The Abdus Salam International Centre for Theoretical Physics

United Nations Educational, Scientific and Cultural Organization

IAEA International Atomic Energy Agency

**1833-48**

# Workshop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

*5 - 16 November 2007*

## Radioactivity measurements and uncertainty evaluation.

Guy RATEL

*Bureau International des Poids et Mesures BIPM*
*Pavilion de Breteuil*
*F-92312 Cedex Sevres*
*FRANCE*

Bureau International des Poids et Mesures


Workshop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

ICTP – Trieste

(5 – 16 November 2007)

# Radioactivity measurements and uncertainty evaluation

**by Guy Ratel**

# Presentation outline

1. Concept of uncertainty
2. Some useful definitions:

   mean, expectation value, variance, median

3. Introduction of three important distributions

   a) rectangular distribution

   b) Poisson distribution

   c) Gaussian distribution

4. Introduction of the Gaussian uncertainty propagation law
5. Application of the preceding concepts to radioactivity measurements
6. Treatment of correlations

## 1. Concept of uncertainty

In metrology the main concern is to measure the value of a given quantity with some appropriate tools.

For instance if someone wants to measure the mass of an amount of substance he will use a balance. The reading of the balance gives a representation of the actual value of the mass in reference to a particular unit, for instance it can be grams (g).

**The value obtained for the mass depends on several factors – among others:**

- **the reproducibility of the method of measurement;**
- **the quality of the standards used to calibrate the balance;**
- **the values and the stability of the physical conditions under which the measurements are carried out, such as pressure, temperature and hygrometry.**

**All these parameters have an influence on the final result of the measurement so that the exact value of the quantity cannot be predicted. Only a most probable value can be attributed to the measurand.**

At the same time as measurements are carried out an estimation of the limits of the domain in which the actual value of the measurand lies should be undertaken.

The differences between the most probable value and each of the two limits (above and below this value) are called uncertainties. The two uncertainties do not have to be equal but in this presentation only symmetrical uncertainties will be considered.

To summarize, all measurement results should always be stated with an uncertainty indicated below:

$$m = 100 \text{ g}; \; u = 0.01 \text{ g}.$$

## 2. Some useful definitions

Often several measurements of the quantity under study are carried out at the same time giving different results. These can be obtained using the same method (replication) or different techniques which have proven to be suitable for the purpose. (Some examples will be given later).

In this case one may wish to characterize the population defined by the entire set of the individual results with some appropriate single figures.

This is usually achieved by means of particular statistical tools, the most frequently used are the arithmetic mean, the median, the weighted mean and the weighted median. Only the first two will be presented in the following.

# Radioactivity measurements and uncertainty evaluation

**a) Arithmetic mean value (also called the expectation value)**

**Let designate one particular result set of *n* values with the variable {$A_i$} (*A* standing for activity), with *i* = 1, .. , *n*.**

**The arithmetic mean is defined as** $\bar{A} = \frac{1}{n}\sum_{i=1}^{n} A_i = E(A).$

**Because** $\bar{A}$ **represents a physical quantity it should be given with an uncertainty.**

**The first idea is to take the uncertainty of one measurement as equal to its deviation from the mean value and, as a consequence, the uncertainty of the population as the mean of all individual deviations:**

$d_i = (A_i - \bar{A})$ **and** $\bar{d} = \frac{1}{n}\sum_{i=1}^{n} d_i.$

# Radioactivity measurements and uncertainty evaluation

The latter expression is by definition equal to 0 and it is not very useful.

To solve the problem an expression equal to $\frac{1}{n}\sum_{i=1}^{n}\left|A_i - \bar{A}\right|$

could have been adopted but this is not mathematically simple.

The next possible choice is to consider the square of the deviation instead of the single deviation to define the uncertainty. The empirical sample variance is then taken as

$$s^2 = E\left(A - \bar{A}\right)^2 = \frac{1}{n-1}\sum_{i=1}^{n}\left(A_i - \bar{A}\right)^2, \; n \neq 1.$$

It follows that the standard deviation *s* is given by the root of the variance.

$$s = \sqrt{\frac{1}{n-1}\sum_{i=1}^{n}\left(A_i - \bar{A}\right)^2}, \; n \neq 1.$$

As expected $s$ has the same dimension as $A_i$.

