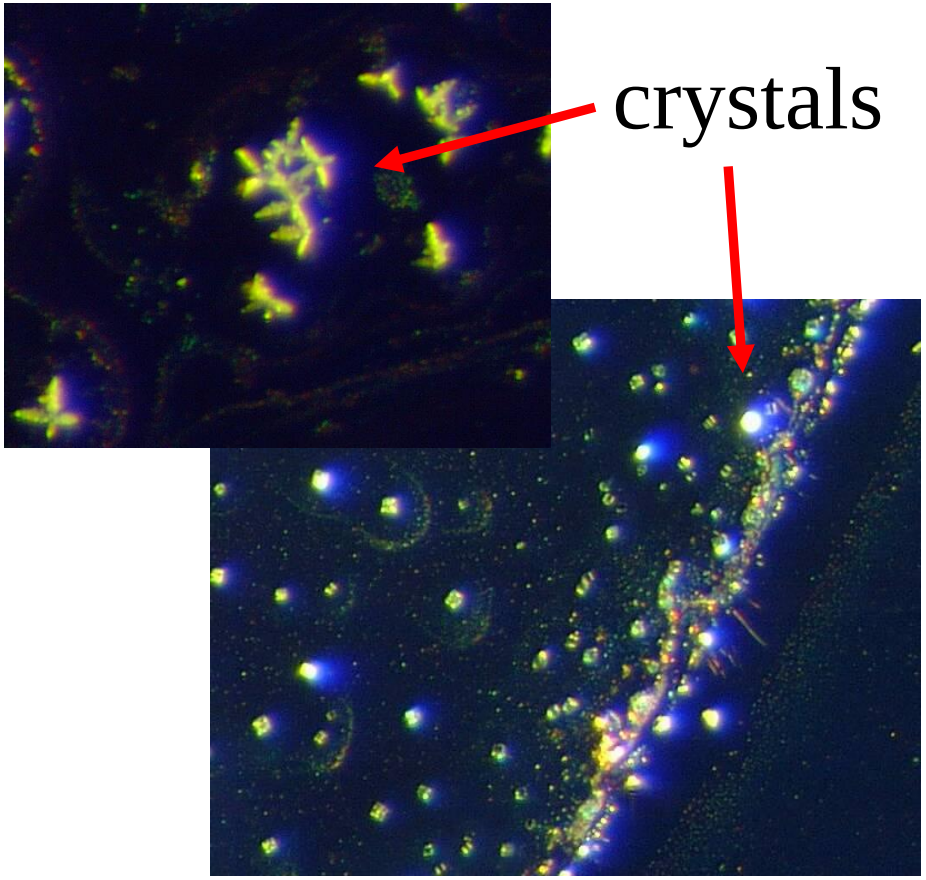
Problematic case

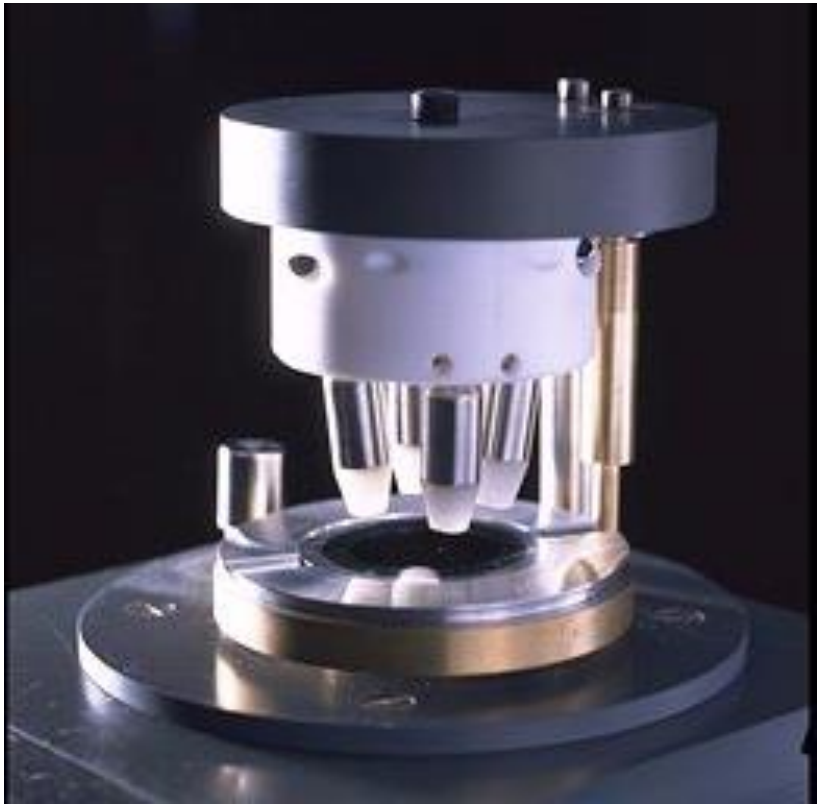


quantitative sources by ‘drop deposition’ with pycnometer



Fast source drying to reduce self-absorption in the sources





Primary Standardisation of activity

Liquid scintillation counting

LSC as a direct measurement method

Activity (Bq)

$$A = \frac{N}{\varepsilon}$$

Counting rate (s^{-1})

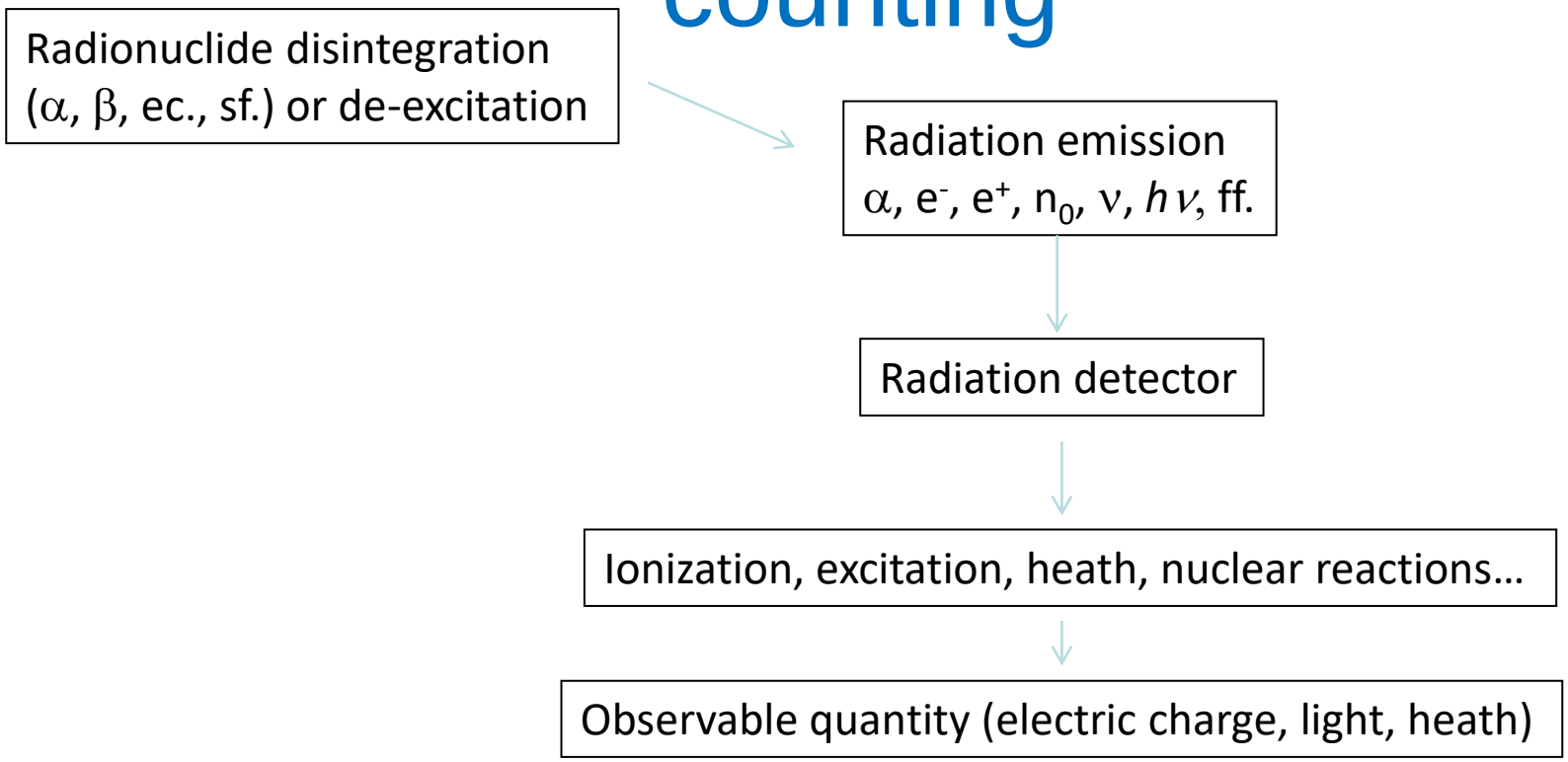
Detection efficiency

Detection efficiency is calculated without using a standard (of the same radionuclide)

Calculation from a model of the various physic-chemical phenomena occurring during the LSC process:

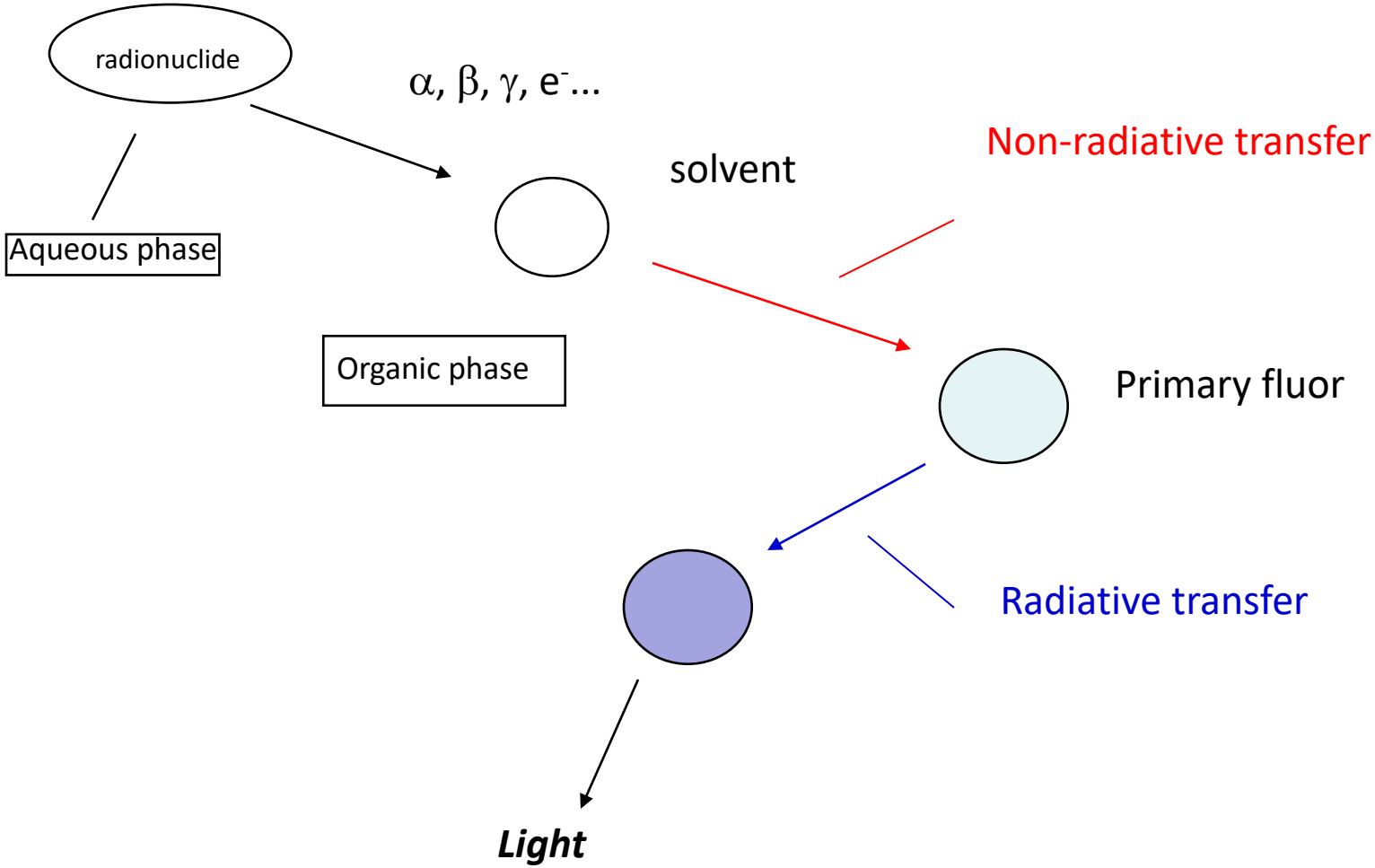
radionuclide decay, radiation emission, radiation absorption by the scintillator, energy transfer in the scintillator, light emission statistics, light propagation, light detection by the photomultiplier tubes, coincidence counting…

Activity measurement, from radionuclide disintegration to pulse counting



The measurande (mean number of disintegrations per time unit) is not directly observable, *sometimes a disintegration produces no directly observable information*

The LS process



Energy transfer

"Eventually, ionizing radiation is transformed into electron energy or heath"

Solvent excitation (main mechanisms, very simplified)

$$S_0 + e^- \rightarrow S_n^* \rightarrow S_1^*$$

$$S_0 + e^- \rightarrow T_n^* \rightarrow T_1^*$$

Energy transfer in the solvent

$$S_1^*(1) + S_0(2) \rightarrow [excimer\ ?] \rightarrow S_0(1) + S_1^*(2)$$

$$T_1^*(1) + T_1^*(2) \rightarrow S_1^*(1) + S_0(2)$$

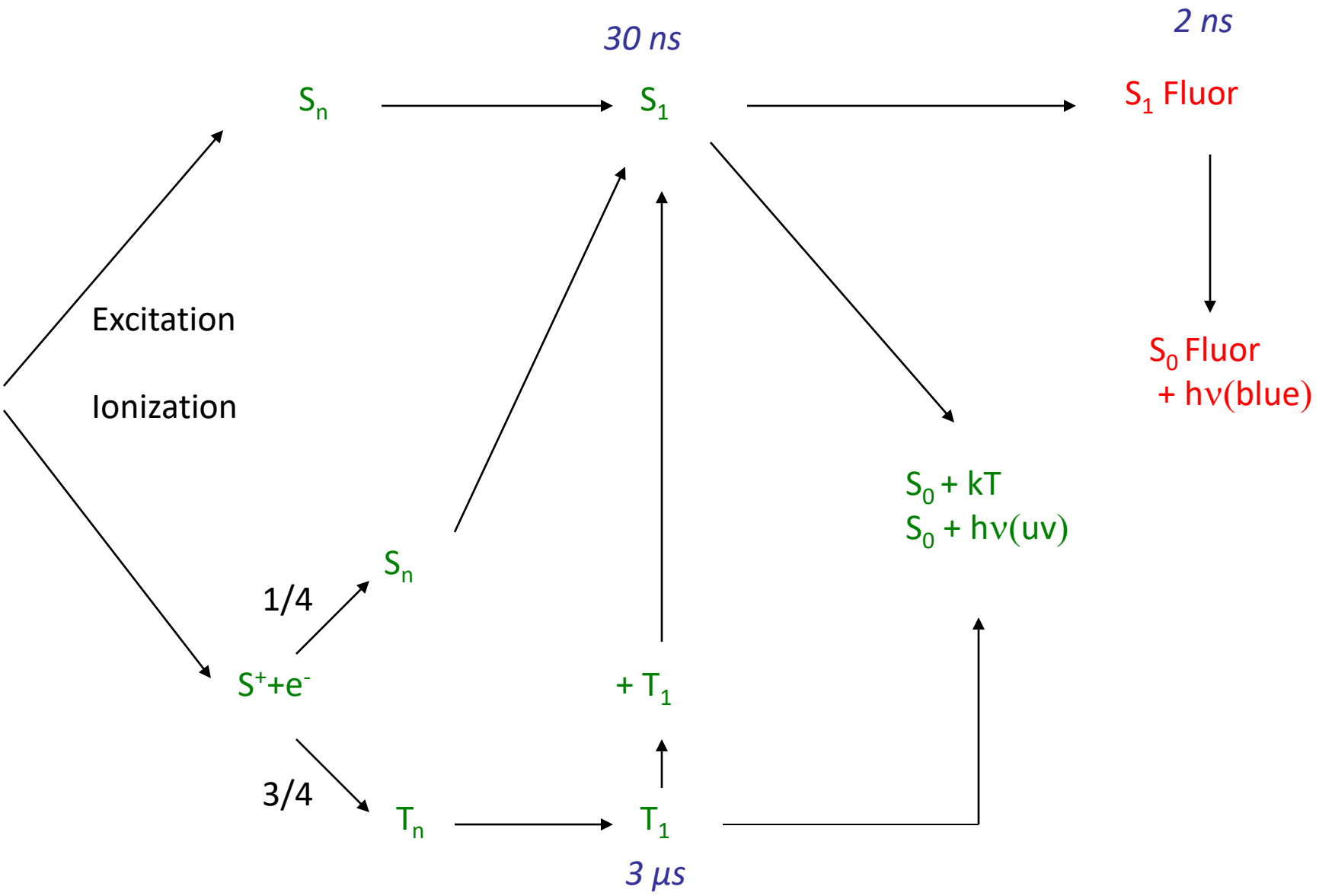
Quenching and light emission

$$S_1^* + M \rightarrow S_0 + M + kT$$ Chemical quenching

$$S_1^*(1) + S_1^*(2) \rightarrow S_0(1) + S_0(2) + kT$$ Ionization quenching

$$S_1^* + F \rightarrow S_0 + F_1^* \rightarrow F + h\nu$$ Light emission!

Example of energy transfer in a toluene-PPO cocktail

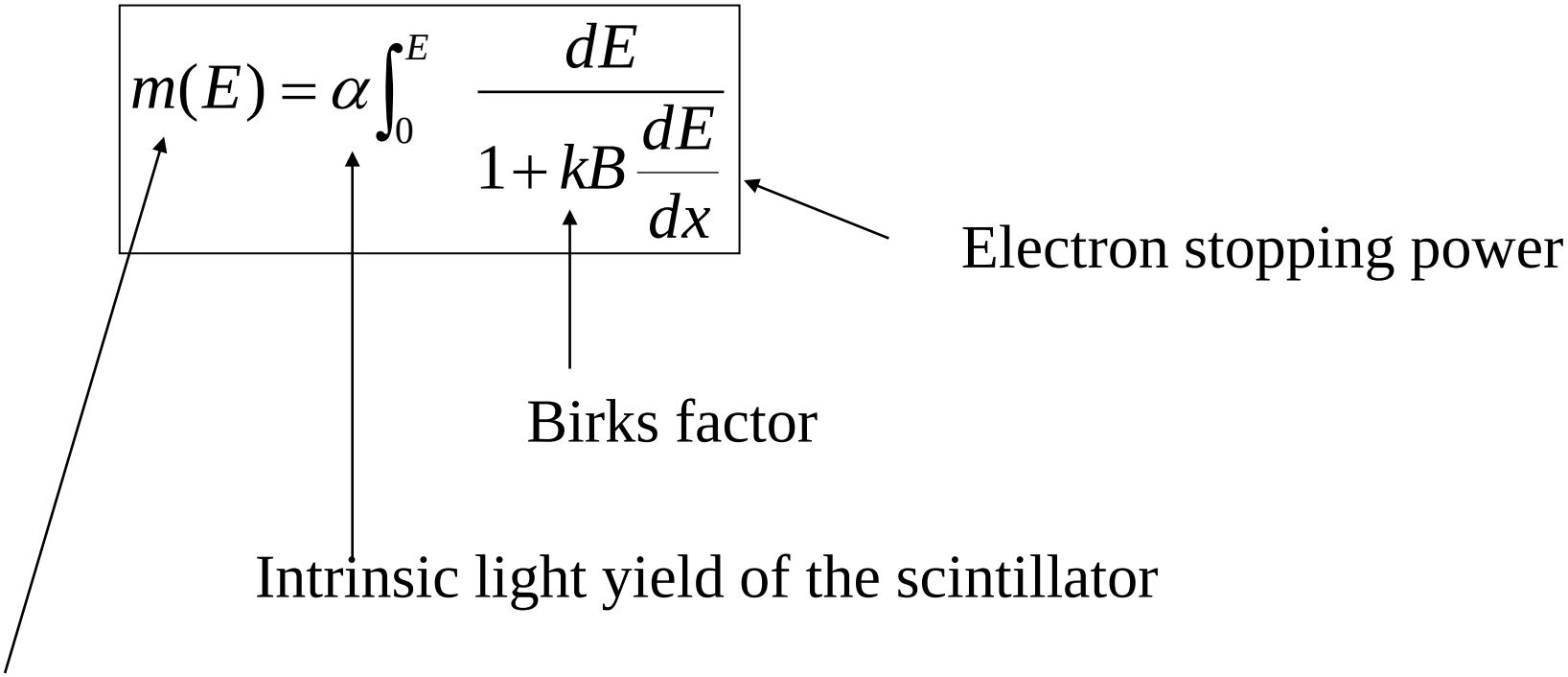


Consequences

- Light emission is a fast process (some ns)
- Light emission yield is low (a keV of energy produces a few photons)
- The number of photons emitted is random
- The mean number of photons emitted is not proportional to the energy

Light emission, Birks formula

If an electron with energy *E* is absorbed by the liquid scintillator, a mean number of *m* photons are emitted



Light emission

The number of photons emitted is a Poisson-distributed random number

$$P(x/m) = \frac{m^x e^{-m}}{x!}$$

Probability of emission of *x* photons for a mean value *m(E)*

Detection efficiency when an energy *E* is absorbed by the liquid scintillator

A photon will produce a photoelectron in the photocathode of the PMT with a probability ν *(quantum efficiency of the photocathode)*

$$P(y\,I\,m) = \frac{(\nu m)^{y} e^{-\nu m}}{y!}$$

Probability to create *y* photoelectrons for a mean value of *m(E)* photons

Detection efficiency (1 PMT)

- The detection efficiency is the detection probability
- The detection probability is 1 minus the non-detection probability
- Non-detection probability : probability of detection of *0* photon when a mean value of *m* is expected

$$\varepsilon = \left[1 - P(0)\right] = 1 - \frac{(\nu m)^0 e^{-\nu m}}{0!}$$

$$\boxed{\varepsilon = 1 - e^{-\nu m}}$$

The detection efficiency is a function of a free parameter, νm, mean number of photoelectrons produced after the absorption of E

Detection efficiency of electrons with energy spectrum S(E) injected in a liquid scintillator

$$\varepsilon = \int_0^E S(E)(1-e^{-\nu\alpha m})dE$$

with

$$m = \int_0^E \frac{dE}{1+kB\frac{dE}{dx}}$$

$\nu\alpha$, free parameter, is the intrinsic efficiency of the detector (scintillator + PMT) in number of photoelectrons per keV

The knowledge of $\nu\alpha$ allows the calculation of ε

How to know $\nu\alpha$?