## a) Median

The median of a sample of ordered values $\{A_i\}$, with $i = 1, .. , n$ and $A_i > A_{i+1}$ is the value for which one half of the data is above, one half below. This implies that the value of the median will change depending on whether the number of data is odd or even.

If $n = 2k + 1$ the median is simply the central value of the ordered data

$$\tilde{A} = A_k.$$

If $n = 2k$ it is usual to define the median as the arithmetic mean of the two central values $A_k$ and $A_{k+1}$

$$\tilde{A} = \frac{1}{2}\left(A_k + A_{k+1}\right).$$

## Uncertainty of the median

The uncertainty of the median is given by

$$u_{\tilde{A}} = C \bullet \mathrm{MAD},$$

with $C = 1.858,$

and where MAD stands for median of the absolute deviations and is defined as

$$\mathrm{MAD} = \mathrm{med}\left\{\left|A_i - \tilde{A}\right|\right\}.$$

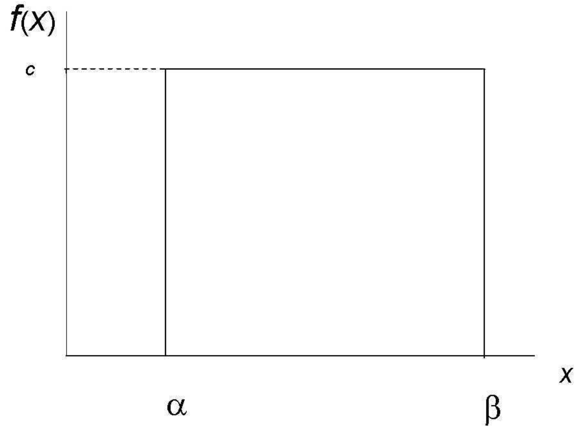
## 3. Three important distributions for radioactivity measurements

### a) Rectangular distribution

$$f(x) = \frac{1}{(\beta - \alpha)} \quad \text{for } \alpha < x < \beta,$$

$$f(x) = 0 \quad \text{for } x < \alpha \text{ or } x > \beta.$$

$$\text{with} \quad c = \frac{1}{(\beta - \alpha)}.$$




**Expectation value :** $E(x) = m = \int_{\alpha}^{\beta} \frac{x}{\beta - \alpha} dx = \frac{\alpha + \beta}{2}.$

**The origin of the x-axis can be chosen at any other convenient place: the distribution can be made symmetrical with regard to the y-axis. It follows that median and mean or expectation value have the same expression.**

**Variance :**

$$\mu_2 = \sigma^2 = E\left[\left(x-m\right)^2\right] = \frac{1}{\left(\beta-\alpha\right)}\int_{\alpha}^{\beta}\left(x-\frac{\alpha+\beta}{2}\right)^2 dx$$

$$= \frac{1}{\left(\beta-\alpha\right)}\int_{\frac{\alpha-\beta}{2}}^{\frac{\beta-\alpha}{2}} y^2 dy = \frac{1}{\left(\beta-\alpha\right)}\frac{2}{3}\left(\frac{\beta-\alpha}{2}\right)^3 = \frac{\left(\beta-\alpha\right)^2}{12}.$$

**Standard deviation :**

$$\sigma = \sqrt{\frac{\left(\beta-\alpha\right)^2}{12}} = \frac{\left(\beta-\alpha\right)}{2\sqrt{3}}.$$

**This distribution applies also to the case of discrete random variables.**

## b) Poisson distribution

**A discrete random variable x, which takes only non negative integer values 0, 1, 2 …, *m*, follows a Poisson distribution when the probability that *x* takes the value *m is***

$$P_m = \frac{\lambda^m}{m!} e^{-\lambda},$$ **where $\lambda$ is a parameter characteristic of the distribution.**