Traditional methods (*ca.* 1988)

- Using a radionuclide as a tracer, the CIEMAT/NIST method
- Using a LS counter with 3 PMT, the TDCR method

Recent developments (after 1995)

- Using a Compton source with a counter with 3 PMT, the CET method
- Using a LS counter with a photodetector giving the histogram of the number of photoelectrons, the HPMT method

The CIEMAT/NIST method

Measurement of a quenched set of ^{3}H standard with a commercial LS counter (2PMT)

$$\varepsilon_{H-3} = \int_0^E S(E)(1-e^{-\nu\alpha m})^2 dE$$

$$m = \int_0^E \frac{dE}{1+kB\frac{dE}{dx}}$$

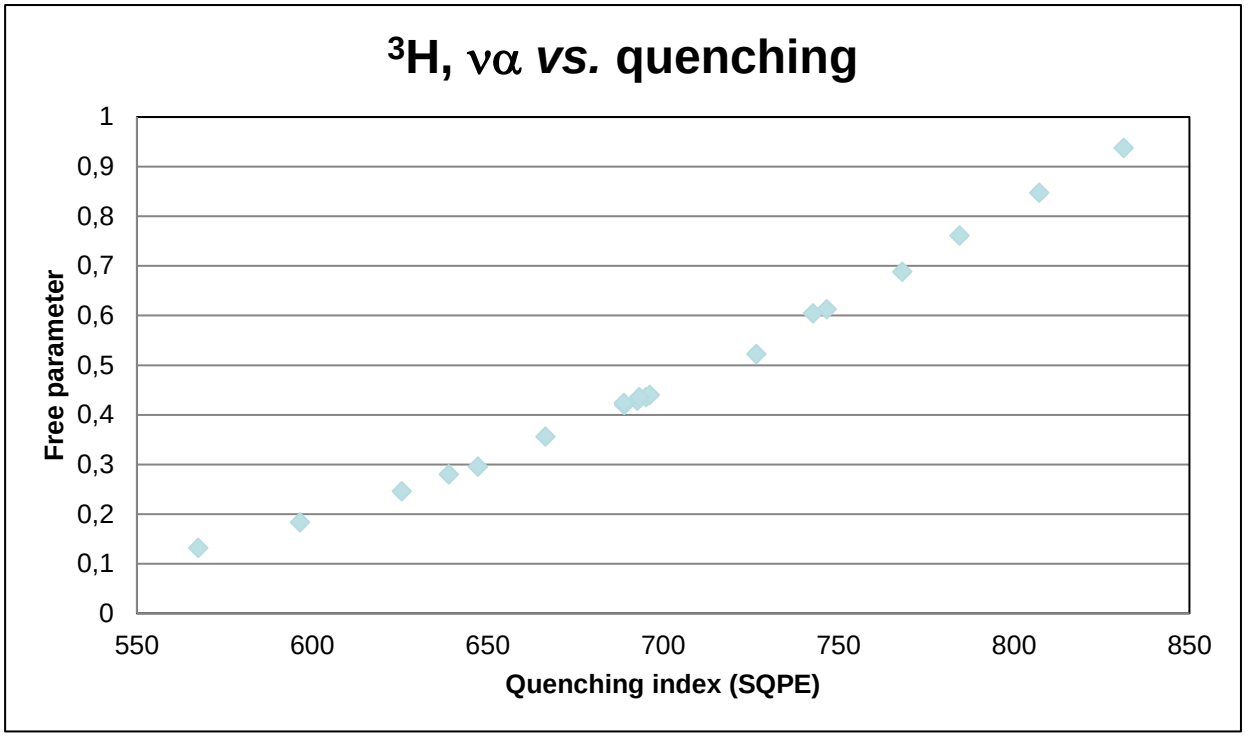
^{3}H detection efficiency

^{3}H spectrum

2 PMT in coincidence

ε is known (activity is known) and thus, the free parameter $\nu\alpha$ can be calculated

$\nu\alpha$ function of the quenching index



If the measurement conditions are kept identical (same vial, scintillator, counter, etc.) the value of this free parameter (function of quenching) can be used for the measurement of another radionuclide

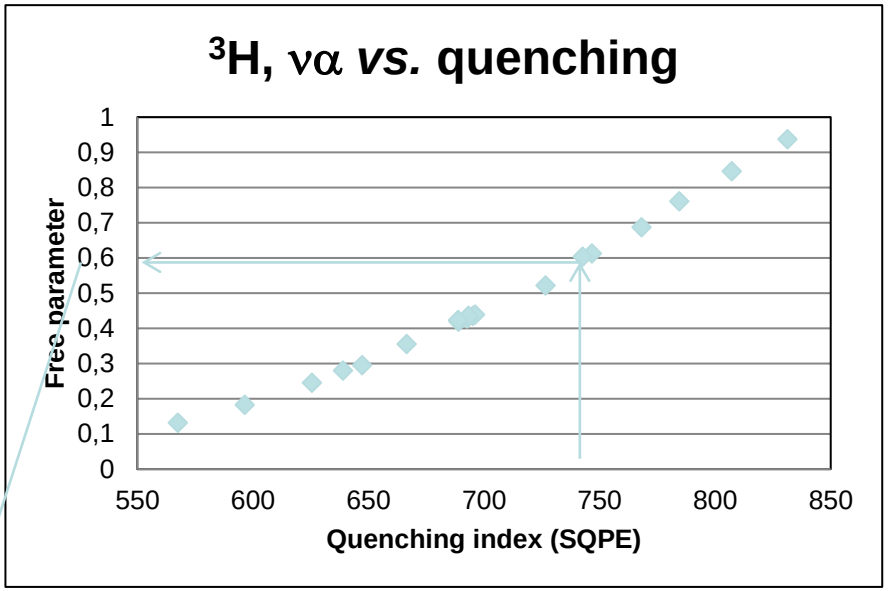
Measurement of radionuclide X

Make a LS source of radionuclide X

Measure the quenching of this source

Get the value of $\nu\alpha$ from the curve

Calculate the detection efficiency for the radionuclide X



$$\varepsilon_X = \int_0^E S(E)(1-e^{-\nu\alpha m})^2\,dE$$

$$m = \int_0^E \frac{dE}{1+kB\frac{dE}{dx}}$$

Spectrum of radionuclide X

The CIEMAT/NIST efficiency tracing method (CNET)

- Calculation of the free parameter using a ^{3}H standard source (for given quenching conditions)

- Use this free parameter to calculate the detection efficiency of any radionuclide… if you know its spectrum *S(E)*

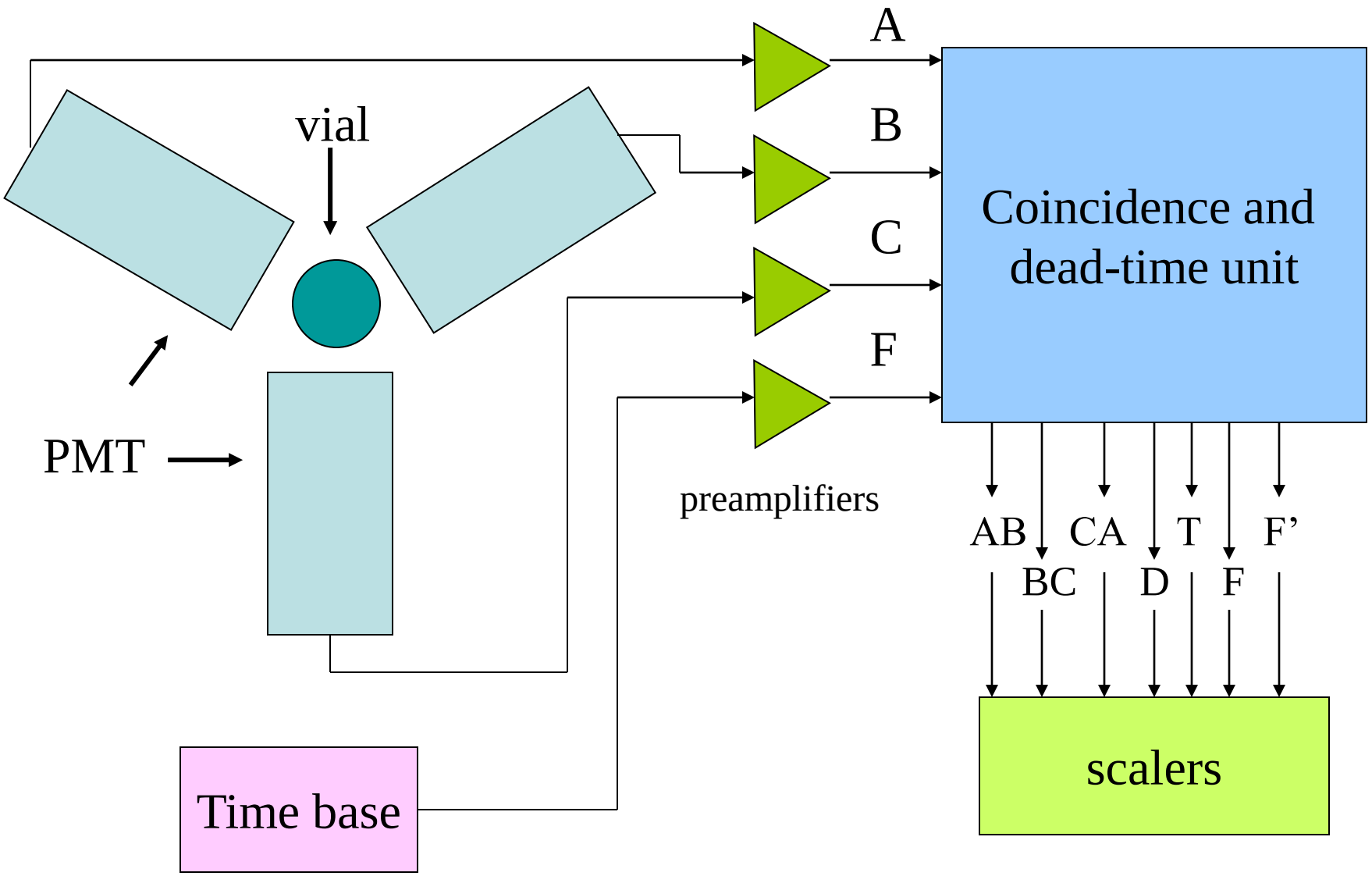
S(E) is the spectrum of the energy absorbed by the scintillator:

- For beta radionuclide, this is the beta spectrum
- For electron capture radionuclides, it must be calculated for each decay path
- For γ- or X-rays, it is calculated using Monte Carlo calculation methods (e.g. PENELOPE)

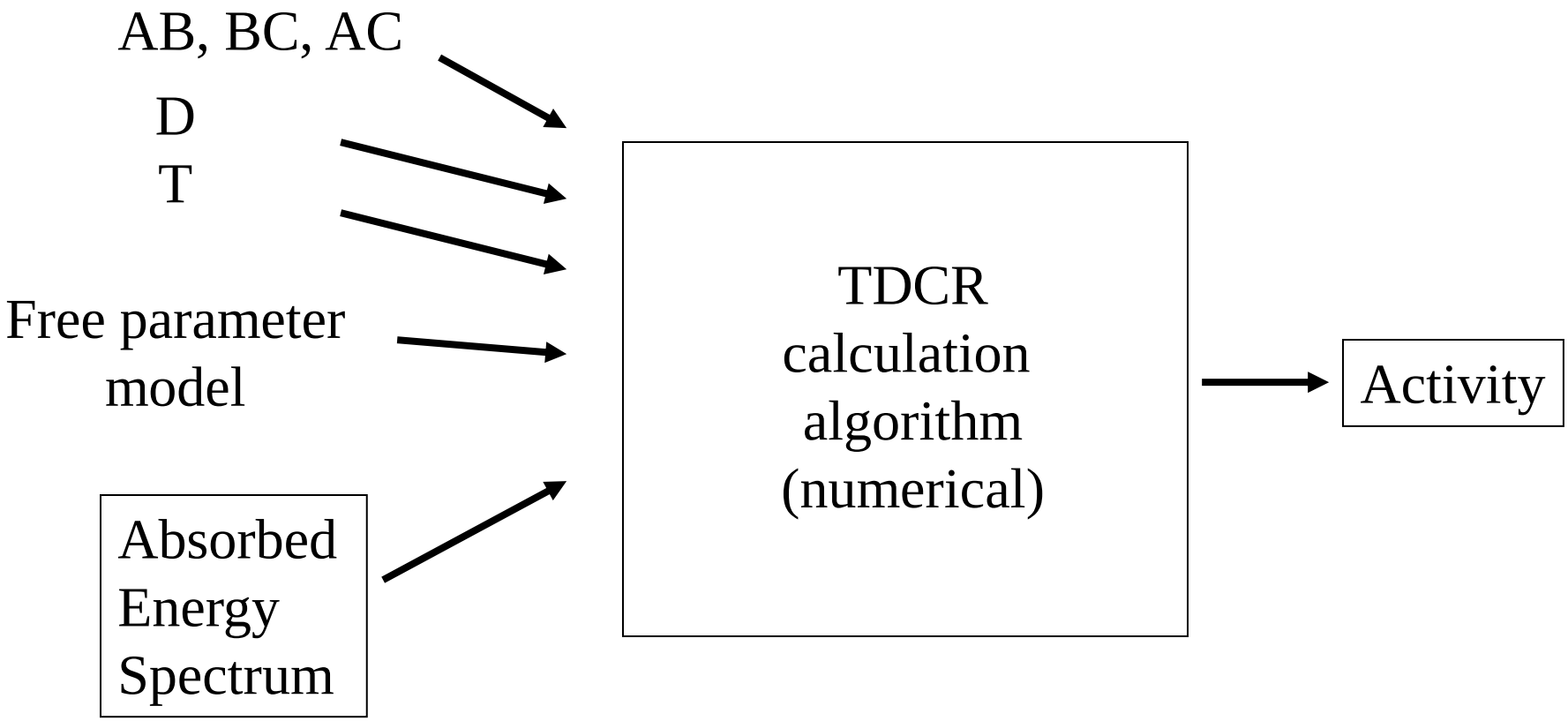
More details in: http://www.nucleide.org/ICRM_LSC_WG/icrmciematnist.htm

The Triple to Double Coincidence Ratio method (TDCR)

LSC TDCR Counter



The TDCR method in short



Radionuclide with normalized spectrum density *S(E)*

Events	Detection efficiency for *S(E)*
2 PMT's in coincidence	$\varepsilon_2 = \int_0^{E_{\max}} S(E)(1-e^{-\frac{\nu\alpha m}{3}})^2 dE$
3 PMT's in coincidence	$\varepsilon_T = \int_0^{E_{\max}} S(E)(1-e^{-\frac{\nu\alpha m}{3}})^3 dE$
Logical sum of double coincidences	$\varepsilon_D = \int_0^{E_{\max}} S(E)(3(1-e^{-\frac{\nu\alpha m}{3}})^2 - 2(1-e^{\frac{\nu\alpha m}{3}})^3) dE$

The ratio of triple to double detection efficiency is:

$$\frac{\varepsilon_T}{\varepsilon_D} = \frac{\int_0^{E_{\max}} S(E)\left(1-e^{-\frac{\nu\alpha m}{3}}\right)^3 dE}{\int_0^{E_{\max}} S(E)\left(3\left(1-e^{-\frac{\nu\alpha m}{3}}\right)^2 - 2\left(1-e^{-\frac{\nu\alpha m}{3}}\right)^3\right) dE}$$

with $$m = \int_0^E \frac{dE}{1+kB\frac{dE}{dx}}$$

For a large number of recorded events, the ratio of frequencies converges towards the ratio of probabilities:

$$\boxed{\frac{\boldsymbol{T}}{\boldsymbol{D}} = \frac{\varepsilon_T}{\varepsilon_D} = \boldsymbol{TDCR}}$$

Resolution algorithm:

Find a value of the free parameter ($\nu\alpha$) giving:

$\varepsilon_T/\varepsilon_D$ calculated = T/D experimental

How many solutions ?

- Monoenergetic electrons: 1 analytical solution
- Pure-beta radionuclides: 1 solution
- Beta-gamma, electron capture: up to 3 solutions...

Detection efficiency (single energy)

Similar PMT's:

Analytical solution

$$\varepsilon D = \frac{27(TDCR)^2}{(1+2(TDCR))^3}$$

PMT's with different quantum efficiencies:

$$m\nu_A = -3Ln(1-\frac{T}{BC})$$

a.s.o. for ν_B and ν_C

$$\varepsilon D = T^2(\frac{1}{BC\cdot AC}+\frac{1}{AC\cdot AB}+\frac{1}{AB\cdot BC}-2\frac{T}{AB\cdot BC\cdot AC})$$

Detection efficiency (multiple energies)

Normalized energy spectrum *S(E)*

Numerical solution: find out νA (*fom*) to solve:

$$TDCR = \frac{\int_{spectrum} S(E)(1-e^{-\frac{\nu\alpha m}{3}})^3 dE}{\int_{spectrum} S(E)((3(1-e^{-\frac{\nu\alpha m}{3}})^2 - 2(1-e^{-\frac{\nu\alpha m}{3}})^3))dE}$$

with

$$m(E) = \int_0^E \frac{dE}{1 + kB\frac{dE}{dx}}$$

If the 3 PMT 's are different (and they generally are!)

$$\frac{\varepsilon_T}{\varepsilon_{AB}} = \frac{\int_0^{E_{\max}} S(E)(1-e^{-\frac{\nu_A \alpha m}{3}})(1-e^{-\frac{\nu_B \alpha m}{3}})(1-e^{-\frac{\nu_C \alpha m}{3}})dE}{\int_0^{E_{\max}} S(E)(1-e^{-\frac{\nu_A \alpha m}{3}})(1-e^{-\frac{\nu_B \alpha m}{3}})dE}$$

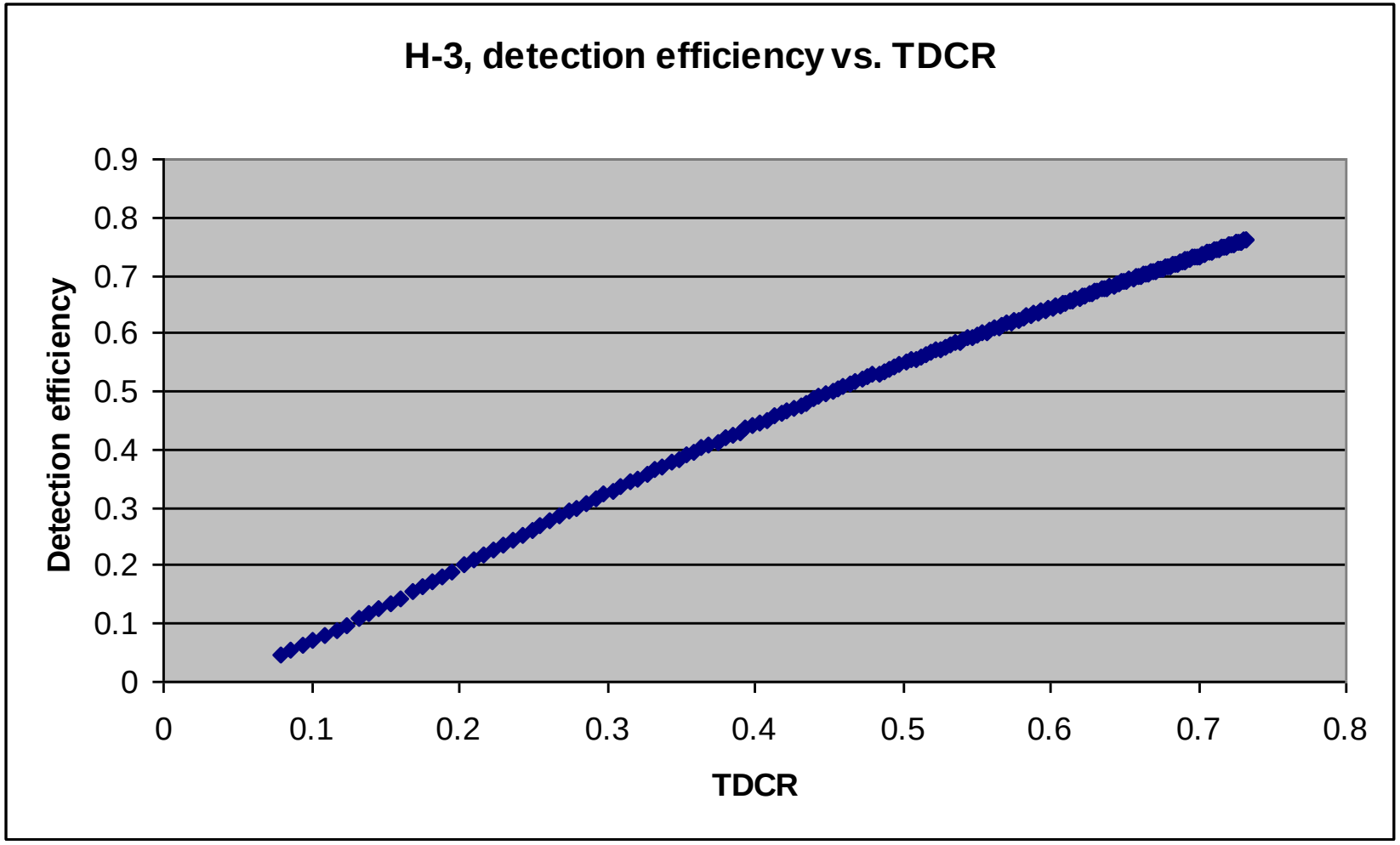
a.s.o. for $\frac{\varepsilon_T}{\varepsilon_{BC}}$ and $\frac{\varepsilon_T}{\varepsilon_{AC}}$

Solution, minimize:

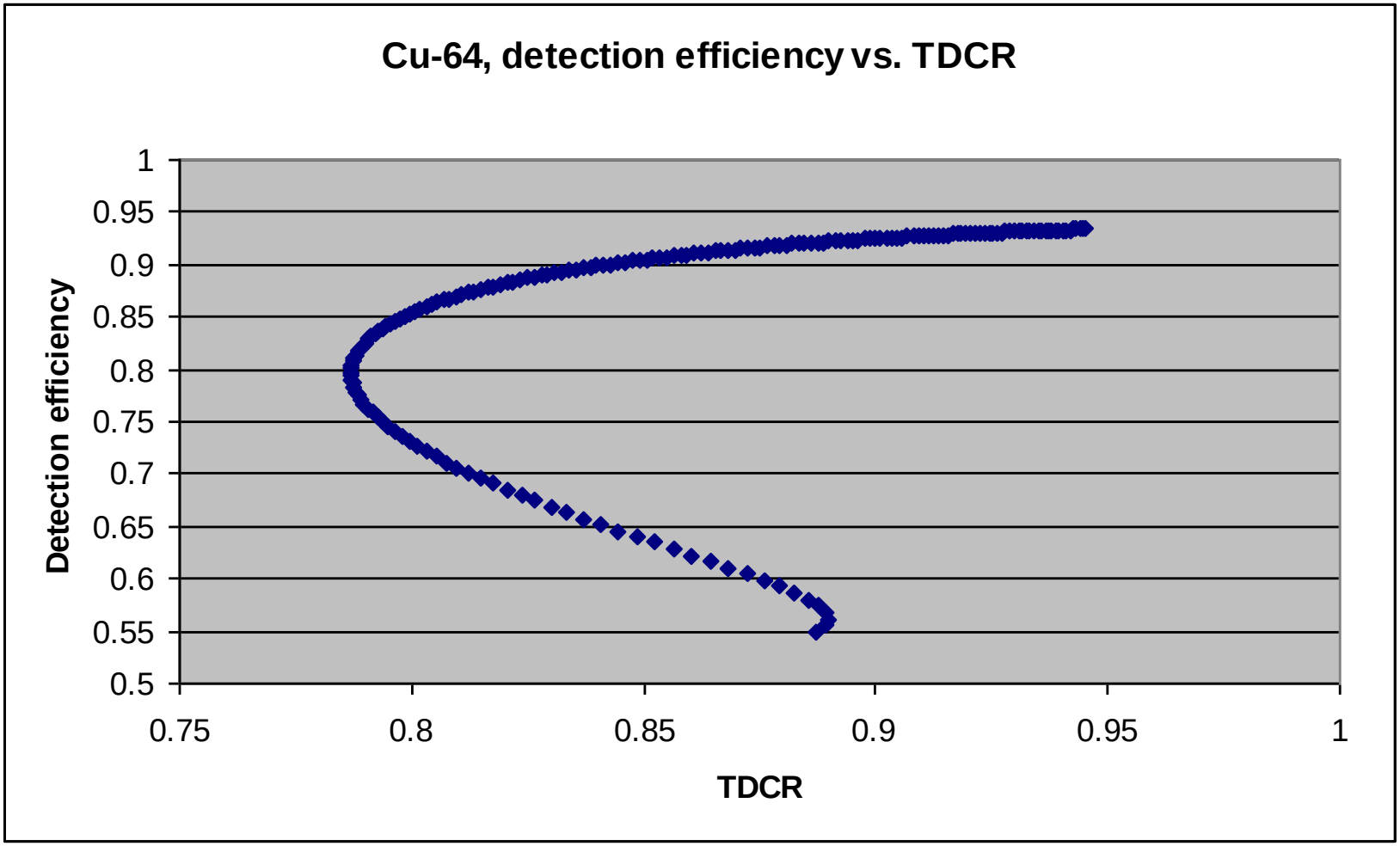
$$\left(\frac{T}{AB}-\frac{\varepsilon_T}{\varepsilon_{AB}}\right)^2+\left(\frac{T}{BC}-\frac{\varepsilon_T}{\varepsilon_{BC}}\right)^2+\left(\frac{T}{AC}-\frac{\varepsilon_T}{\varepsilon_{Ac}}\right)^2$$

This gives the detection efficiency and free parameter for of each PMT

Example of calculation, ^{3}H



More complicated example, ^{64}Cu (β^+, β^-, e.c.)