**Normalization :** $$\sum_{m=0}^{\infty} P_m = \sum_{m=0}^{\infty} \frac{\lambda^m}{m!} e^{-\lambda} = e^{-\lambda} \sum_{m=0}^{\infty} \frac{\lambda^m}{m!} = 1.$$

**Expectation value :** $$E(x) = \sum_{m=0}^{\infty} m P_m = \sum_{m=1}^{\infty} m P_m = e^{-\lambda} \sum_{m=1}^{\infty} m \frac{\lambda^m}{m!} = \lambda e^{-\lambda} \sum_{m=1}^{\infty} \frac{\lambda^{m-1}}{(m-1)!} = \lambda e^{-\lambda} \sum_{k=0}^{\infty} \frac{\lambda^k}{k!} = \lambda.$$

**The parameter $\lambda$ is therefore the mean value of the random variable *x*.**

# Radioactivity measurements and uncertainty evaluation

**Variance :**

$$E\left[(x-\lambda)^2\right] = E\left(x^2\right) - \lambda^2,$$

$$\begin{aligned} E\left(x^2\right) &= \sum_{m=0}^{\infty} m^2 P_m = e^{-\lambda}\sum_{m=0}^{\infty} m^2 \frac{\lambda^m}{m!} = \lambda e^{-\lambda}\sum_{m=1}^{\infty} m \frac{\lambda^{m-1}}{(m-1)!} \\ &= \lambda e^{-\lambda}\left[\sum_{m=1}^{\infty}(m-1)\frac{\lambda^{m-1}}{(m-1)!} + \sum_{m=1}^{\infty}\frac{\lambda^{m-1}}{(m-1)!}\right] \\ &= \lambda e^{-\lambda}\left[\lambda\sum_{m=2}^{\infty}\frac{\lambda^{m-2}}{(m-2)!} + \sum_{m=1}^{\infty}\frac{\lambda^{m-1}}{(m-1)!}\right] \\ &= \lambda e^{-\lambda}\left(\lambda e^{\lambda} + e^{\lambda}\right) = \lambda^2 + \lambda. \end{aligned}$$

**For a Poisson distribution the variance is equal to the expectation value.**

**Standard deviation :** $\sigma = \sqrt{E\left[(x-\lambda)^2\right]} = \sqrt{\lambda}.$

**A particularly well-known example for a Poisson distribution is the decay of a radioactive substance; the uncertainty of the measurement of the number of disintegrations per time is simply given by the square root of the count rate.**

## c) Gaussian or normal distribution

This statistical law plays an important role in experimental science. This distribution represents a limit to which all other useful distributions approach under specific conditions often encountered in practical applications.

In particular, the values obtained for a quantity through a series of experiments follows closer a Gaussian distribution as the number of determinations becomes larger.

The p.d.f. (probability density function) of the Gaussian distribution is symmetric:

$$f(x) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{x-m}{\sigma}\right)^2},$$

Normalization :

$$\int_{-\infty}^{\infty} f(x)dx = \frac{1}{\sigma\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{-\frac{1}{2}\left(\frac{x-m}{\sigma}\right)^2} dx = \frac{\sigma}{\sigma\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{-\frac{1}{2}t^2} dt = 1.$$

**Expectation value :**

$$\int_{-\infty}^{\infty} x f(x) dx = \frac{1}{\sigma\sqrt{2\pi}} \int_{-\infty}^{\infty} x e^{-\frac{1}{2}\left(\frac{x-m}{\sigma}\right)^2} dx = \frac{1}{\sigma\sqrt{2\pi}} \int_{-\infty}^{\infty} \left(\sigma\sqrt{2}t + m\right) e^{-\frac{1}{2}\left(\frac{x-m}{\sigma}\right)^2} dt$$

$$= \frac{1}{\sqrt{\pi}} \left[ \sigma\sqrt{2} \int_{-\infty}^{\infty} t e^{-t^2} dt + m \int_{-\infty}^{\infty} e^{-t^2} dt \right] = \frac{\sqrt{\pi}}{\sqrt{\pi}} m = m,$$

**Variance :**

$$E\left[(x-m)^2\right] = \int_{-\infty}^{\infty} (x-m)^2 f(x) dx = \frac{1}{\sigma\sqrt{2\pi}} \int_{-\infty}^{\infty} (x-m)^2 e^{-\frac{1}{2}\left(\frac{x-m}{\sigma}\right)^2} dx$$

$$= \frac{2\sqrt{2}\sigma^3}{\sigma\sqrt{2\pi}} \int_{-\infty}^{\infty} t^2 e^{-t^2} dt = = \frac{\sigma^2}{\sqrt{\pi}} \left( \int_{-\infty}^{\infty} t 2t e^{-t^2} dt \right)$$

$$= \frac{\sigma^2}{\sqrt{\pi}} \left\{ -t e^{-t^2} \Big|_{-\infty}^{\infty} + \int_{-\infty}^{\infty} e^{-t^2} dt \right\} = \frac{\sqrt{\pi}\sigma^2}{\sqrt{\pi}} = \sigma^2.$$

**Standard deviation :**

$$\mu_2 = \sqrt{E\left[(x-m)^2\right]} = \sqrt{\sigma^2} = \sigma.$$

**In the area of radioactivity measurements, the normal distribution describes typically the lines appearing in the spectrum registered by a spectrometer based on a hyper-pure Germanium detector placed in the field of a radionuclide decaying by gamma emissions.**

## 4. Introduction of the Gaussian uncertainty propagation law

Up to now only the uncertainty of a measured quantity has been considered. However, usually the physical laws are functions of one or even several quantities, the value of which can be determined by appropriate measurements.

As a consequence these determinations are not exact but are obtained with uncertainties. The question arises then of knowing how to combine these individual uncertainties to calculate the resulting uncertainty of the physical function. The German mathematician Gauss pioneered this field in 1794 and developed in the following years a theoretical formalism, which is still in use at present and is known as the Gaussian uncertainty propagation law.

# Radioactivity measurements and uncertainty evaluation

In the two following slides the outline of the theory will be presented.

- *Case of one variable*

  Consider first a physical quantity $x$ known with an uncertainty $\Delta x$. $\Delta x$ should be taken as small ($\Delta x \ll 1$).

  Assume that a physical law is function of the quantity $x$ and of some constant parameters.

  So $f = f(x, a_1, \ldots a_n)$.

  Knowing the value of $f$ for $x$, the function has to be evaluated for $x + \Delta x$.

  The function can then be developed in a Taylor series and, as $\Delta x$ is small, the higher order in $\Delta x$ can be neglected.

# Radioactivity measurements and uncertainty evaluation

Therefore $f$ becomes

$$f(x + \Delta x, a_1, \ldots a_n) = f(x, a_1, \ldots a_n) + \Delta x\, f_x'(x, a_1, \ldots a_n) + O(\Delta x^2)$$

$$= f + \Delta f.$$

The change in $f$ induced by the change $\Delta x$ of $x$ is

$$\Delta f = \Delta x\, f_x'(x, a_1, \ldots a_n).$$

As before only positive changes are considered and the variance is taken as

$$(\Delta f)^2 = (\Delta x\, f_x'(x, a_1, \ldots a_n))^2$$

and the standard deviation is

$$u_f = \sqrt{\left(\Delta x\, f_x'\right)^2}$$

## *Some applications*

For $f(x, a_1,\dots a_n) = x$ then $\Delta f = \Delta x$

For $f(x, a_1,\dots a_n) = x^2$ then $\Delta f = 2\Delta x\, x$

or $\Delta f / f = 2\Delta x / x$ ,

which are all well-known relationships.

Further if the function represents the decay of a radioactive source

$f = e^{-\mathrm{Ln}(2)t/T1/2}$ then $\Delta f = \mathrm{Ln}(2)t\, \Delta T_{1/2} /(T_{1/2})^2\, e^{-\mathrm{Ln}(2)t/T1/2}$

or $\Delta f / f = \mathrm{Ln}(2)t / T_{1/2}(\Delta T_{1/2} / T_{1/2})$.