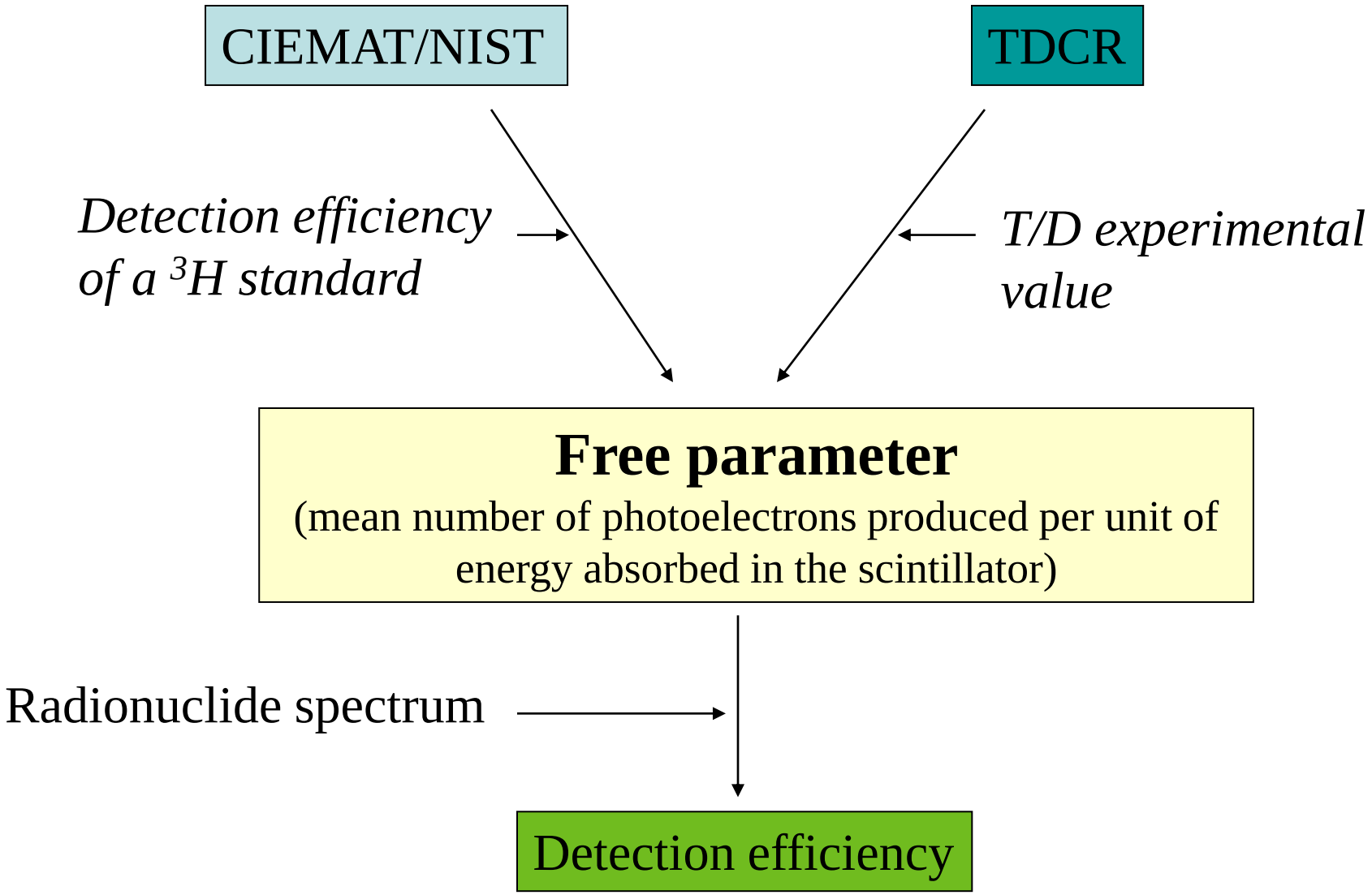


Typical TDCR uncertainty budget

Uncertainty component	**Relative uncertainty (k=1)**
Weighing	~ 0.1 %
Counting statistics	ALARA (e.g. 0.1 %)
Background	ALARA (e.g. 0.01 %)
Detection efficiency	0.1 % - 1 % function of *E*
Sources variability	Generally ~ 0.2 %
Total	From a few 0.1 % to a few %

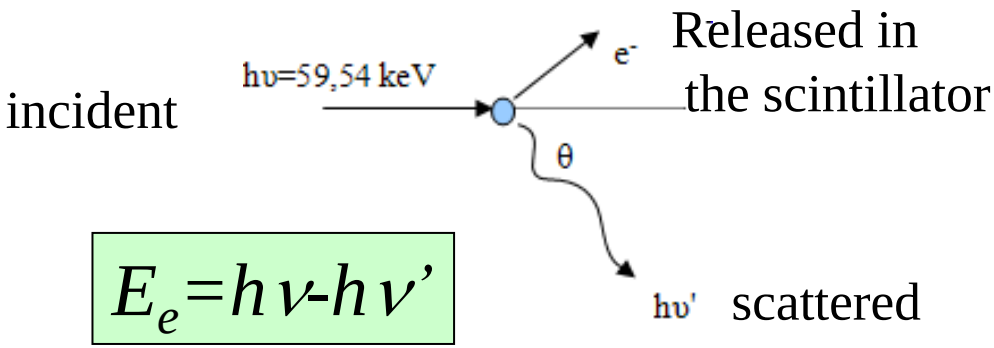
The Compton Efficiency Tracing method (CET)

LSC radionuclide standardization methods



Alternative: efficiency tracing with a virtual radionuclide

Principle: produce a temporary electron source inside a LS vial using the Compton interaction



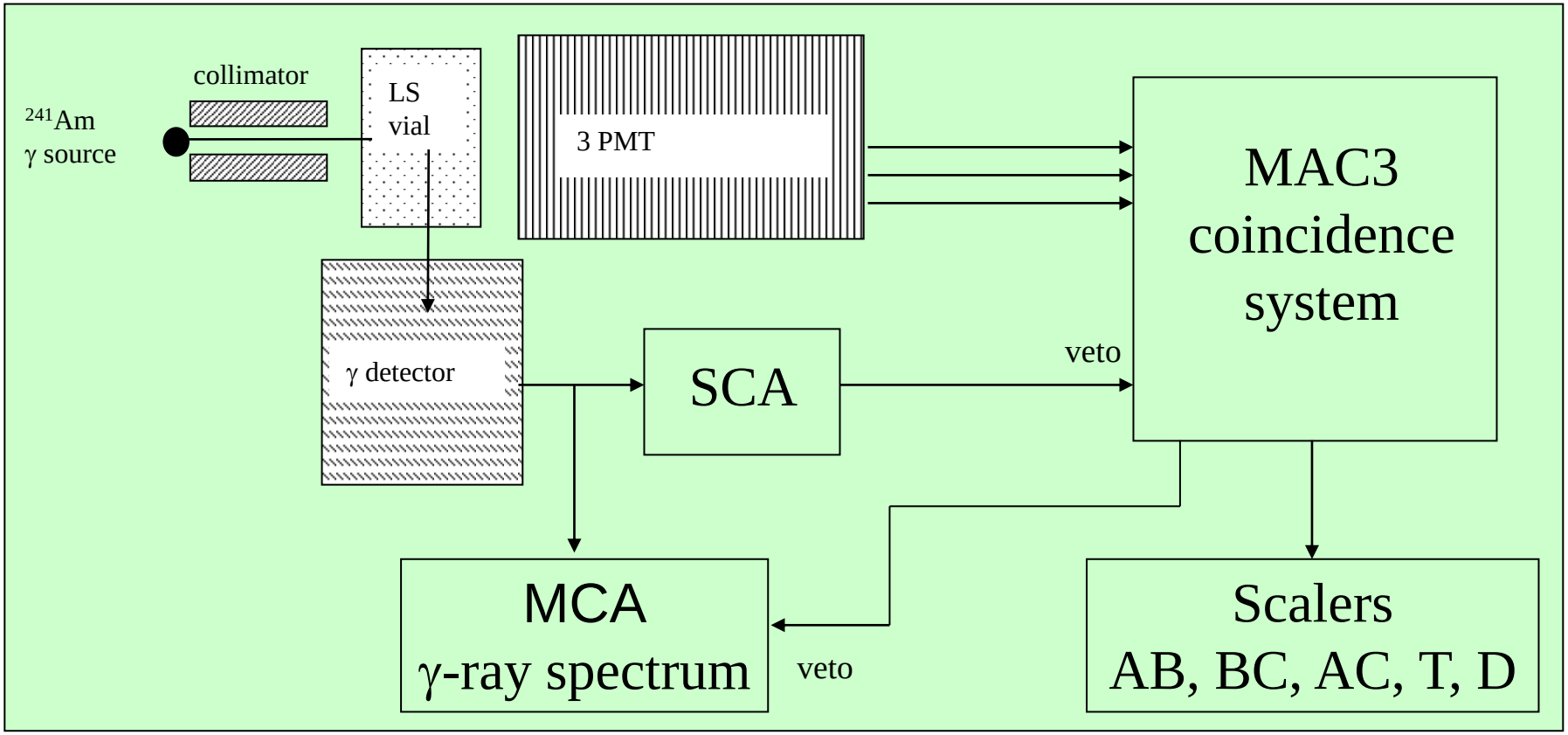
$$E_e = h\nu - h\nu'$$

This source is switchable and its energy spectrum is measurable (can be either monoenergetic or polyenergetic)

Experimental setup

External 59.5 keV γ source (filtered ^{241}Am solid source)

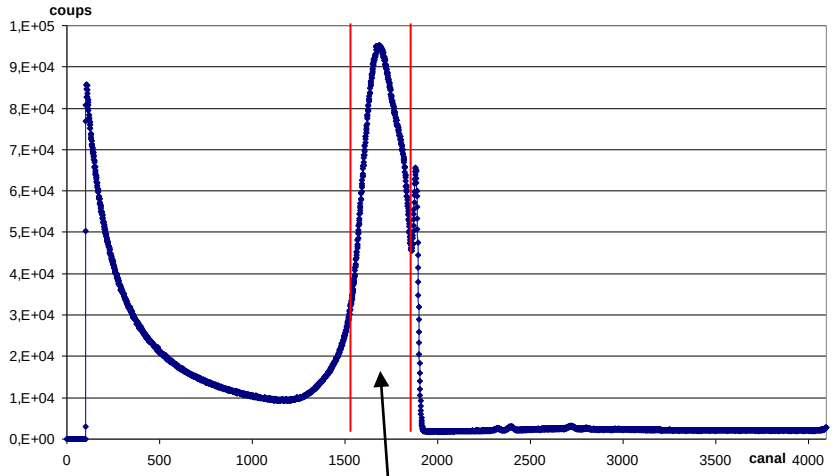
- γ-ray detector with calibrated energy
- Coincident measurement (rejection of other electron sources)
- Energy of the electron source controlled by energy selection of the diffused Compton photon spectrum



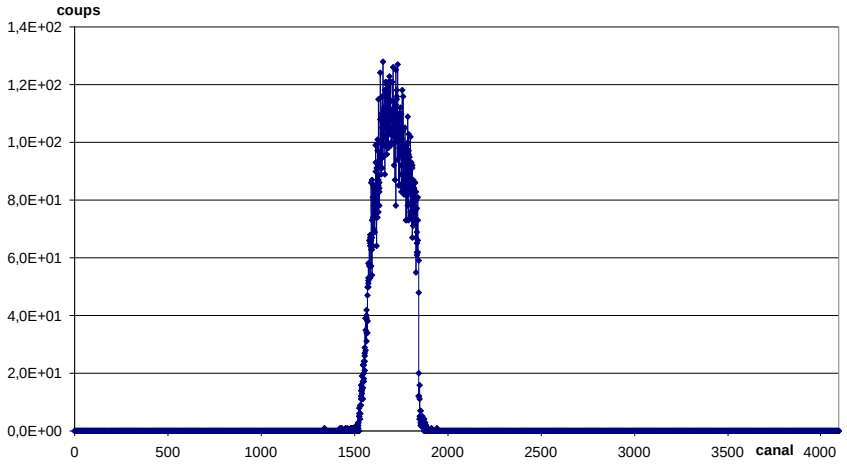
Internal low-energy tracer source

The geometry of the system defines the possible detectable Compton diffused photon

Experimental photon spectrum

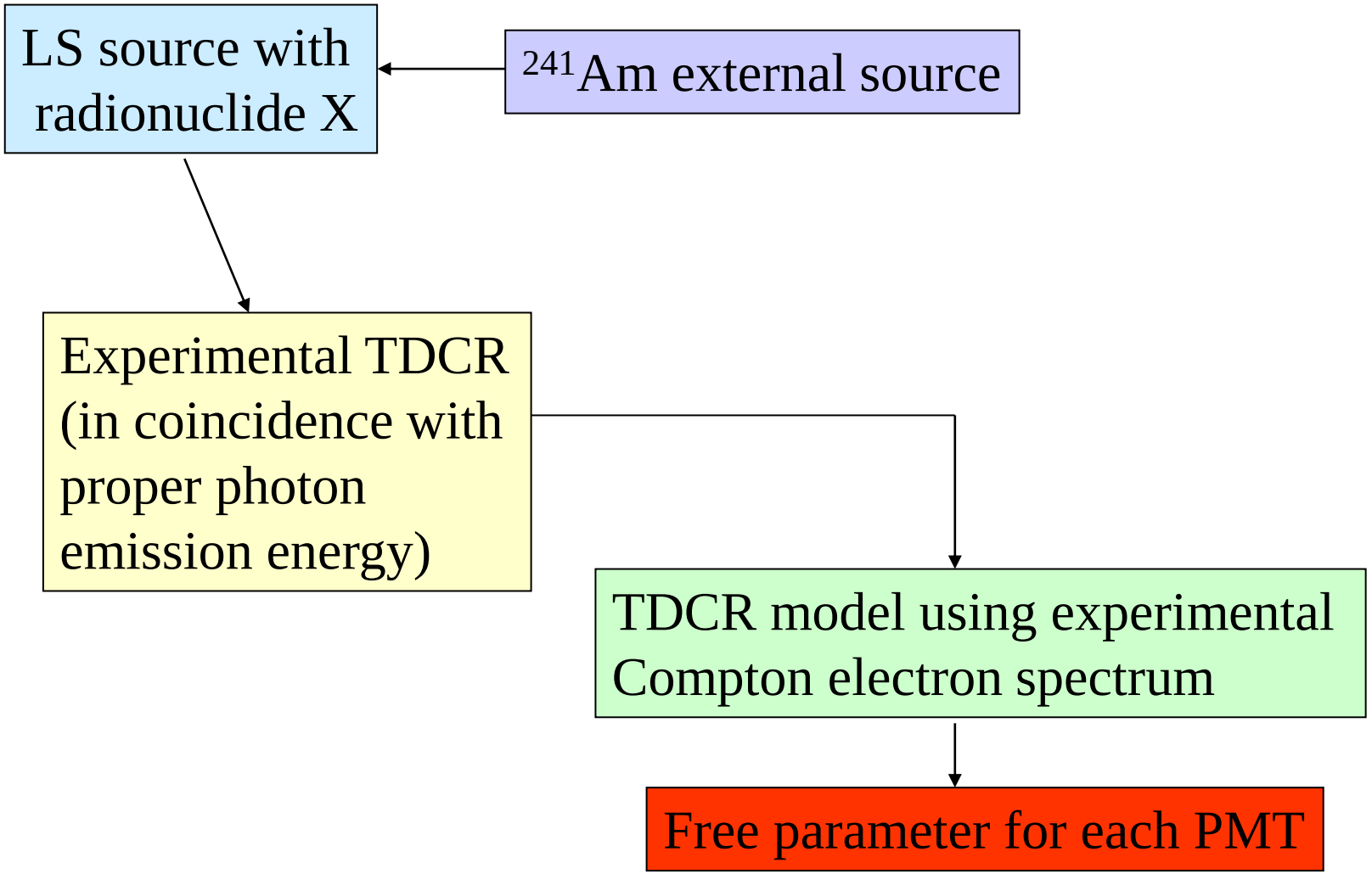


Selected spectrum

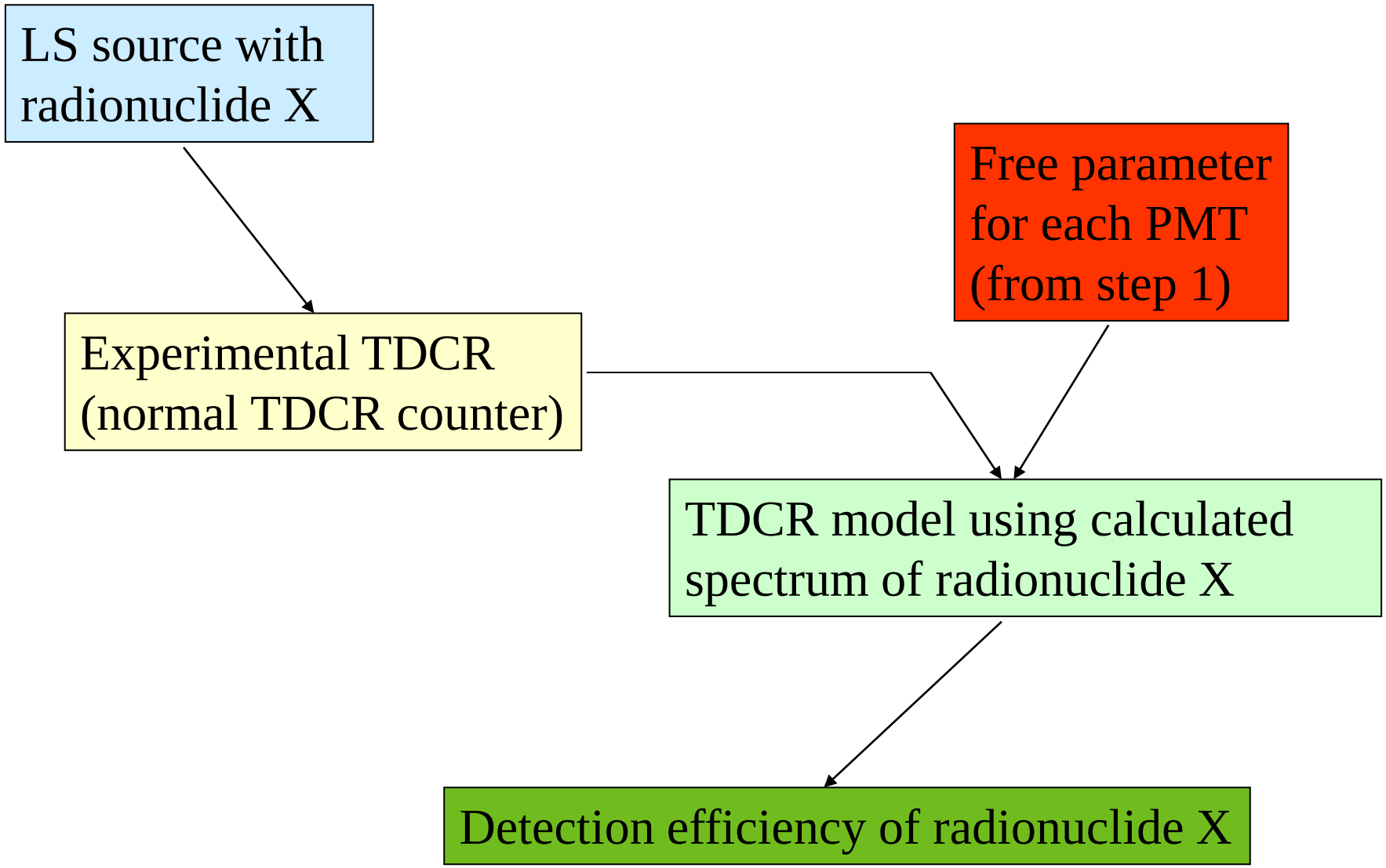


The useful portion of this spectrum is selected using the SCA

Compton source tracer method, step 1



Compton source tracer method, step 2



The CET is, in principle, similar to the CIEMAT/NIST method but:

- Free parameter deduced from the real source to measure (no quenching parameter to consider)
- Tracer from internal source created *in situ* by Compton effect
- The counting rate of the Compton source does not matter
- The spectrum of the tracer source is only defined by the geometry of the detector and can be measured
- The detection efficiency can also be calculated using the traditional TDCR model with the same data set

Recent developments of the Compton efficiency tracing method

Cooperation between the Sofia University and Laboratoire National Henri Becquerel: Krasimir Mitev, Chavdar Dutsov, Benoit Sabot

Development of a miniature 3D-printed Compton TDCR spectrometer with list-mode digital acquisition

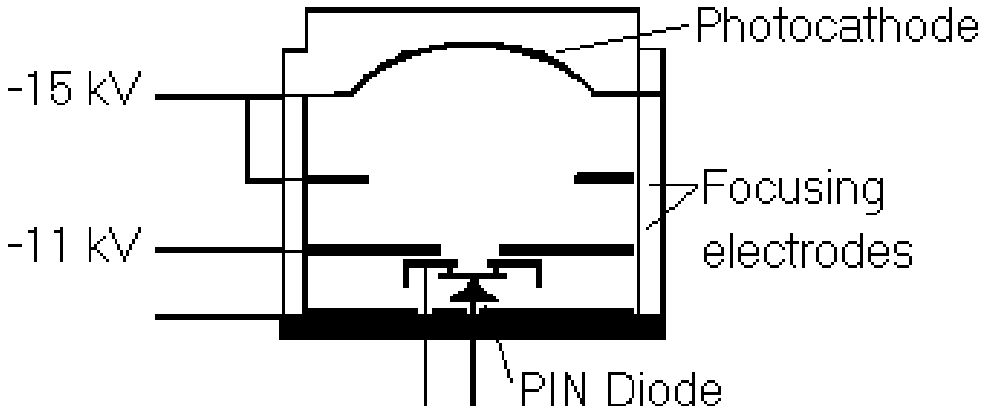
Advantages:

- no direct interaction between the excitation source and the PMTs
- Reduction of the acquisition time (many energies covered at the same time)

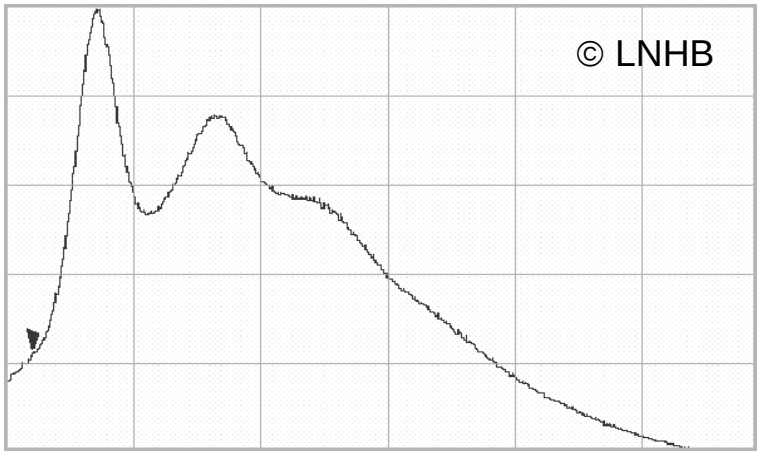
- A common publication is coming soon…

Other method in LSC using high-resolution photodetectors

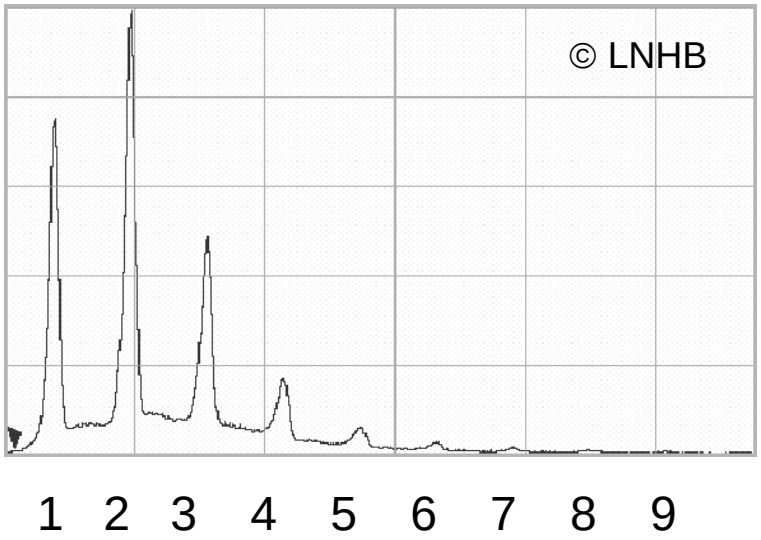
HPMT



Resolution of photodetectors



Traditional PMT (a very good one!)