- ***Case of several variables***

**Often the functions encountered in measurement depend on more than just one variable, let say for instance temperature, pressure and hygrometry in the case of mass measurements. The question arises then to evaluate the uncertainty of the function when the uncertainties of all individual quantities are taken into account.**

**First let the function *f* be a function of three variables *x*, *y* and *z* with respective uncertainties $\Delta x$, $\Delta y$, $\Delta z$. These uncertainties are supposed small so that the function *f* can be developed into a Taylor series which can be interrupted after the first order.**

# Radioactivity measurements and uncertainty evaluation

Hence

$f(x + \Delta x, y + \Delta y, z + \Delta z, a_1, \ldots a_n) = f(x, y, z, a_1, \ldots a_n) + \Delta f$

with $$\Delta f = \Delta x \frac{\partial f}{\partial x} + \Delta y \frac{\partial f}{\partial y} + \Delta z \frac{\partial f}{\partial z} + \text{terms of higher order.}$$

As in the case of one variable we can evaluate the variance taken as $(\Delta f)^2$

$$(\Delta f)^2 \simeq \left(\Delta x \frac{\partial f}{\partial x} + \Delta y \frac{\partial f}{\partial y} + \Delta z \frac{\partial f}{\partial z}\right)^2 = \left(\Delta x \frac{\partial f}{\partial x}\right)^2 + \left(\Delta y \frac{\partial f}{\partial y}\right)^2 + \left(\Delta z \frac{\partial f}{\partial z}\right)^2$$
$$+ 2\Delta x \Delta y \frac{\partial f}{\partial x}\frac{\partial f}{\partial y} + 2\Delta x \Delta z \frac{\partial f}{\partial x}\frac{\partial f}{\partial z} + 2\Delta y \Delta z \frac{\partial f}{\partial y}\frac{\partial f}{\partial z}.$$

If the cross terms are neglected this reduces to

$$(\Delta f)^2 \simeq \left(\Delta x \frac{\partial f}{\partial x}\right)^2 + \left(\Delta y \frac{\partial f}{\partial y}\right)^2 + \left(\Delta z \frac{\partial f}{\partial z}\right)^2.$$

# Radioactivity measurements and uncertainty evaluation

The uncertainty is then given by $u \simeq \sqrt{\left(\Delta x \frac{\partial f}{\partial x}\right)^2 + \left(\Delta y \frac{\partial f}{\partial y}\right)^2 + \left(\Delta z \frac{\partial f}{\partial z}\right)^2}$.

The derivatives appearing in the preceding equation are called sensitivity coefficients. The above expression is rigorously true if the variables in ***f*** are independent.

***Example***

The ionization of an ionization chamber can be expressed by

***I = CU/t***. Its uncertainty becomes then $u_I = \sqrt{\left(\Delta C \frac{U}{t}\right)^2 + \left(\Delta U \frac{C}{t}\right)^2 + \left(\Delta t \frac{CU}{t^2}\right)^2}$

or when it is expressed with the relative uncertainties of the variables

$$\frac{u_I}{I} = \sqrt{\left(\frac{\Delta C}{C}\right)^2 + \left(\frac{\Delta U}{U}\right)^2 + \left(\frac{\Delta t}{t}\right)^2}.$$

**General formulae for the propagation of uncertainties when some correlations between the variables exist can be obtained in a more direct approach.**

**For this purpose consider a function *f* of *i* = 1, … , *n* variables, $x_i$, each of them having a mean value $E(x_i) = \mu_i$ and an uncertainty $u(x_i)$. As previously, for small deviations of the variable around its mean value the function *f* can be expanded in a first-order Taylor series. All higher orders are assumed to be negligible.**

$$f - \bar{f} = \sum_{i=1}^{n} \frac{\partial f}{\partial x_i}(x_i - \mu_i)$$

**The square of the above expression is**

$$\left(f - \bar{f}\right)^2 = \left( \sum_{i=1}^{n} \frac{\partial f}{\partial x_i}(x_i - \mu_i) \right)^2$$

# Radioactivity measurements and uncertainty evaluation

or after development

$$\left(f-\overline{f}\right)^2=\sum_{i=1}^{n}\left(\frac{\partial f}{\partial x_i}\right)^2(x_i-\mu_i)^2+\sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\frac{\partial f}{\partial x_i}\frac{\partial f}{\partial x_j}(x_i-\mu_i)(x_j-\mu_j).$$

It becomes after the expectation of both sides are taken

$$E[(f-\overline{f})^2]=\sum_{i=1}^{n}\left(\frac{\partial f}{\partial x_i}\right)^2 E\left[(x_i-\mu_i)^2\right]+\sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\frac{\partial f}{\partial x_i}\frac{\partial f}{\partial x_j}E\left[(x_i-\mu_i)\right]E\left[(x_j-\mu_j)\right],$$

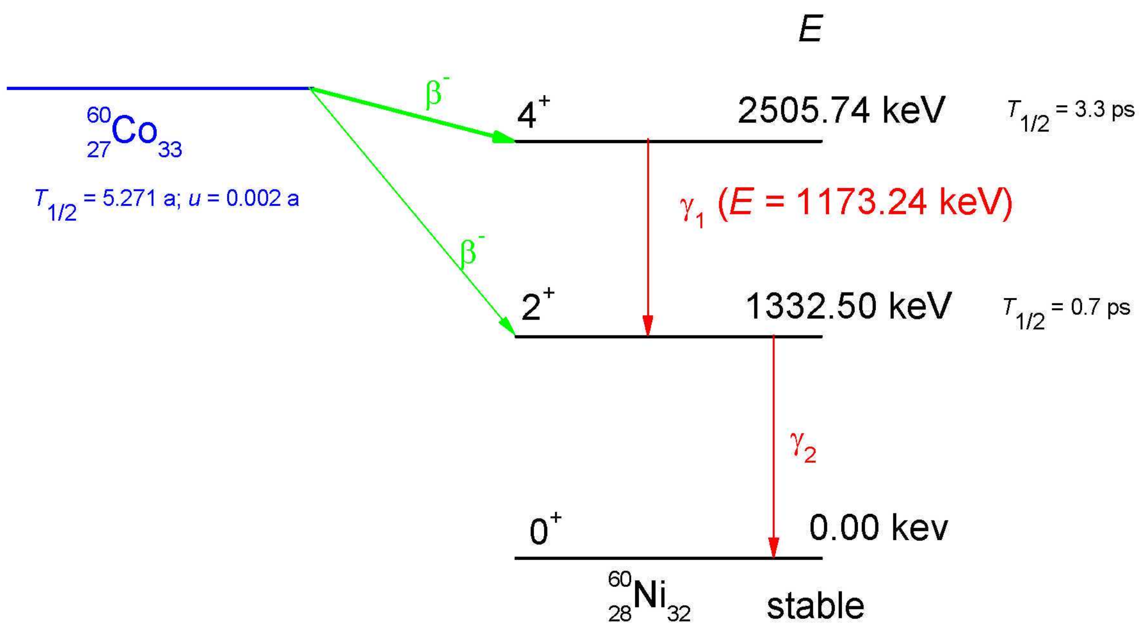
which is the value of the variance of the function ***f*** and can be written using the usual symbol for variance as

$$\sigma_f^2=\sum_{i=1}^{n}\left(\frac{\partial f}{\partial x_i}\right)^2\sigma_i^2+\sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\frac{\partial f}{\partial x_i}\frac{\partial f}{\partial x_j}\underbrace{E\left[(x_i-\mu_i)\right]E\left[(x_j-\mu_j)\right]}_{\text{covariance}}.$$