Hybrid PMT

Analysis of the PMT spectrum

Convolution of:

- single photoelectron spectrum
- Radionuclide spectrum
- Statistical distribution of the number of photons
- Backscattering

The statistical distribution of photons can be calculated by deconvolution

This statistical distribution allows the calculation of the free parameter

More details in: http://www.nucleide.org/ICRM_LSC_WG/icrmhpmt.htm

Towards a no-parameter approach, the ZoMBieS method (L. Bignell, ANSTO)

Zer**o** **M**odel **B**y using Co**i**ncidenc**e** **S**cintillation

- 3 PMT detector with Compton spectrometer
- Compton spectrometer used to generate monoenergetic Compton electrons in the LS source, with variable energy

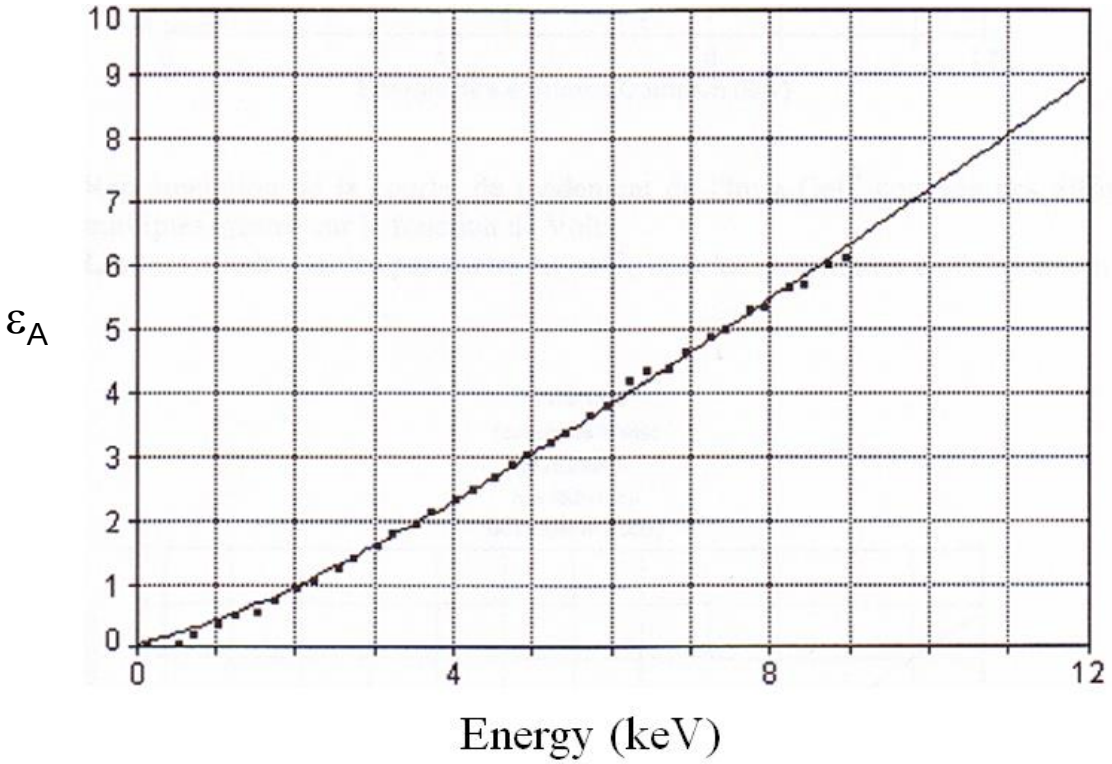
Ratio of detection efficiency in triple and double coincidence:

$$\frac{T}{AB} = \frac{\varepsilon_A \cdot \varepsilon_B \cdot \varepsilon_C}{\varepsilon_A \cdot \varepsilon_B} = \varepsilon_C$$

Idem for BC and AC

By varying the energy, one can plot ε_A, ε_B and ε_C *vs.* the energy

ZoMBieS method



$$\varepsilon_T = \int_0^E S(E) \cdot \varepsilon_A(E) \cdot \varepsilon_B(E) \cdot \varepsilon_C(E) dE$$

The detection efficiency can be calculated *without* free parameter model!

ZoMBieS method

Advantage:

- No-parameter model, no standard, no calibrated tracer
- Minimum model assumptions (no Birks equation, no Poisson law)

Drawbacks:

- Complicated experimental system (monoenergetic Compton electrons selection difficult to adjust)
- Very long acquisition time

Future developments:

- Optimized counter with higher efficiency in γ channel

Conclusions for LSC

The free parameter model is the main tool for the use of LSC in radionuclide metrology (TDCR, CIEMAT/NIST, efficiency tracing techniques)

- **Some open issues still remain:**
  - Optimal statistical model
  - Calculation of the scintillator non-linearity
  - Calculation of detection efficiency in coincidence when there is a correlation between PMT signals
  - Cerenkov light emission model
  - Light propagation anisotropy model (color quenching)

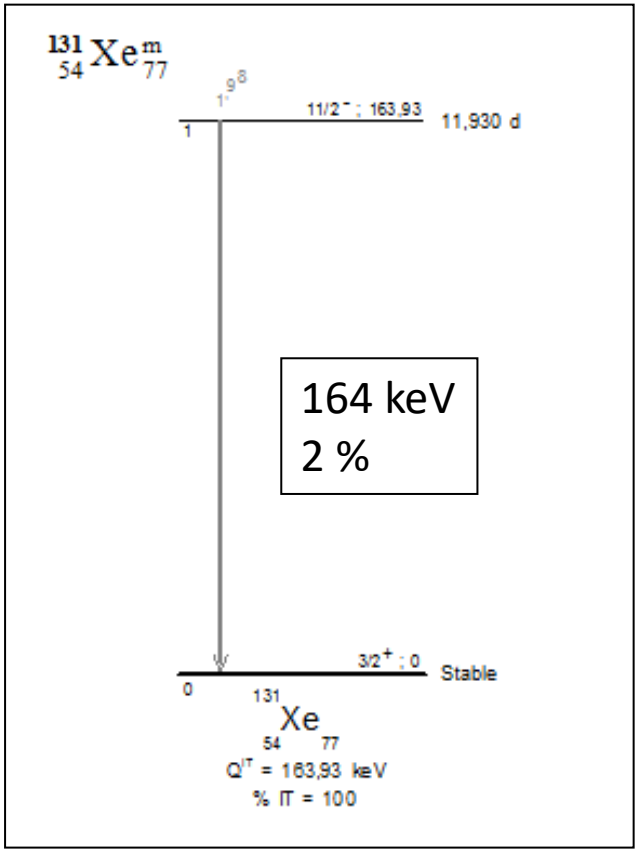
- **New developments are pending:**
  - Monte Carlo simulation including light propagation and detection
  - Experimental study of scintillator non-linearity

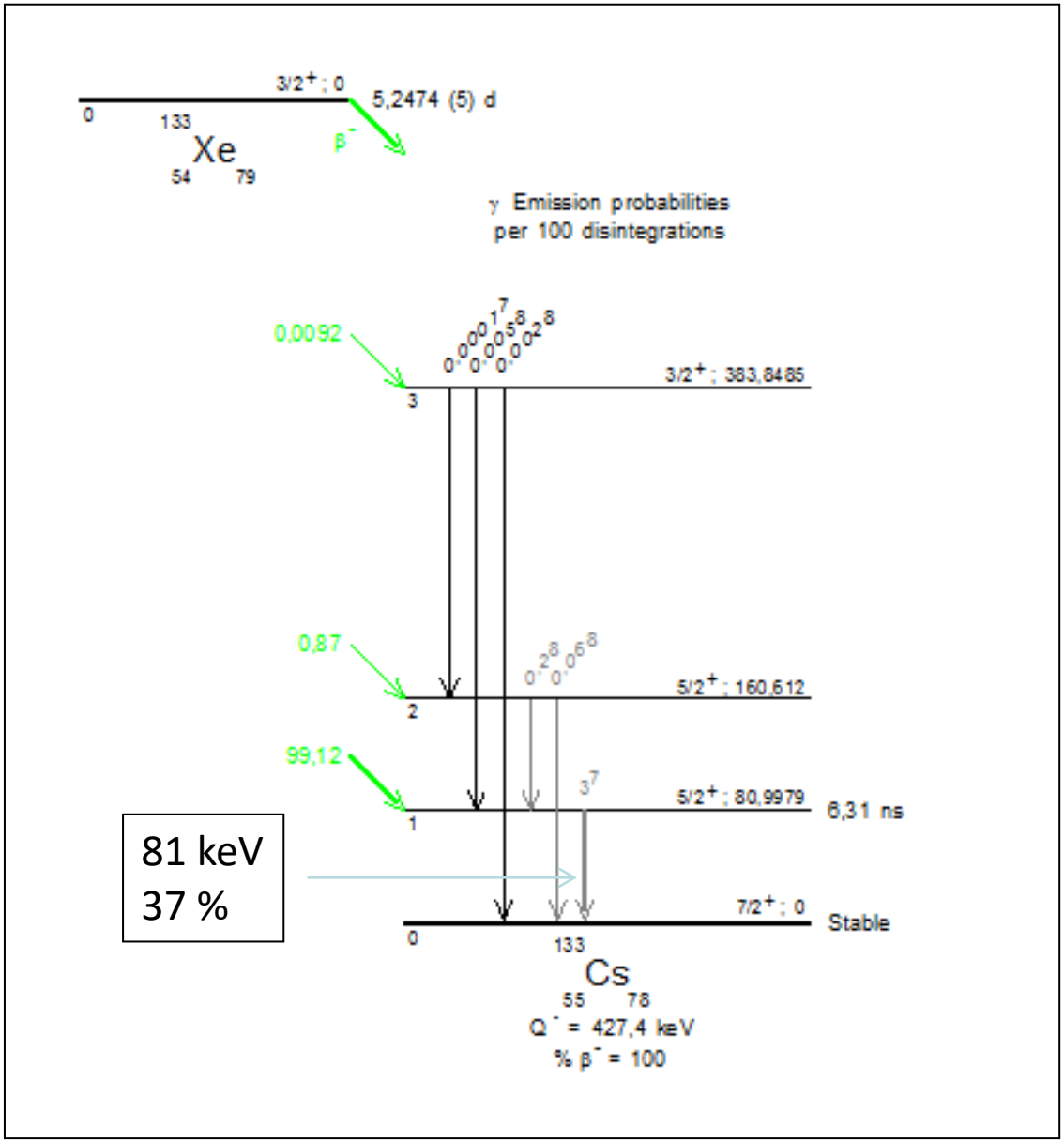
The free parameter model will remain the cornerstone of the use of LSC in radionuclide metrology… until the proof that the no-model approach is mature

Primary Standardisation of activity

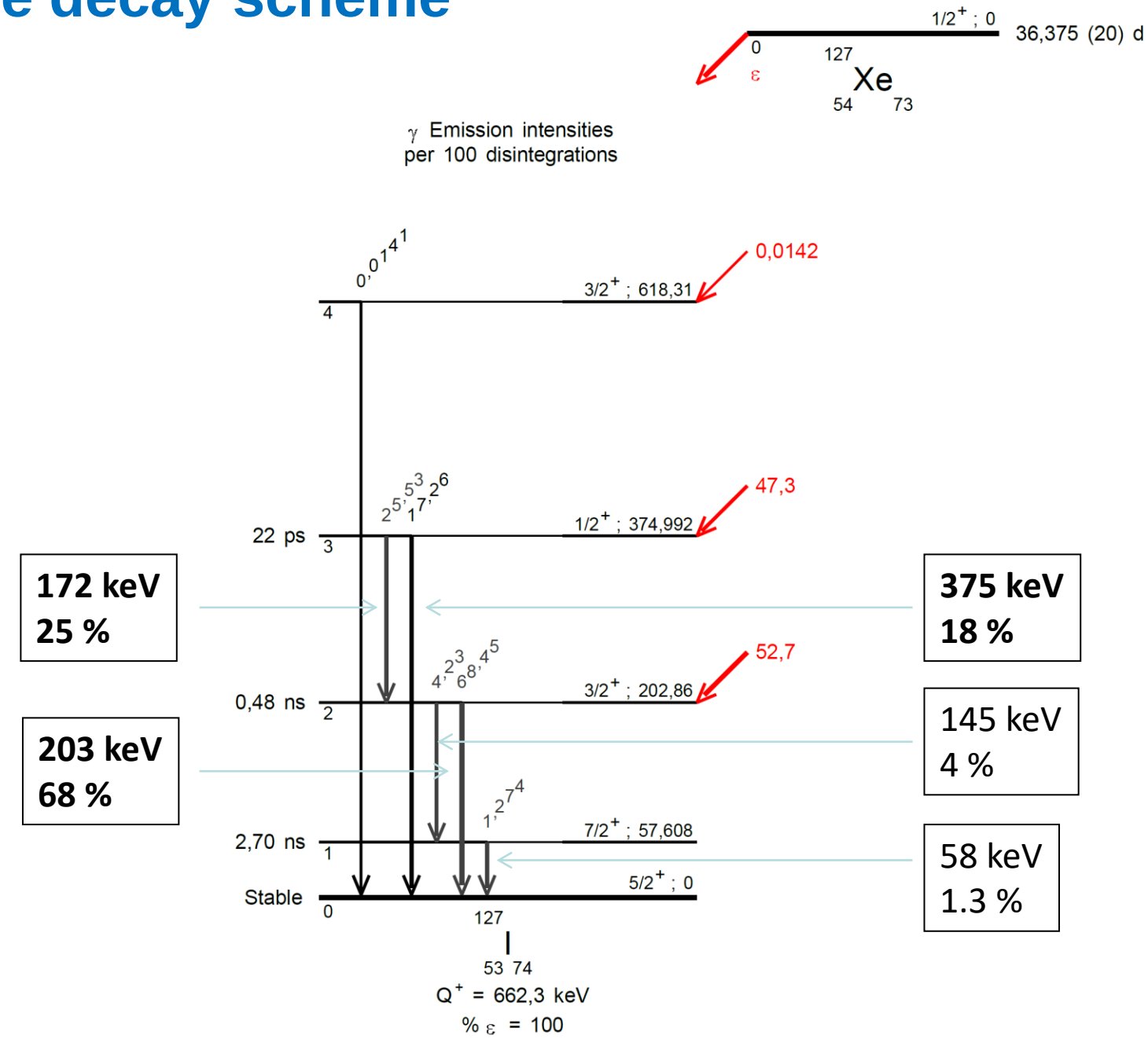
Radioactive gas (noble gas and tritium)

^{131m}Xe and ^{133}Xe decay schemes





^{127}Xe decay scheme



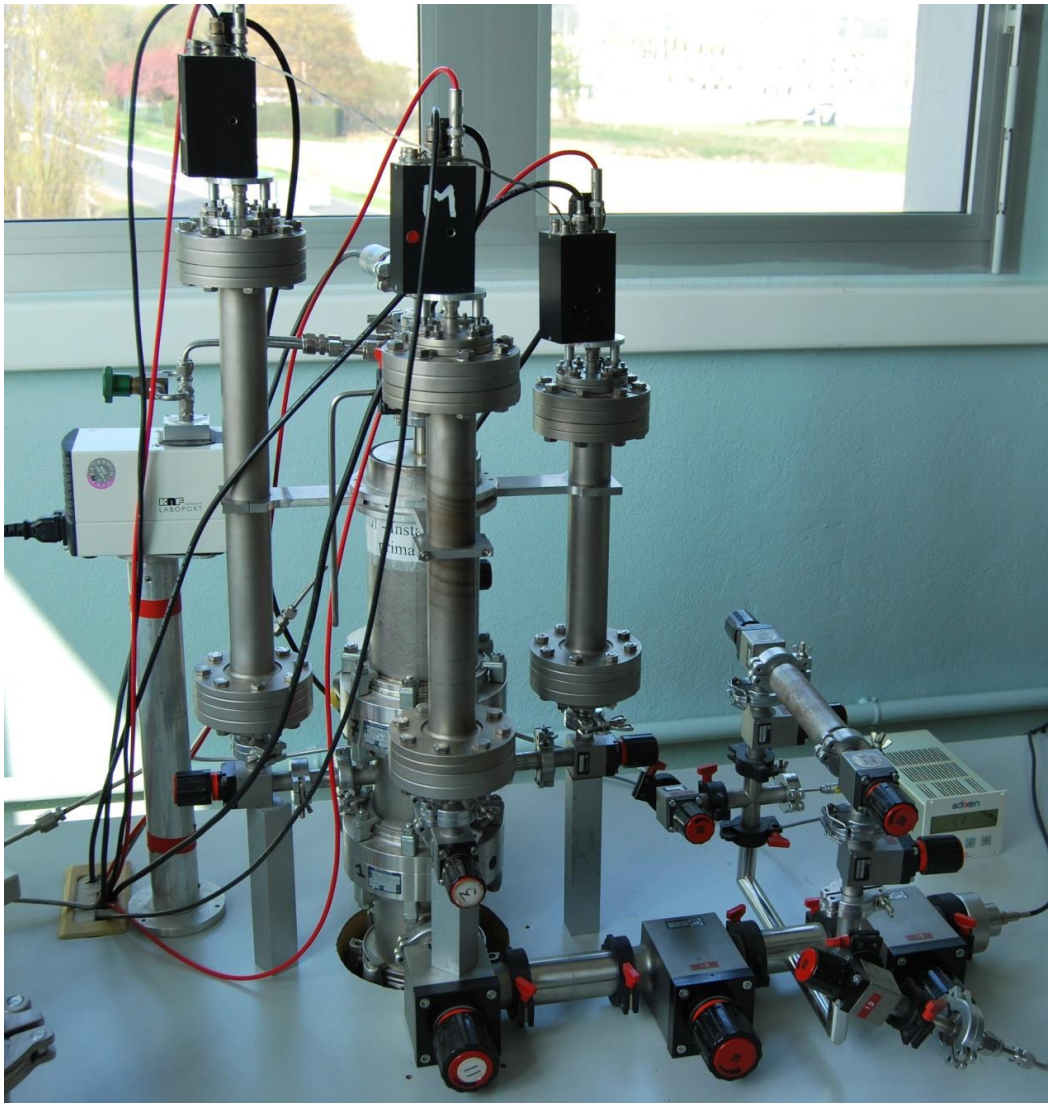
Example: Standardization of ^{127}Xe

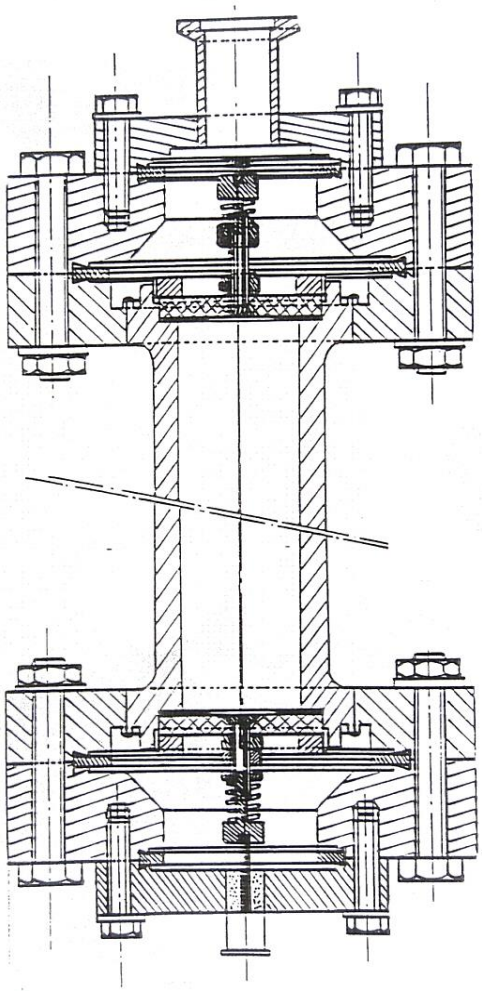
Internal gas counting system:

- 3 proportional counters, identically constructed but with different lengths

- counting gas: propane

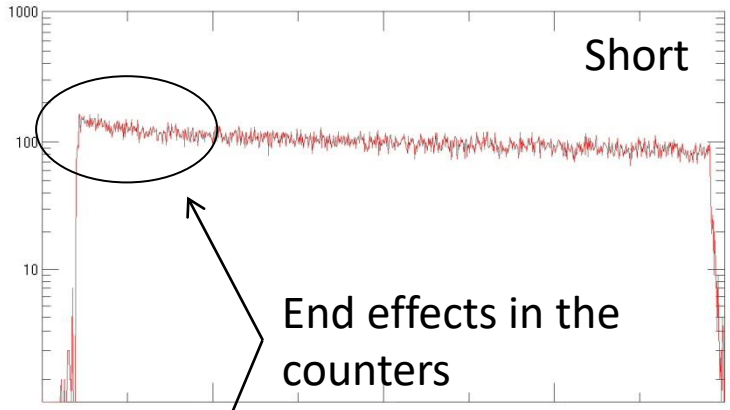
Counters



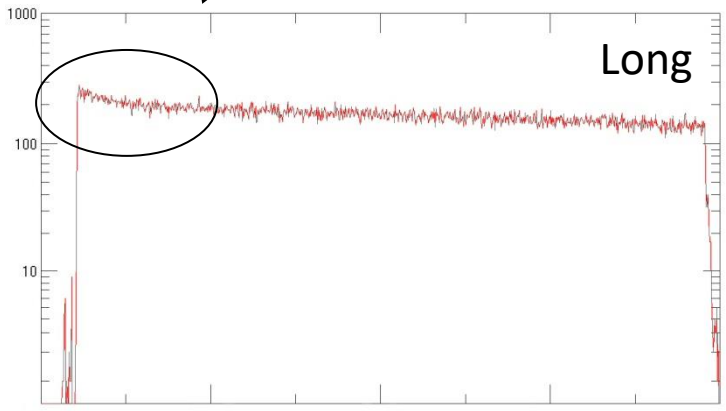


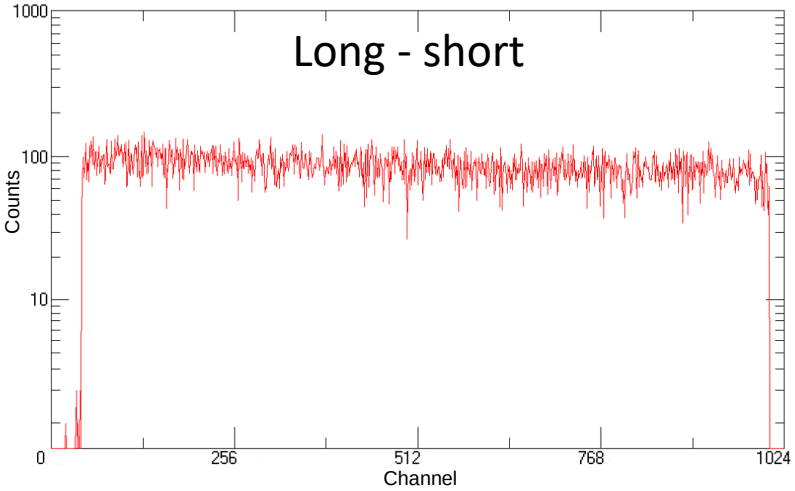
Differential measurement

Long – short ≈ virtual cylindrical counter without end effects



End effects in the counters





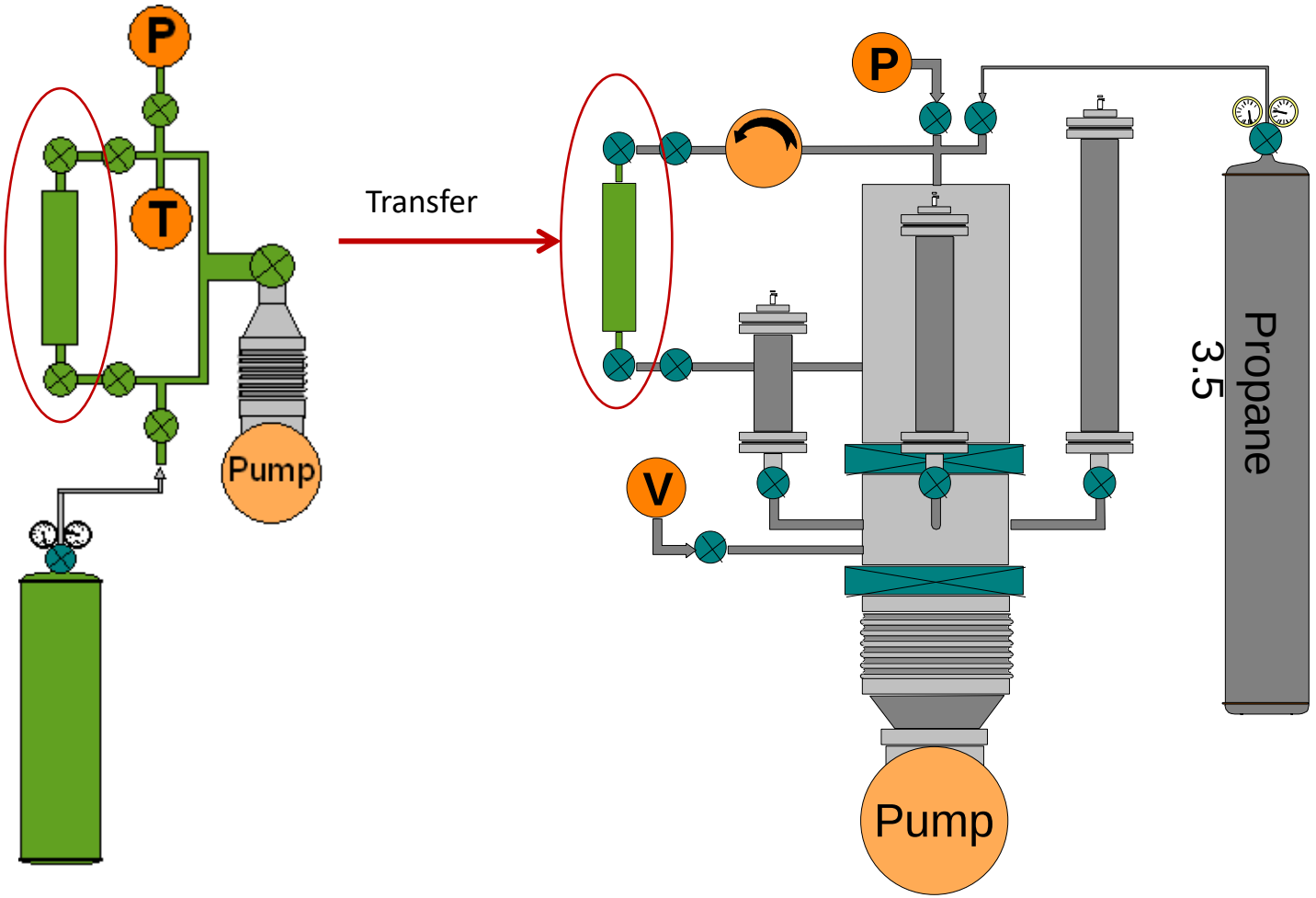
Measurement method

Sample preparation

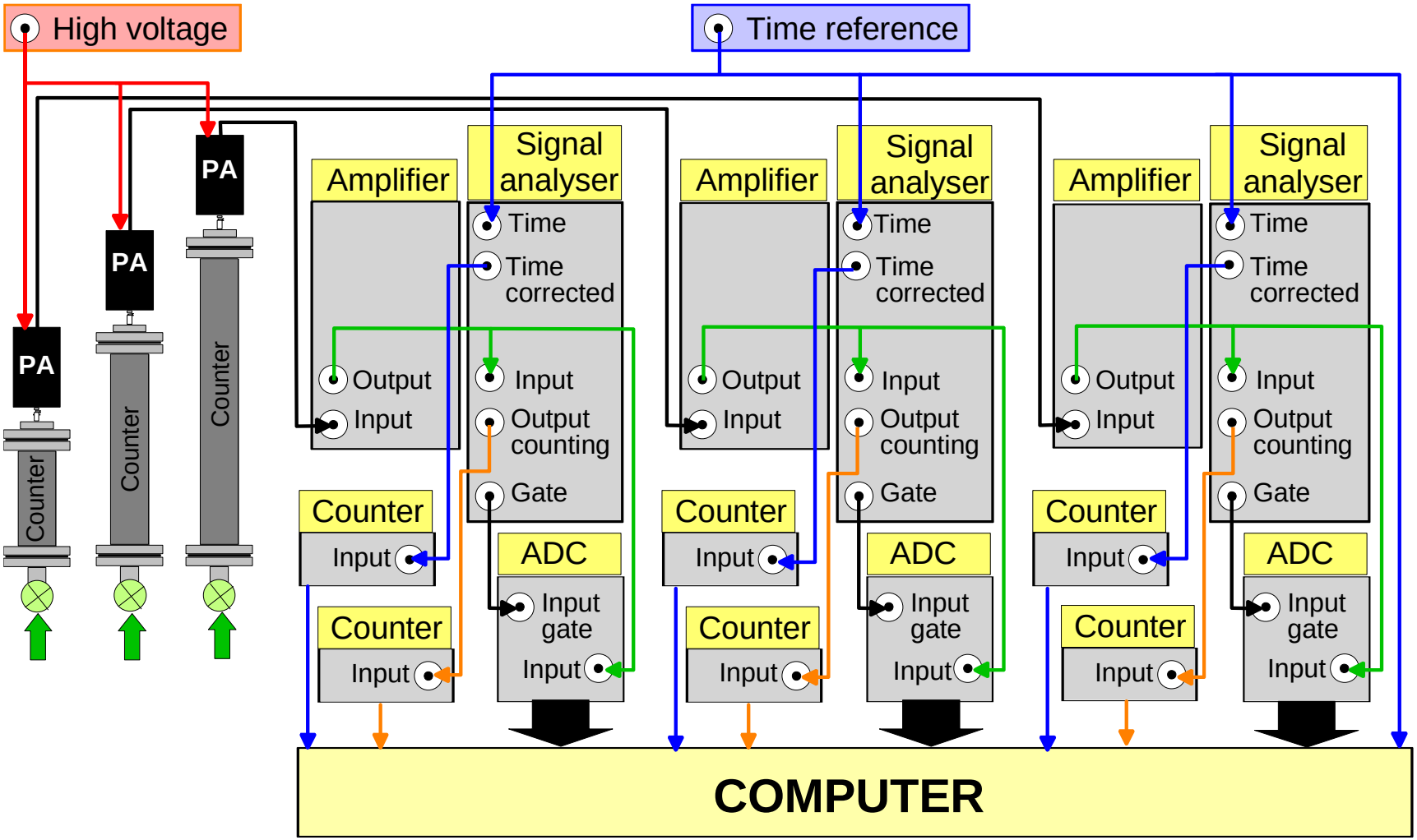
Sample measurement

Sampling volume
119.3 cm^3

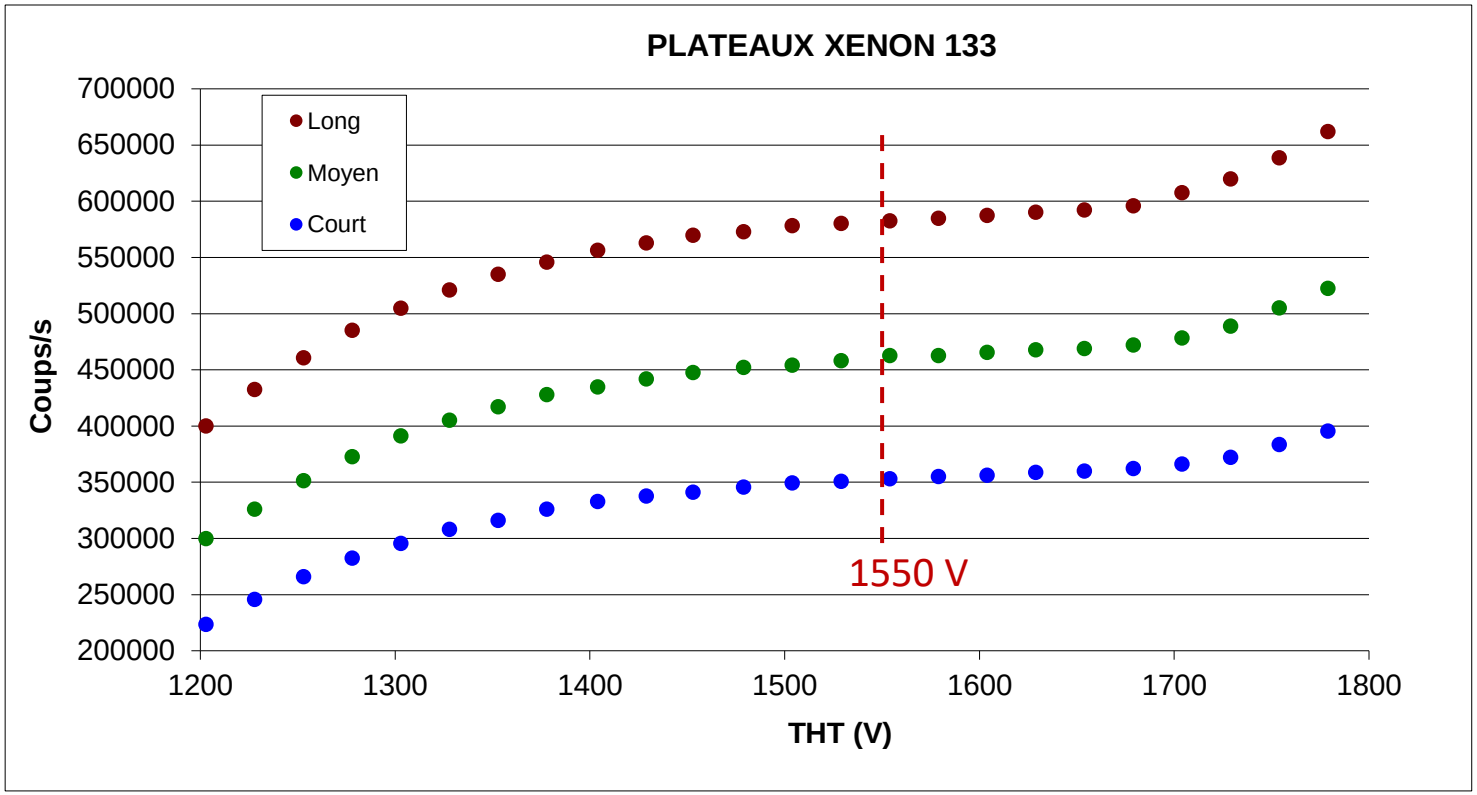
Active gas



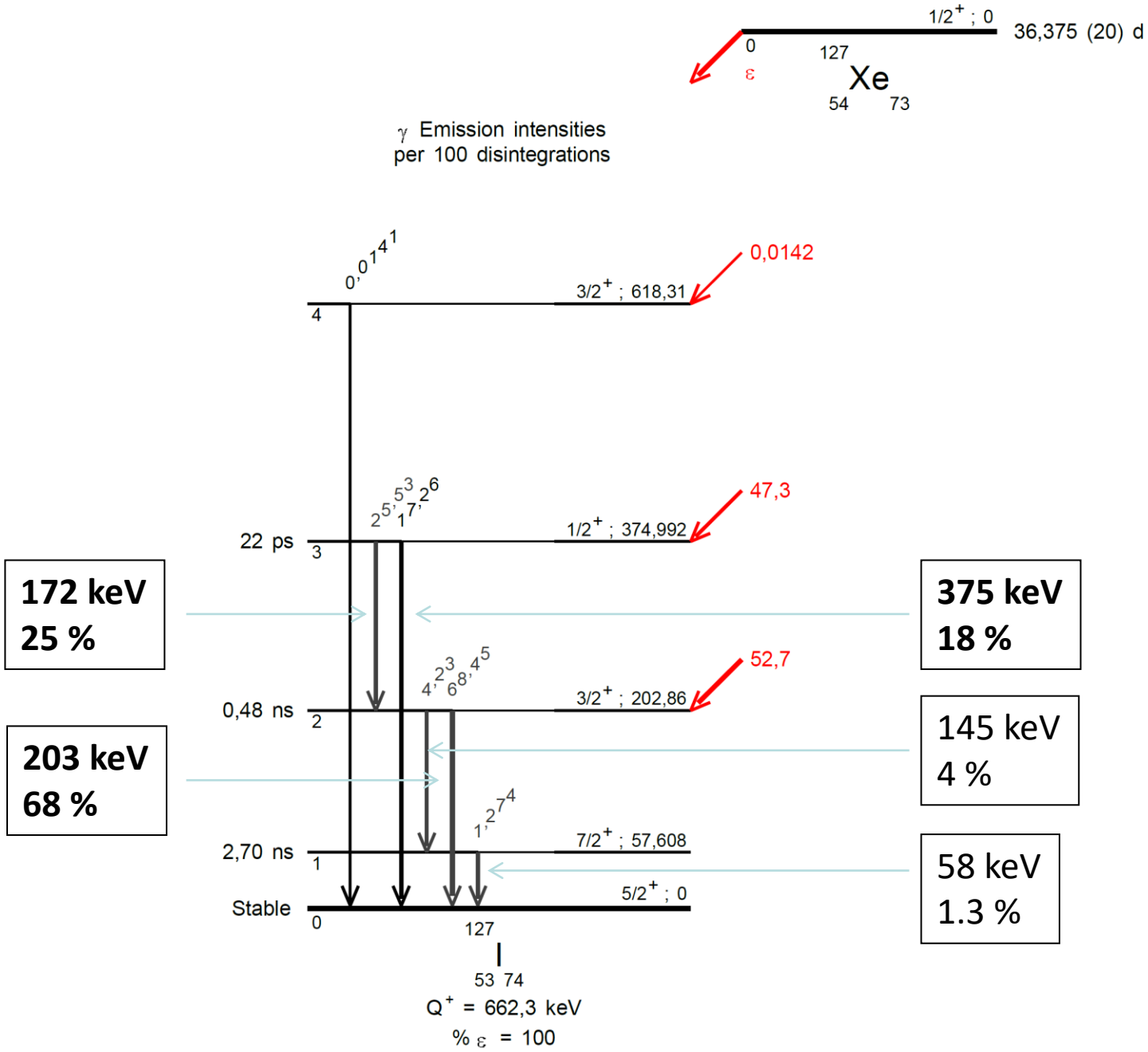
Data acquisition system



Optimum operating voltage



Calculation of detection efficiency



Calculation of detection efficiency 1

- Energy threshold: 200 eV (calibrated using a ^{37}Ar source with 200 eV Auger electrons)

- Imposed extending-type dead-time (50 μs) to mitigate the effect of saturated pulses

- Live-time measured with a live-time clock (no calculated dead-time correction)

- Detection efficiency = 1-probability of undetected radiations

- Atomic rearrangement using detailed atomic levels: L1 to L3, M1 to M5, N1 to N5, O subshells considered

Calculation of detection efficiency 2

- Electron vacancy in N and O shells: maximum Auger electrons energy of 136 eV. ε_N=0
- Electron vacancies in M shells. ε_M=1
- Electron vacancy in L shells. No detection of some Coster-Kronig transitions E<200 eV. ε_L= 0.989 for L3, 0.980 for L2 and 0.982 for L1
- Electron vacancy in K shell. ε_K=0.962

→ Detection efficiency of electron capture: 0.960

Calculation of detection efficiency 3

Gamma and X-rays emissions:
Monte Carlo simulation, PENELOPE code, taking into account the emission intensities

58 keV: ε=0.792
203 keV: ε=0.162
375 keV: ε=0.184

Global detection efficiency of ^{127}Xe: ε = 0.967

Calculation of detection efficiency 4

•Uncertainty calculation: composition of uncertainties due to atomic and nuclear data and uncertainties due to Monte Carlo simulation of photons absorption

•Uncertainties on atomic and nuclear data: from « table des radionucléides », LNHB

•Uncertainties on Monte Carlo simulation: conservative value of 10 %

•Low influence of atomic and nuclear data (robust model)

•Small influence of Monte Carlo simulation results

• **Combined relative standard uncertainty of detection efficiency: 0.4 %**

Global uncertainty budget

Component	Relative standard uncertainty %
Counting statistics	0.38
Counters volumes	0.23
Reference volume and STP correction	0.30
Decay corrections	0.05
Discriminator threshold	0.15
Detection efficiency	0.4
Combined standard uncertainty	**0.7**

Primary Standardisation of activity

Isothermal calorimetry

Basic relationship between rate of energy (heat) input , or power *P*, and activity *A*

$$dH/dt = P = A\,\hat{E}$$

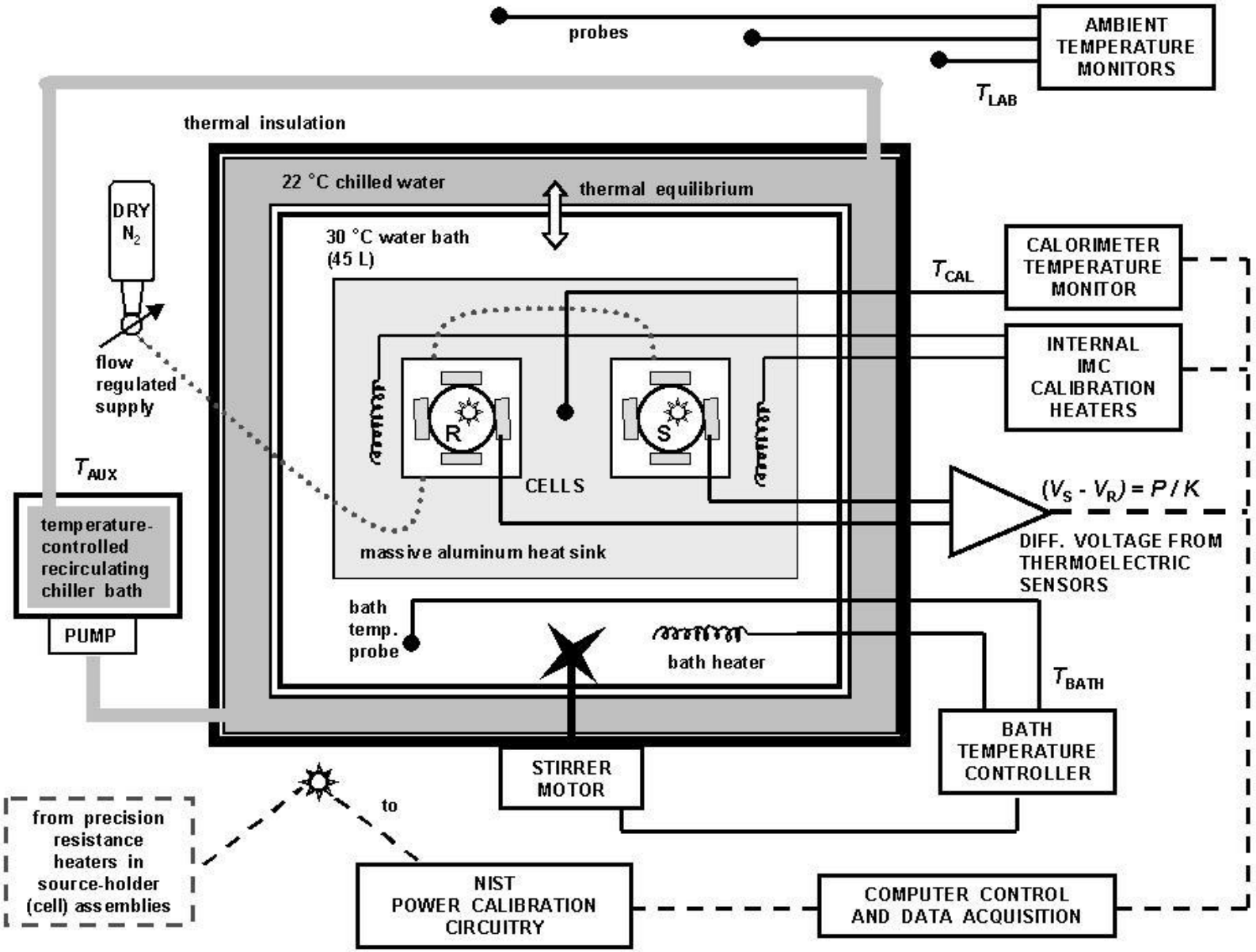
$\hat{E}$ = average energy per decay

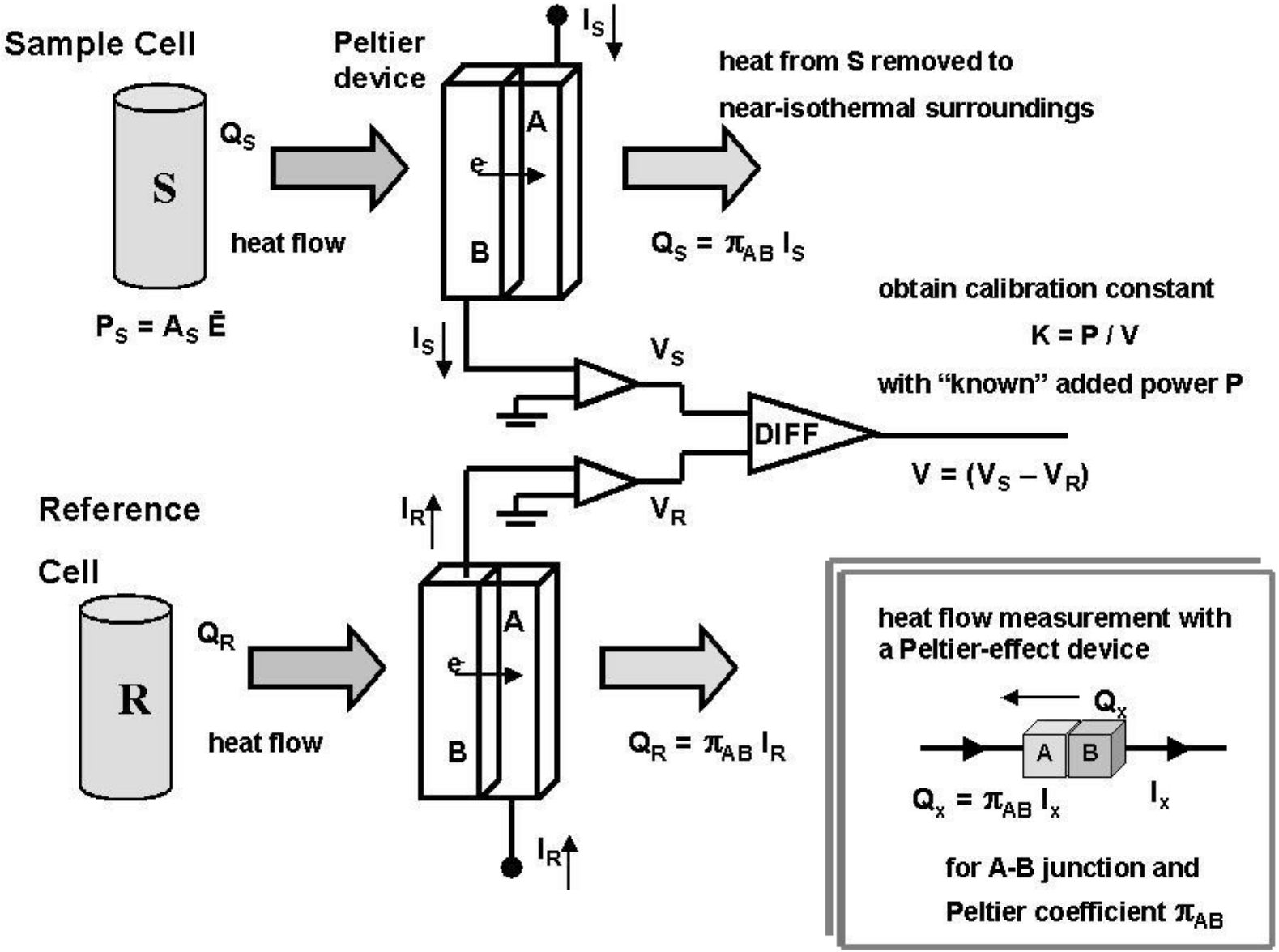
^{3}H / ^{55}Fe	0.9 $\mu W \cdot GBq^{-1}$
^{103}Pd / ^{125}I	9.
^{32}P	111.
^{90}Sr-^{90}Y	181.
^{226}Ra	4338.