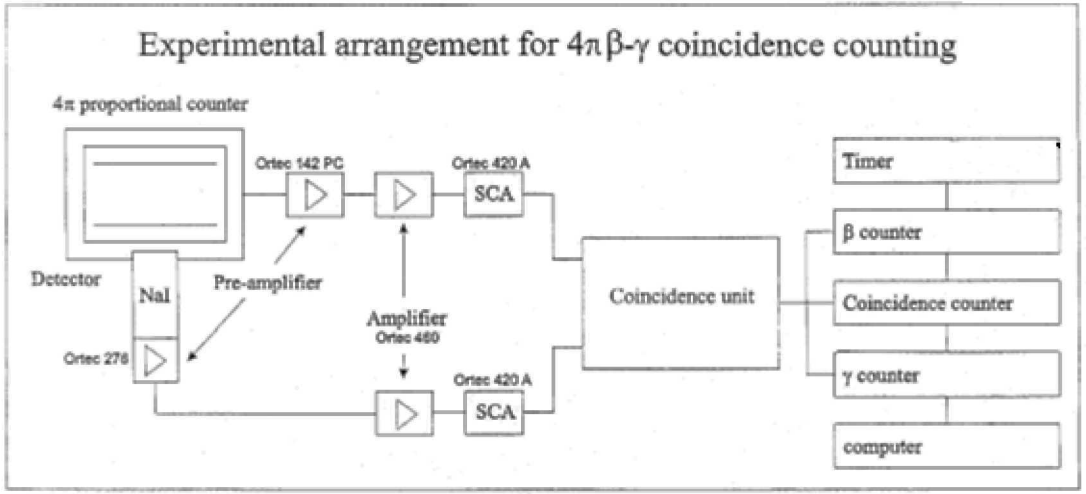
# 5. Applications

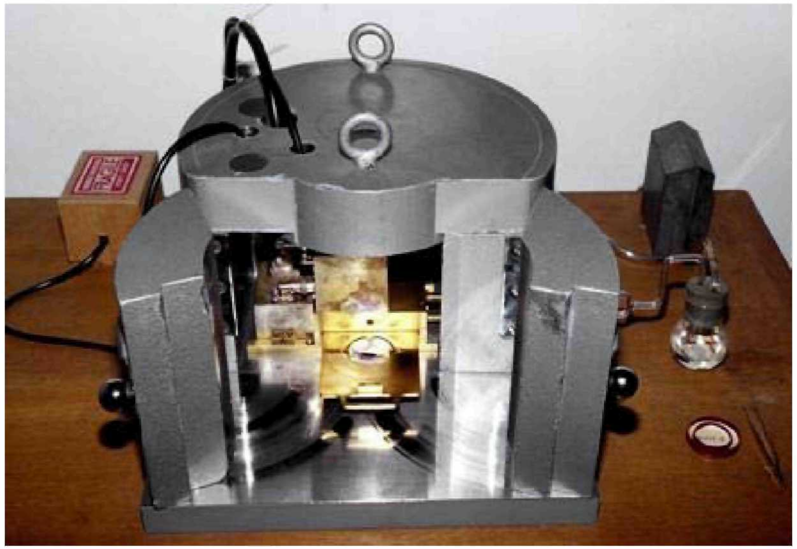
## a) Coincidence measurement of a β-γ emitter