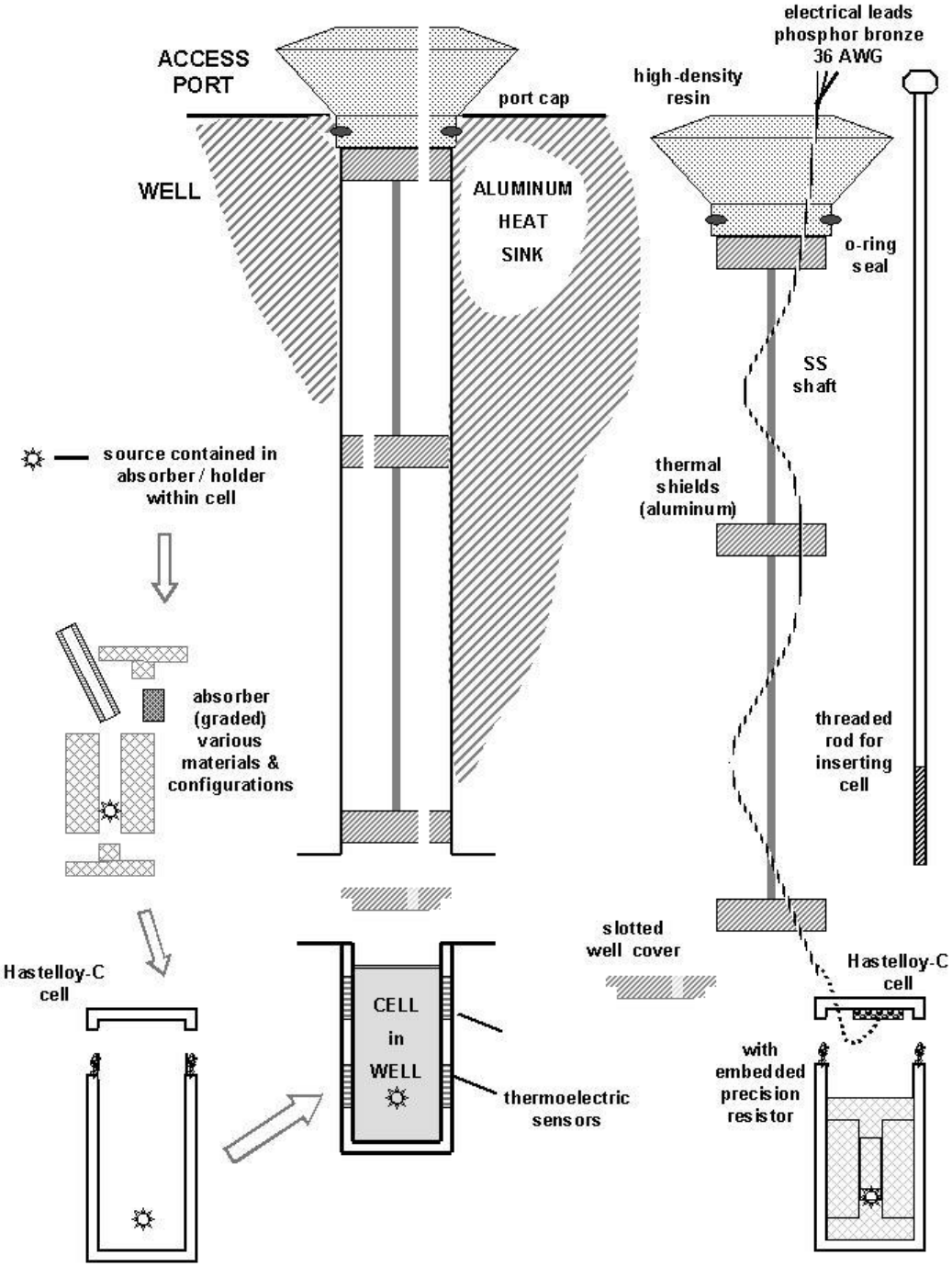
Assumes absorb & measure ALL ionizing radiation (no losses)

And no “heat defect” effects (I.e., no chemistry)

CSC “Isothermal Microcalorimeter (IMC)”

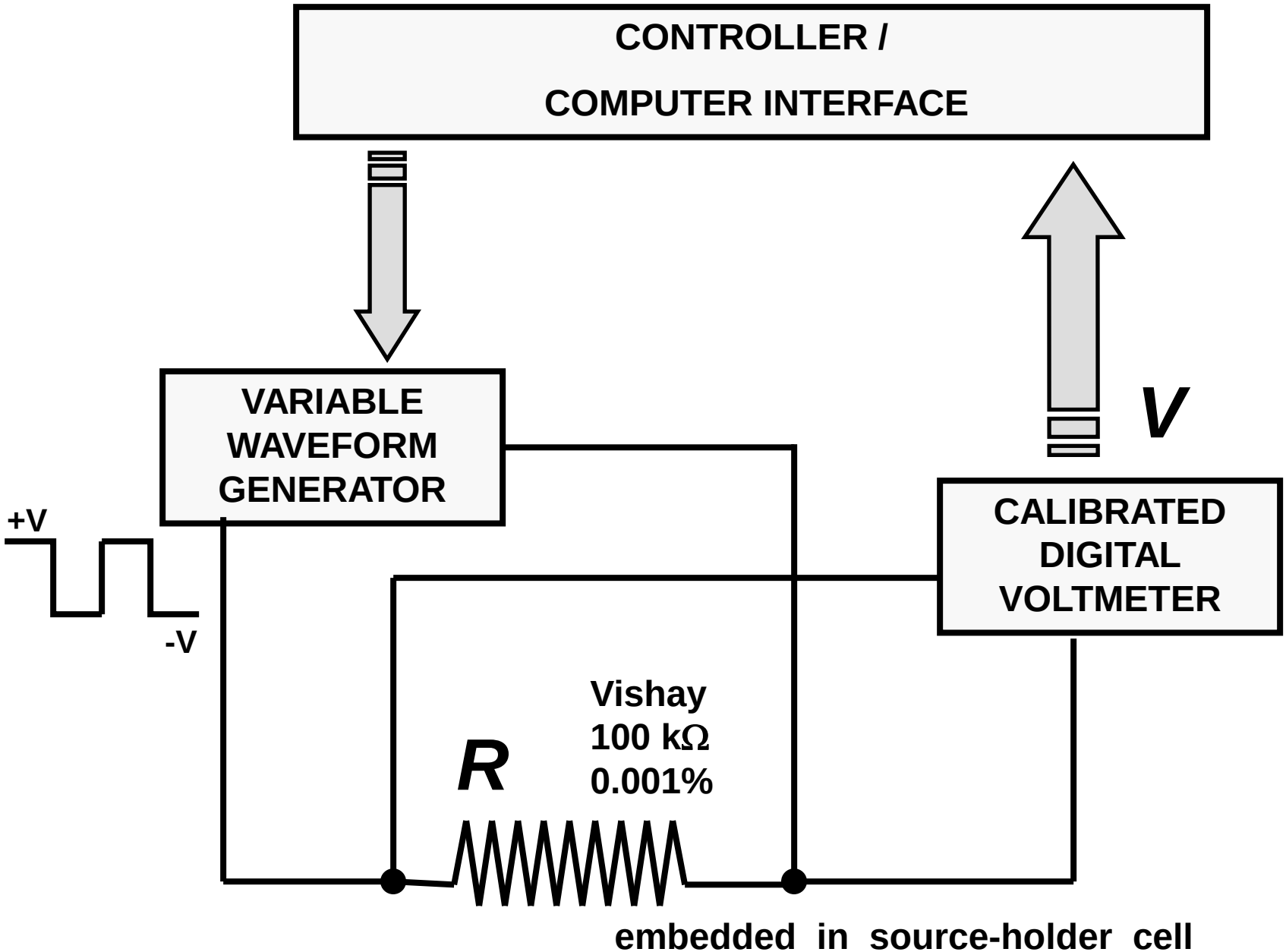

probes
AMBIENT TEMPERATURE MONITORS
T_{LAB}
thermal insulation
22 °C chilled water
thermal equilibrium
30 °C water bath (45 L)
DRY N_2
flow regulated supply
T_{CAL}
CALORIMETER TEMPERATURE MONITOR
INTERNAL IMC CALIBRATION HEATERS
R
S
CELLS
massive aluminum heat sink
T_{AUX}
temperature-controlled recirculating chiller bath
PUMP
$(V_S - V_R) = P / K$
DIFF. VOLTAGE FROM THERMOELECTRIC SENSORS
bath temp. probe
bath heater
T_{BATH}
BATH TEMPERATURE CONTROLLER
STIRRER MOTOR
from precision resistance heaters in source-holder (cell) assemblies
to
NIST POWER CALIBRATION CIRCUITRY
COMPUTER CONTROL AND DATA ACQUISITION

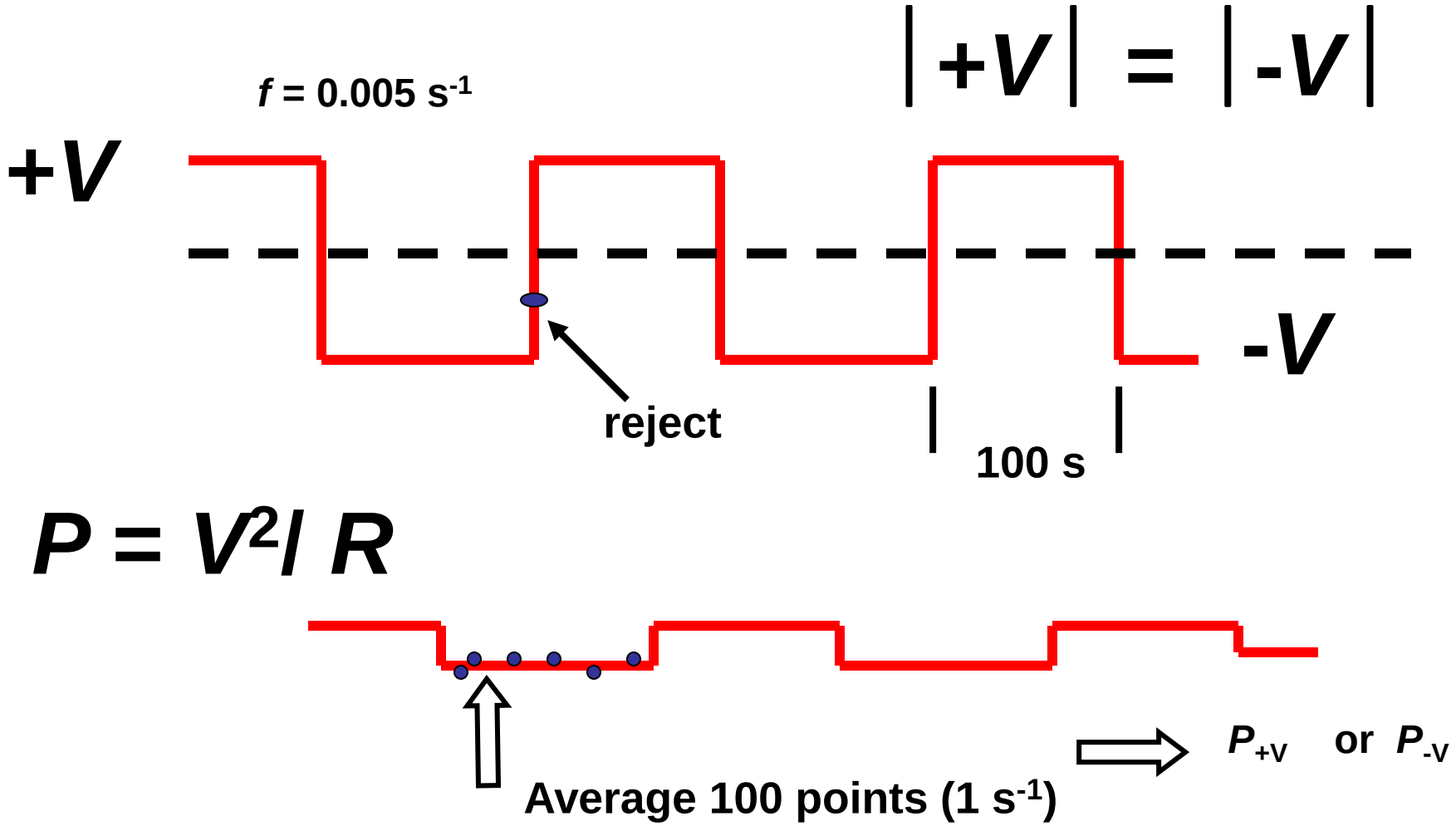

Sample Cell
S
Q_S
heat flow
$P_S = A_S \bar{E}$
Peltier device
I_S
A
e
B
heat from S removed to near-isothermal surroundings
$Q_S = \pi_{AB} I_S$
obtain calibration constant
K = P / V
with "known" added power P
I_S
V_S
DIFF
$V = (V_S - V_R)$
I_R
V_R
Reference Cell
R
Q_R
heat flow
A
e
B
$Q_R = \pi_{AB} I_R$
I_R
heat flow measurement with a Peltier-effect device
Q_x
A
B
$Q_x = \pi_{AB} I_x$
I_x
for A-B junction and Peltier coefficient π_{AB}


ACCESS PORT
port cap
WELL
ALUMINUM HEAT SINK
source contained in absorber / holder within cell
absorber (graded) various materials & configurations
Hastelloy-C cell
CELL in WELL
thermoelectric sensors
electrical leads phosphor bronze 36 AWG
high-density resin
o-ring seal
SS shaft
thermal shields (aluminum)
threaded rod for inserting cell
slotted well cover
Hastelloy-C cell
with embedded precision resistor

port assemblies -- source (absorbers) holders & cells

$P = V^2/R$


CONTROLLER /
COMPUTER INTERFACE
VARIABLE
WAVEFORM
GENERATOR
V
CALIBRATED
DIGITAL
VOLTMETER
+V
-V
R
Vishay
100 kΩ
0.001%
embedded in source-holder cell



$P = V^2 / R$

mean (P) = 1/2 mean (P_{+V}) + 1/2 mean (P_{-V})

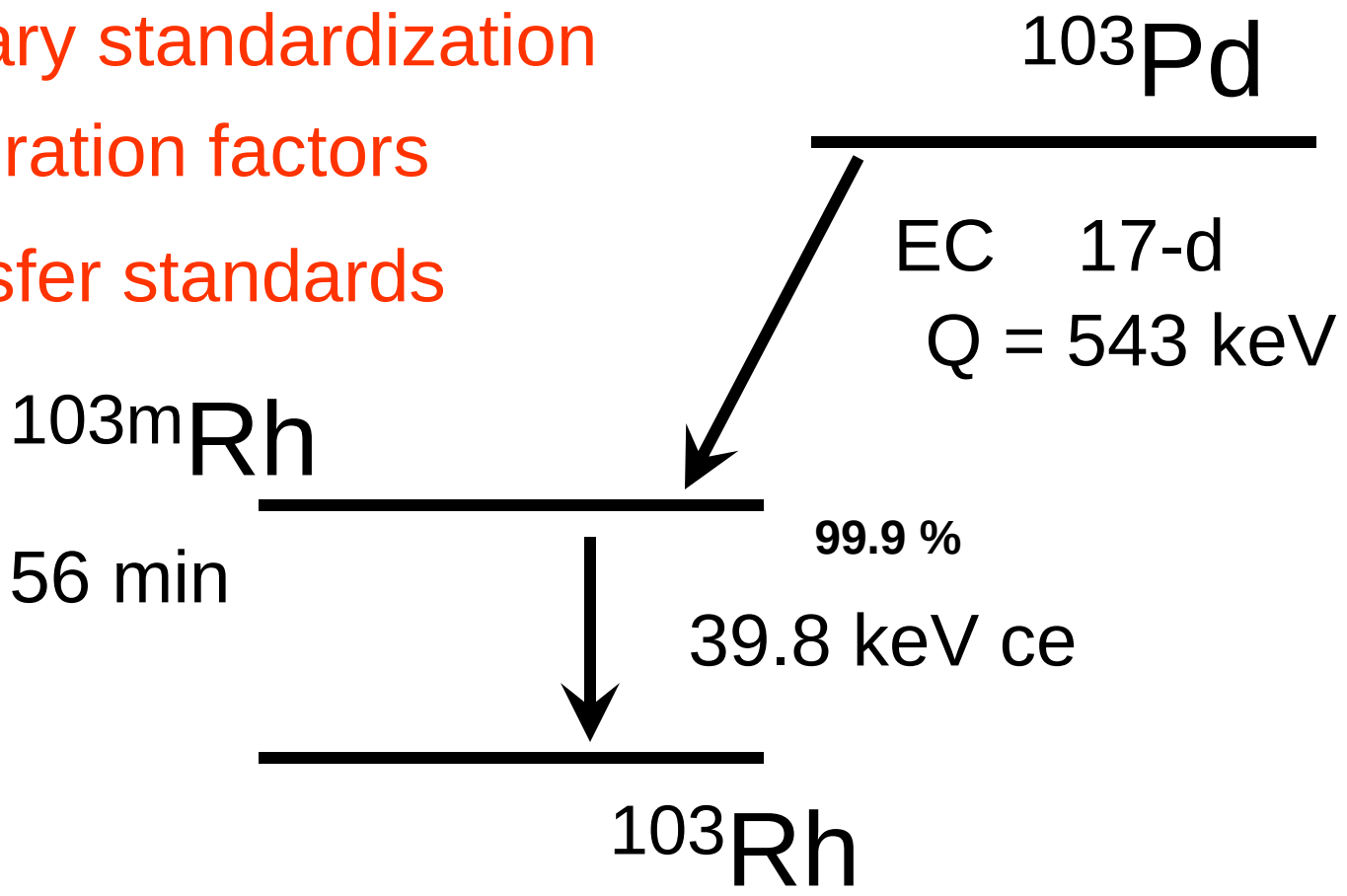
var (P) = 1/2 var (P_{+V}) + 1/2 var (P_{-V}) + covar (P_{+V}, P_{-V})

+ autocorr(P_{+V}) + autocorr(P_{-V})

need primary standardization

+ calibration factors

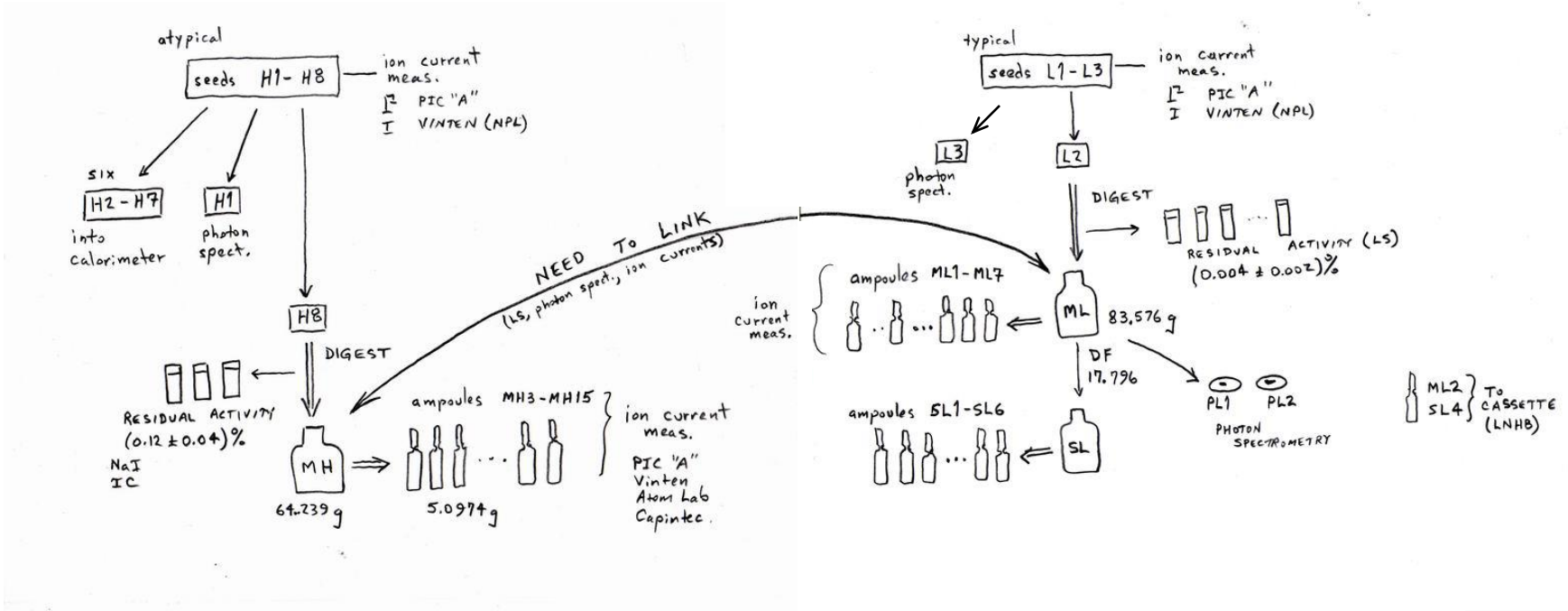
+ transfer standards

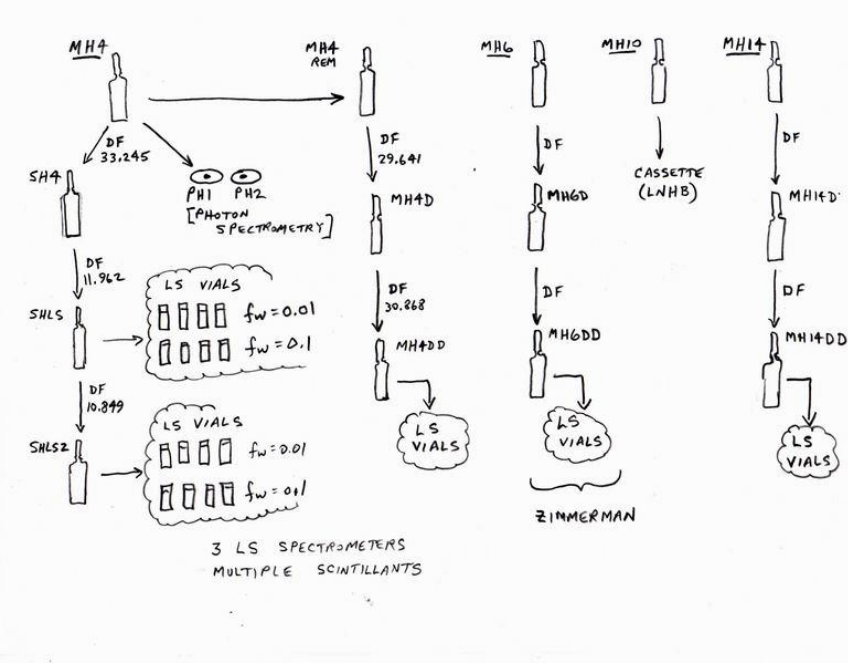


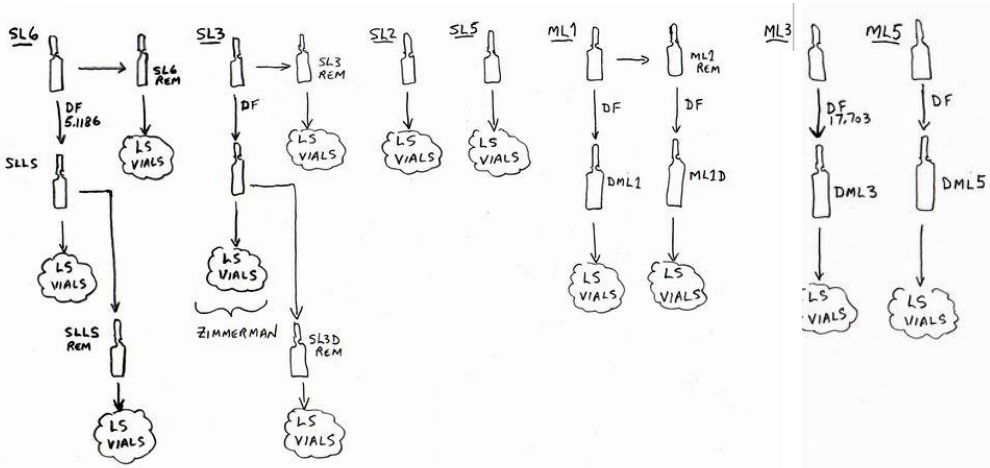
Brachytherapy source

used to treat prostate cancer

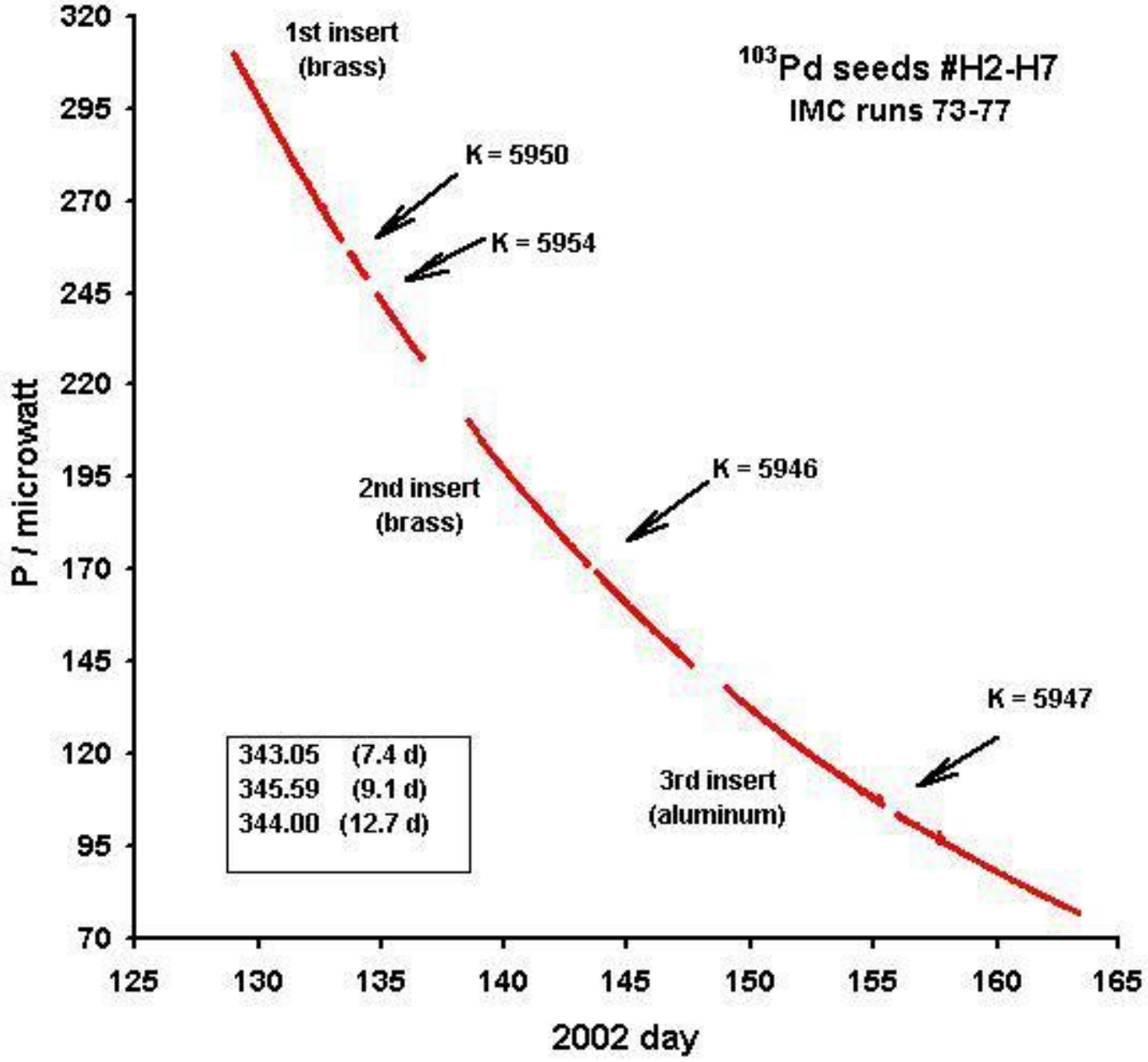
candidate for intravascular use





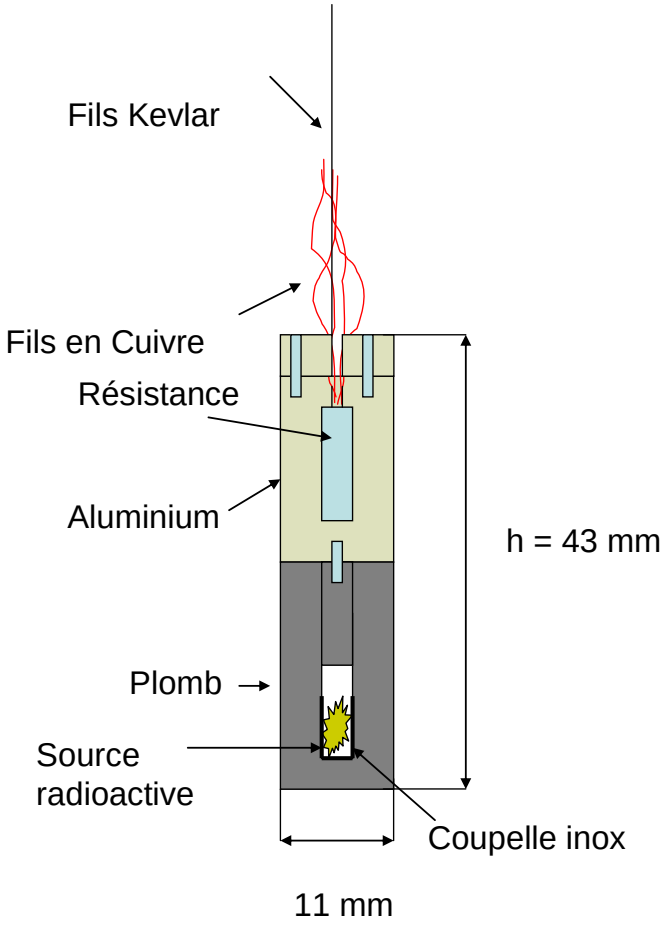


^{103}Pd standardization scheme


320
295
270
245
220
195
170
145
120
95
70
P / microwatt
125
130
135
140
145
150
155
160
165
2002 day
1st insert
(brass)
^{103}Pd seeds #H2-H7
IMC runs 73-77
K = 5950
K = 5954
2nd insert
(brass)
K = 5946
K = 5947
3rd insert
(aluminum)
343.05 (7.4 d)
345.59 (9.1 d)
344.00 (12.7 d)

Calorimeter at LNHB

Source holder



Example : standardization of a ^{241}Am source

$$ {}^{A}_{Z}X_{N} \rightarrow {}^{A-4}_{Z-2}Y_{N-2} + {}^{4}_{2}He_{2} $$

$$ Q_{\alpha} = E_{\alpha i} + E_{i} + E_{ri} $$

- $E_{\alpha i}$: energy of the α particle
- E_i: excited level energy
- E_{ri}: recoil energy

100% alpha decay towards ^{237}Np: Q_{α}= (5637,81± 0,12)keV

γ et X-ray emission

Emission of 131 γ photons with energies between 26 keV and 1 MeV) but main emission at **59,54 keV** (emission intensity 36%)

Secondary X-rays emission between 11,9 keV and 118,4 keV

Monte Carlo simulation of photon interactions with the absorber cell (PENELOPE)

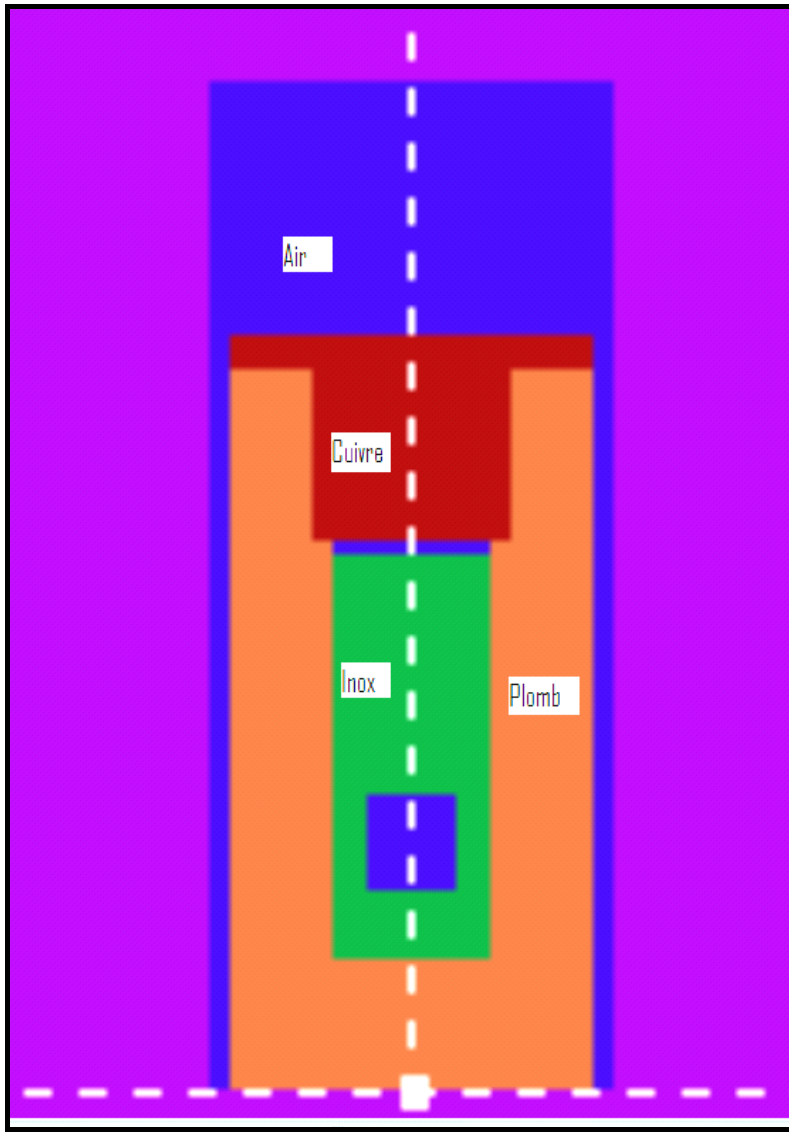
Determination of the absorbed power

Results for A = 10 MBq

Power released: $P_D = (9{,}0205 \pm 0{,}0002)\ \mu W$

Relative absorption $P_{abs} = 99{,}9999350\ \%$

Geometry of the absorber cell



Activity of the ^{241}Am source

Calorimetry $P = (11{,}633 \pm 0{,}041)\ \mu W$

$$A = \frac{P}{E_D} \quad \text{and} \quad E_D = 5637{,}81\ keV$$

$$A = (12{,}896 \pm 0{,}045)\ MBq$$

Comparative measurement: LSC after quantitative dissolution of the source

$$A_{source} = (12{,}8884 \pm 0{,}0045)\ MBq$$

Relative difference: 0,06 %

Comments about calorimetry

Calorimetry is SLOW

needs long time to thermally stabilize

typically need multiple determinations

different / absorbers / Monte Carlo calc. verifications

Accuracy is in range of $\pm$ 1 or 2 percent

Largely due to baseline instabilities and

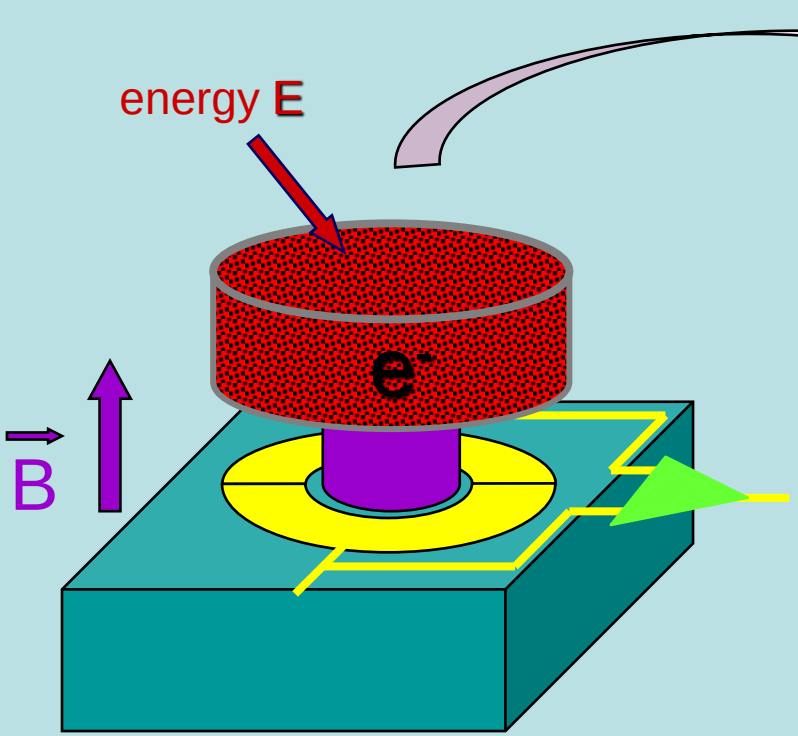
uncertainties in establishing baselines to get ΔP

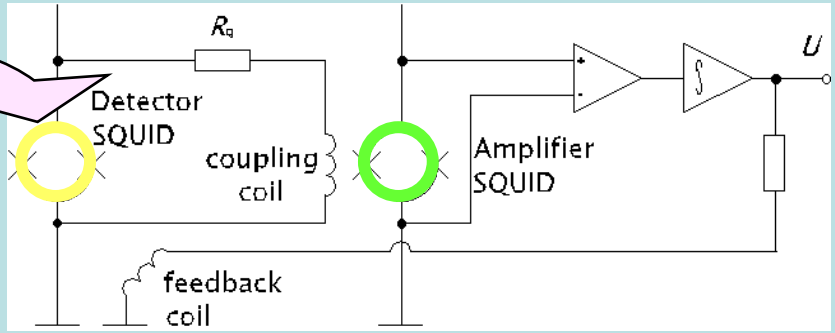
Power may be measured very accurately

But still need average energy per decay to get Activity

Low-temperature calorimeters (bolometers)

Magnetic metallic calorimeters (MMC)





metallic Au absorber

temperature rise

$$\Delta T = E / C$$

magnetic thermometer (Au:Er) in B - field

variation of magnetic moment

1st SQUID : sensor read-out (variation of flux)

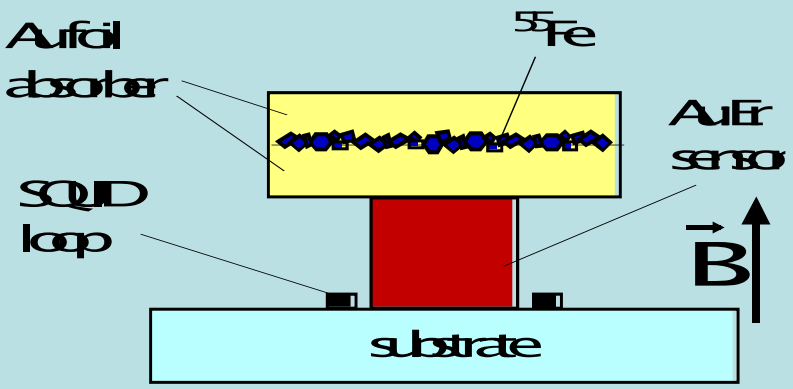
2nd SQUID : signal amplification ΔV

Why very low temperatures (< 100 mK) ?
Heat capacity C = g T and thermodynamic fluctuations

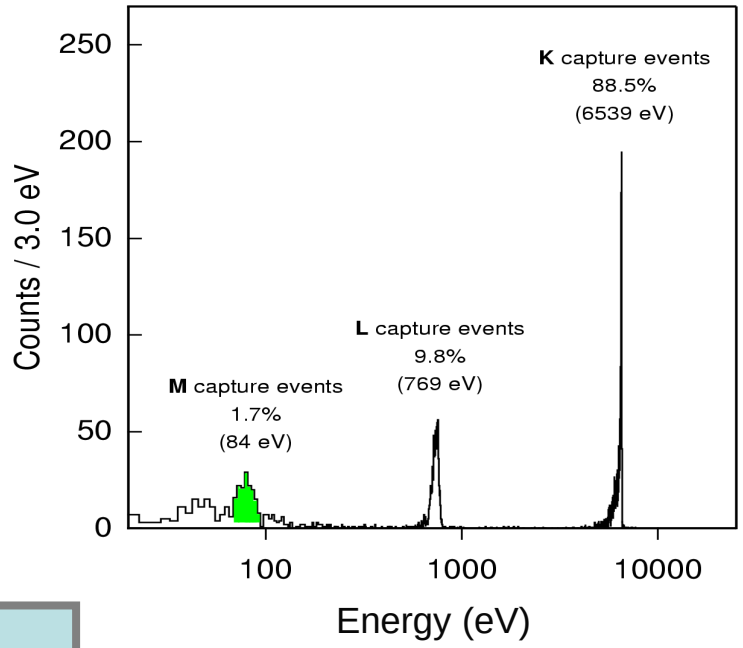
What do we measure? Incident energy converted into detectable energy

Detector	Physics of the detectors	minimum energy for a carrier of information
Scintillators	Visible light photons	**0.1 to 1 keV**
Proportional counters	Ionisation	**10 to 30 eV**
Semi-conductors	Electron-hole pairs	**3 - 4 eV**
At very low temperature (<100 mK)		
Superconducting Tunnel Junctions	Breaking of Cooper pairs → **quasiparticles**	10^{-3} eV
Dielectric calorimeters	**Thermal phonons production**	10^{-5} to 10^{-4} eV
Metallic magnetic calorimeters	**Thermal excitation of conduction electrons**	$< 10^{-5}$ eV

Absolute activity measurement with MMC



Total absorption spectrum of an enclosed ^{55}Fe source



- Efficient thermalization of electron energy :
  - ✓ K, L and M captures perfectly separated
- Good energy resolution
- Energy detection threshold : < 100 eV
- Detection efficiency for photons of 100 eV - 6,5 keV : 99 %

Primary Standardisation of activity

Neutron emission rate

Neutron sources

Measurand:

Emission rate under 4π sr (unit : s^{-1})

Neutron sources:

- (α,n) : AmBe, PuBe, RaBe…
- spontaneous fission: ^{252}Cf, ^{242}Cm…
- mixtes : $^{244}CmBe$…
- (γ,n) : PuBe…

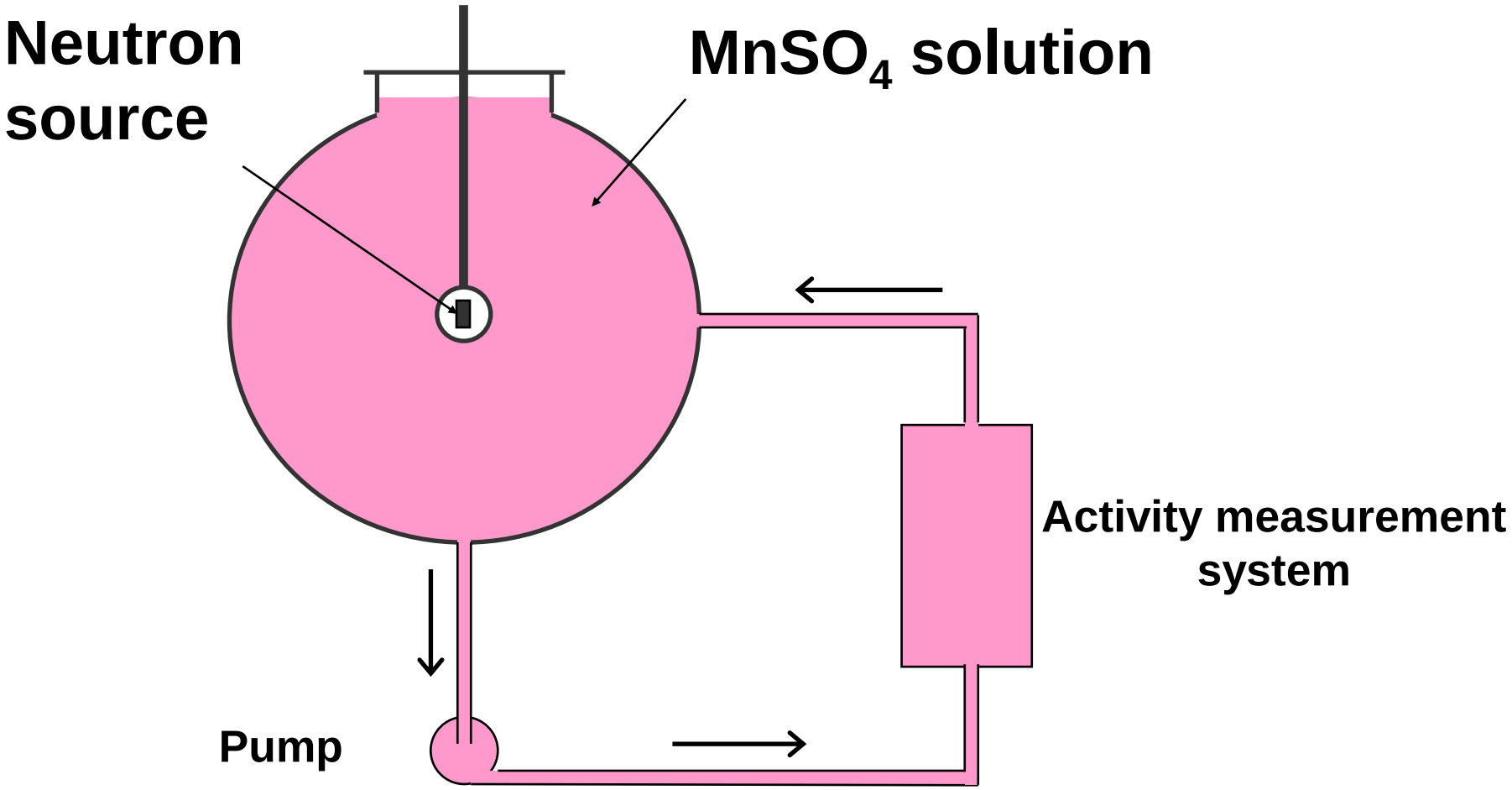
Range:

- from 10^5 to 10^9 s^{-1}

Target uncertainty:

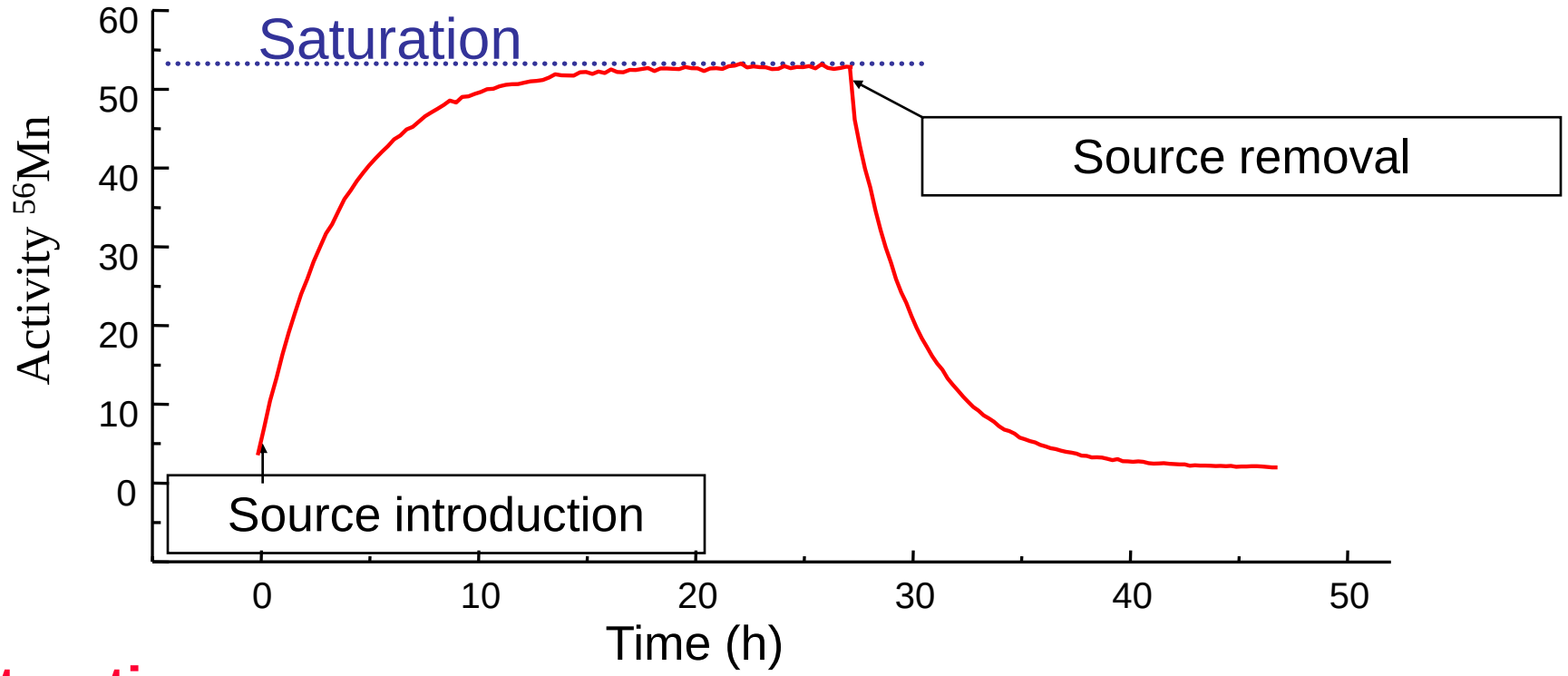
- about 1 %

Measurement principle



Activation kinetics

$$^{55}Mn + n_0 \rightarrow {}^{56}Mn + \gamma$$



Saturation :

^{56}Mn production by neutron capture = ^{56}Mn radioactive decay → ^{56}Mn constant activity

Neutron emission rate

Φ : number of neutrons emitted by time unit under 4π sr

$$\Phi = \frac{A_{sat}}{R}$$

A_{sat} : mesured

R : calculated

Φ : emission rate under 4π sr, s^{-1}

A_{sat} : ^{56}Mn activity in the bath at saturation, *Bq*

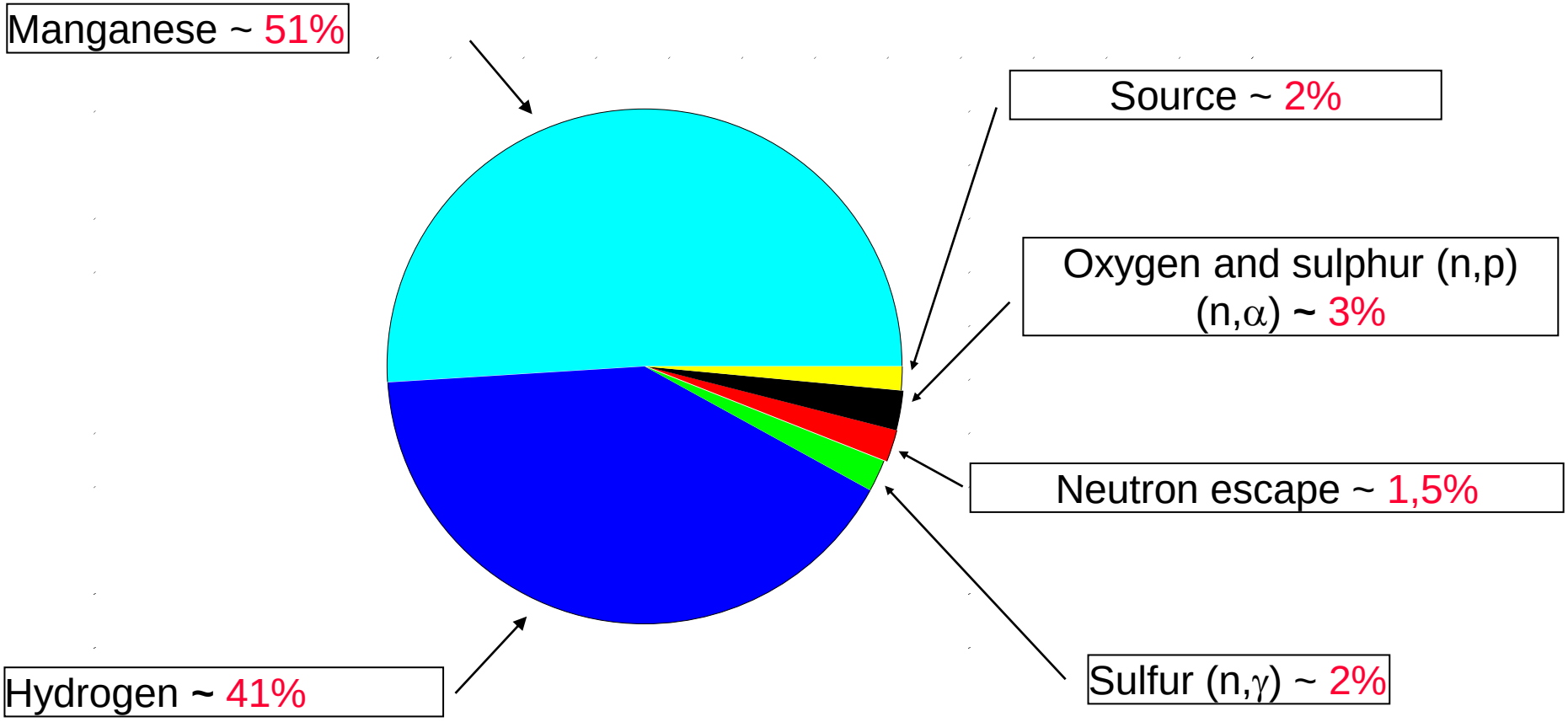
R : efficiency of the bath (probability of creation of one ^{56}Mn atom per neutron)

Relative uncertainty

$$\frac{u_\Phi}{\Phi} = \sqrt{\frac{u_{A_{eq}}^2}{A_{eq}^2} + \frac{u_R^2}{R^2}}$$

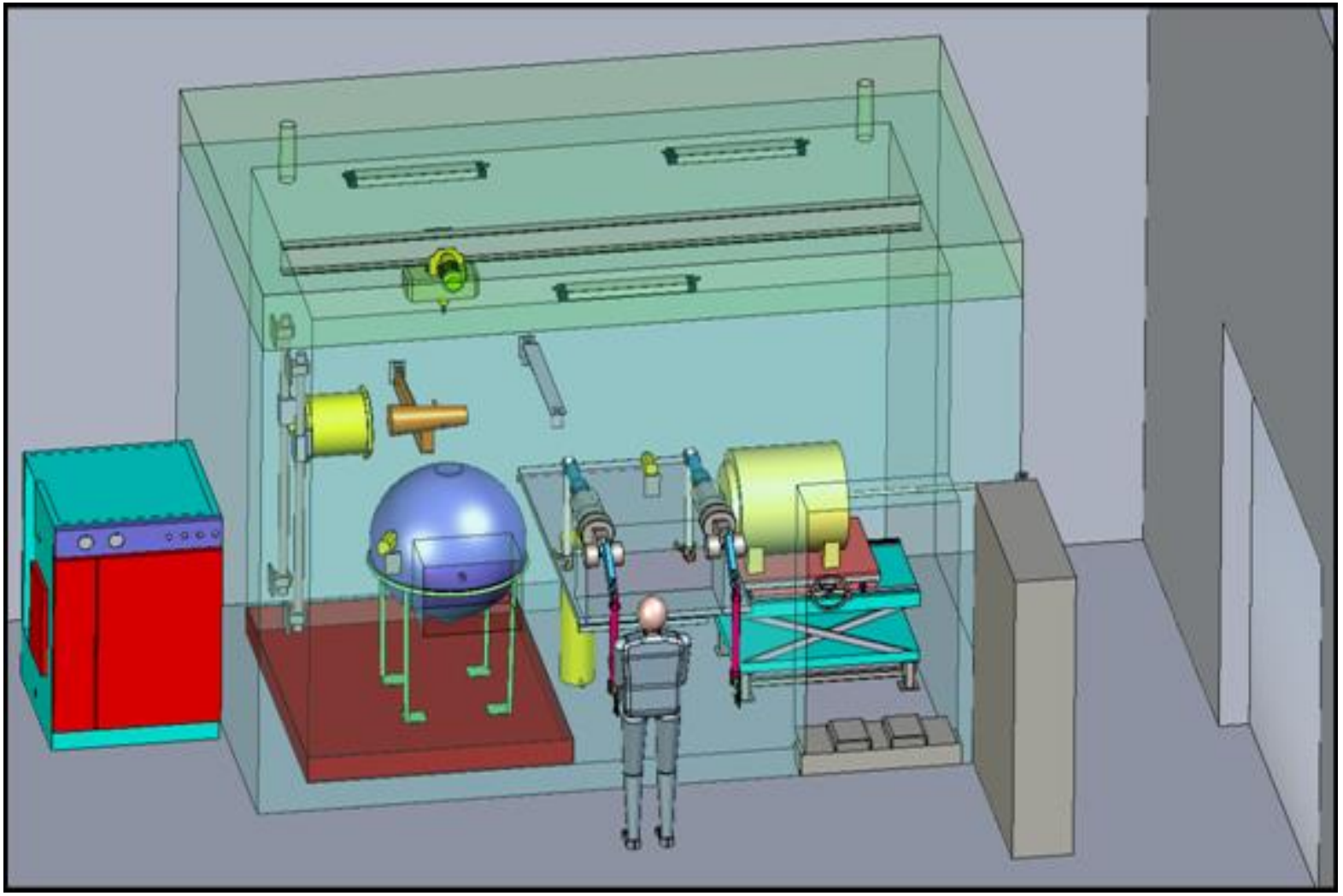
Neutron interaction with the bath

Example X3 AmBe source



Interaction probabilities

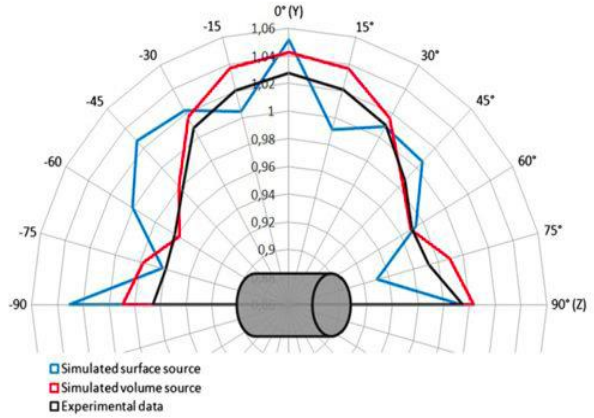
Facility at LNHB



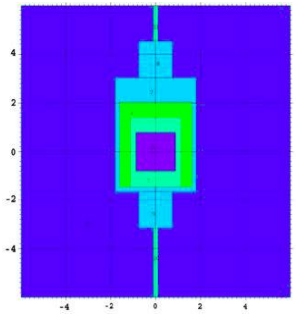
Inside view

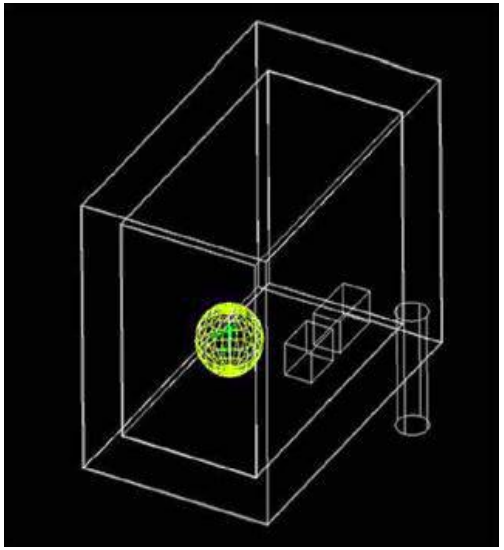
Monte Carlo model of the bath

• Source model:
volume source (composition of point sources with emission spectrum from ISO 8529-1)



• detailled model of the source with structures





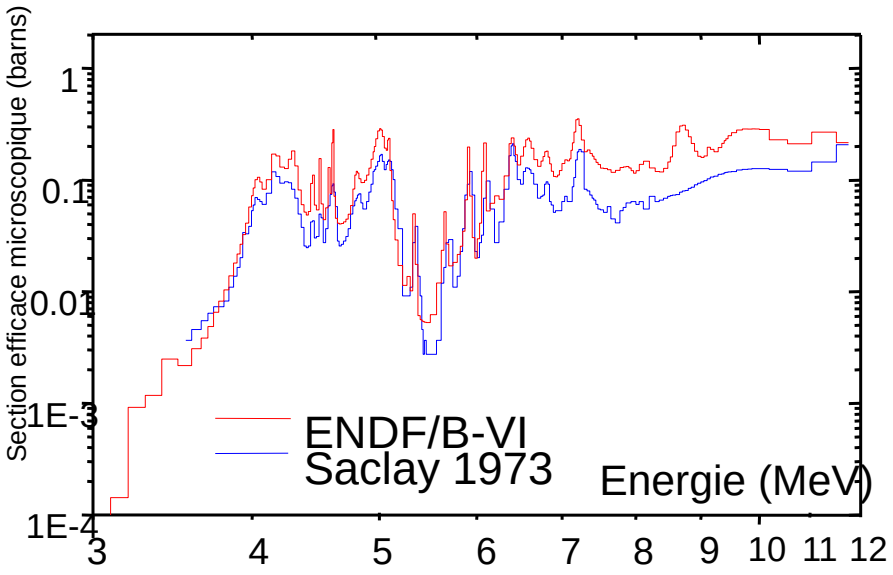
Monte Carlo calculation

- Choice of cross-section libraries

Selected library: ENDF/B VII-0

<u>Major problem on oxygen cross-sections (disparities between libraries)</u>

0,5 % difference on the calculated bath efficiency between two cross-sections libraries



Comparison between Monte Carlo codes

For the same input data (geometry, cross sections, source) results of MCNPX, FLUKA et GEANT4

Code	Mn	H
MCNPX	42,72 (1)	51,00 (1)
GEANT4	45,25 (2)	48,66 (2)
FLUKA	43,47 (7)	51,92 (7)

Probability of interaction with the atoms of the bath
(uncertainties are the Monte Carlo fluctuations)

Known issues

1. Oxygen cross-sections

2. Bias between MCNPX and FLUKA (up to 1,8 % difference on the bath efficiency)

MCNPX is the standard code for laboratories using the manganese bath method

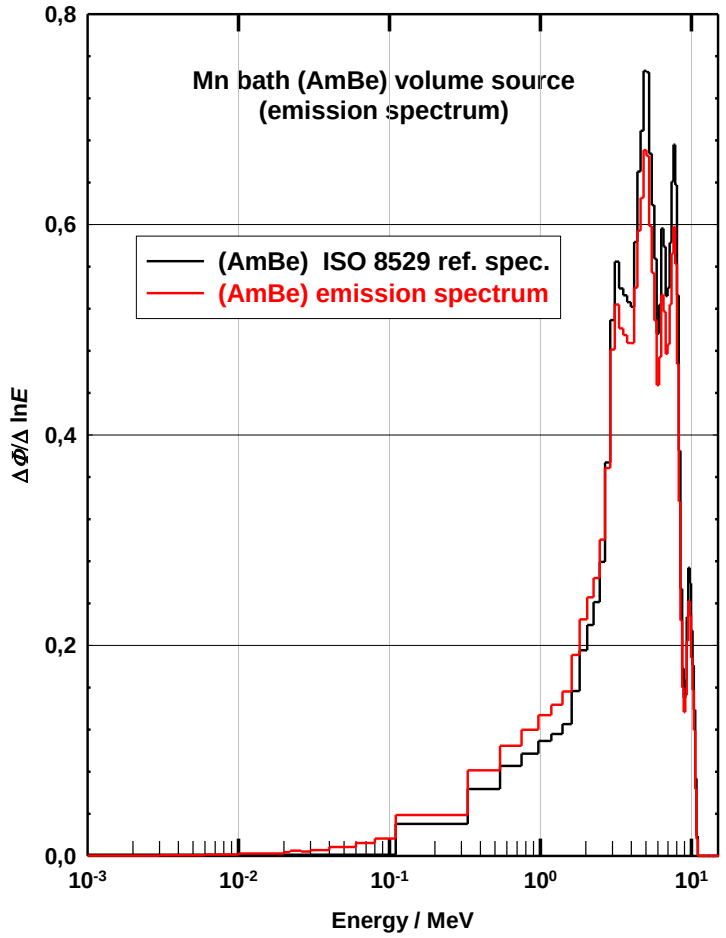
Thus, if durng an international comparison all participants use MCNPX and the same cross-section library (e.g. CCRI(III) K9 AmBe1 comparison)… a good consensus can be reached!

But an experimental validation is necessary. This validation is extremely complicated.

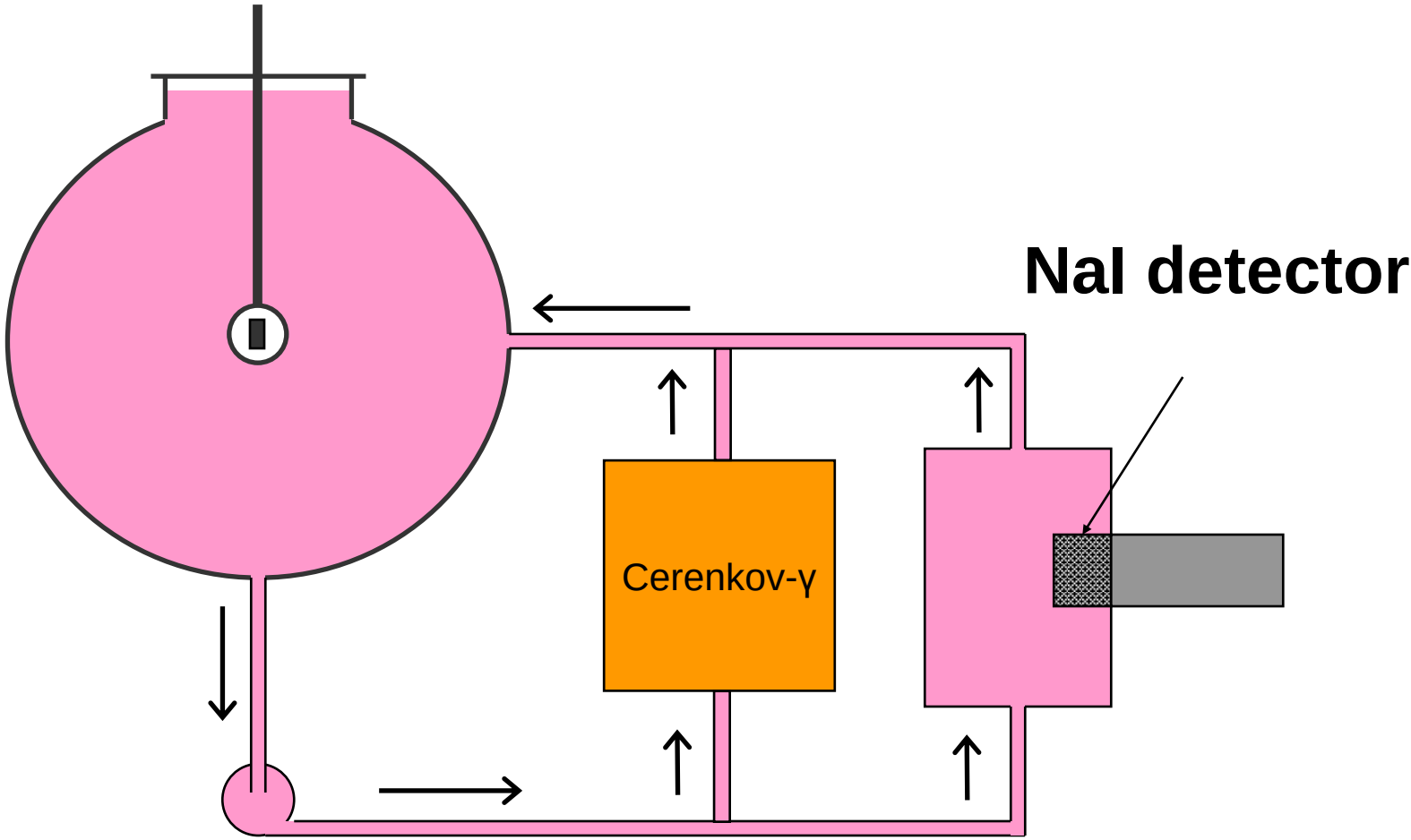
Validation difficulties

Neutron measurement is difficult

- non-directly ionizing radiation
- scattering
- Very large energy range (from a few meV to several tenths of MeV)
- Measurement instruments are generally calibrated with standards… calibrated with the manganese bath method!

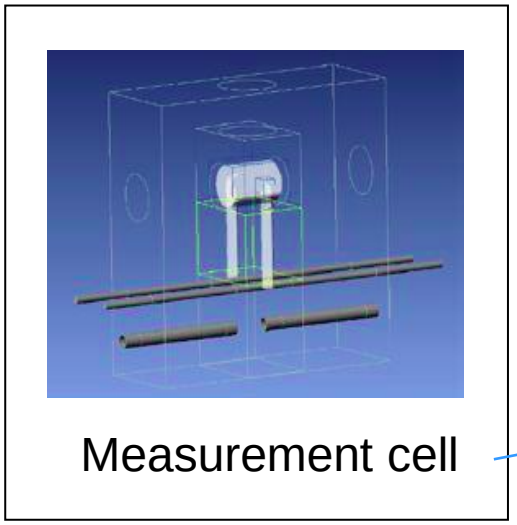


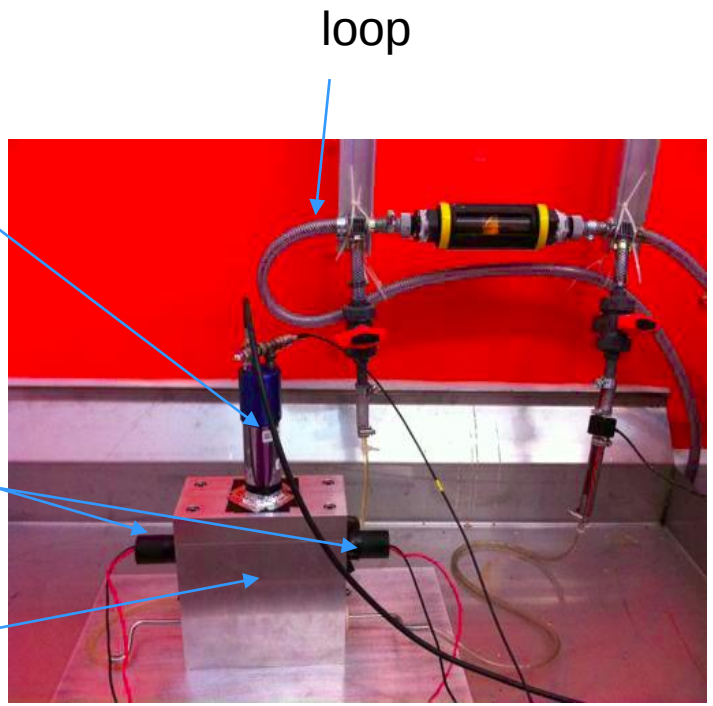
Recent developments: online measurement of the ^{56}Mn activity by Cerenkov-γ coincidence



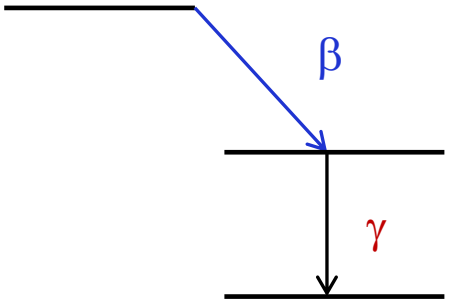
Online ^{56}Mn activity measurement

Advantage: avoid the calibration of the NaI detector



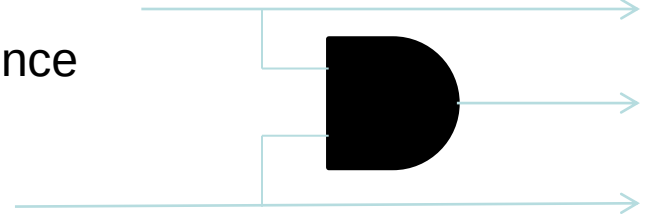


^{56}Mn coincidence counting



Cerenkov light detected by two PMTs in coincidence

γ detected by NaI(Tl) detector



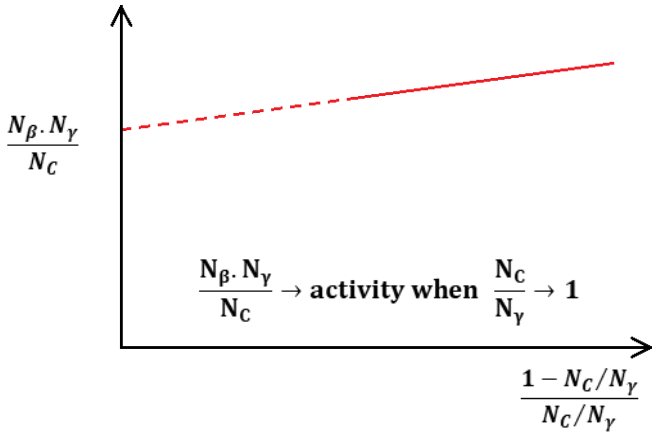
With some β–γ cross-talk

$$N_\beta = A\left[\varepsilon_\beta + (1-\varepsilon_\beta)\varepsilon_{\beta\gamma}\right]$$

$$N_\gamma = A\varepsilon_\gamma$$

$$N_c = A\left[\varepsilon_\beta \varepsilon_\gamma\right]$$

$$and \quad N_\beta = A\left[1-\left(1-\frac{N_C}{N_\gamma}\right)\varepsilon_{\beta\gamma}\right] = A.F\left(1-\frac{N_C}{N_\gamma}\right)$$



Conclusion on primary measurement methods

- Necessity of various instrumentation and measurement methods due to the various properties of radioactivity
- Precise primary activity measurement is always a difficult task
- Final relative standard uncertainty can be lower than 1 %, depending on the method
- The more precise methods are based on coincidence counting or defined solid angle counting