Simplified decay scheme of $^{60}Co$



# Radioactivity measurements and uncertainty evaluation



# Radioactivity measurements and uncertainty evaluation

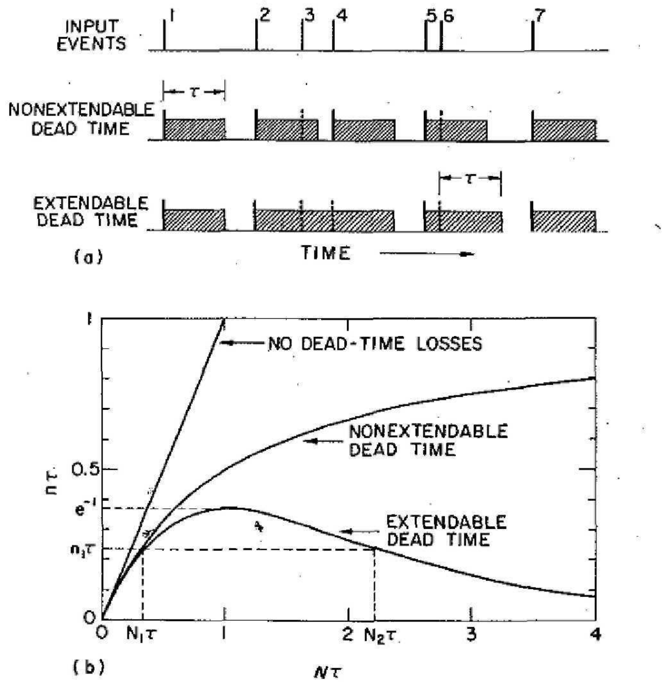
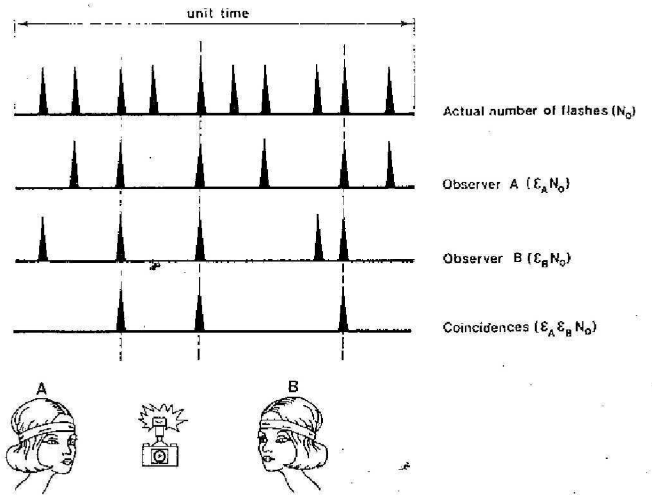



**Fig. 18.** Nonextendable and extendable dead times. (a) A sequence of input events (top row) is shown with the corresponding outputs from a nonextendable dead time (middle row) and an extendable dead time (bottom row). With the nonextendable dead time, events 3 and 6 are lost but 4 survives because it is separated by more than $\tau$ from 2, the last preceding event to produce an output. With the extendable dead time, event 4 is lost also because it is separated by less than $\tau$ from 3 which extended the dead time initiated by 2. Event 6 extended the dead time initiated by 5 but not by enough to block the following event. (b) The observed output rates as functions of the input rate for the two types of dead time are shown as plots of $n\tau$ *vs* $N\tau$. (This is equivalent to plotting $n$ *vs* $N$ both in units of $\tau^{-1}$.) Note that with an extendable dead time, the output rate $n_1$ could be observed for two input rates, $N_1$ or the many times larger $N_2$.

# Radioactivity measurements and uncertainty evaluation




Efficiency of observer A $\varepsilon_A = \frac{N_A}{N_o} = 60\%$ (A observes $0.6\,N_o$ flashes in unit time)

Efficiency of observer B $\varepsilon_B = \frac{N_B}{N_o} = 50\%$ (B observes $0.5\,N_o$ flashes in unit time)

on average B observes 50% of those flashes that are observed by A

(i.e. $0.3\,N_o$ flashes in unit time)

i.e. number of coincidences $= 0.3\,N_o = \varepsilon_A \varepsilon_B N_o = \frac{N_A}{N_o} \cdot \frac{N_B}{N_o} \cdot N_o$

or $N_o = \frac{N_A \cdot N_B}{\text{number of coincidences}} = \frac{6 \times 5}{3} = 10$

Fig. 5-17 The principle of activity measurement by coincidence counting. As was done in the early days, each observer used a low-power telescope and recorded the occurence of a flash on the ZnS(Ag) screen by pressing a key that caused a needle to move across a strip of smoked paper.

$$N_{\beta\text{cor}} = \frac{N_\beta}{(1 - N_\beta \tau_\beta)}$$

$$N_{\gamma\text{cor}} = \frac{N_\gamma}{(1 - N_\gamma \tau_\gamma)}$$

$$N'_\beta = N_{\beta\text{cor}} - B_\beta,$$

$$N'_\gamma = N_{\gamma\text{cor}} - B_\gamma.$$

$$N'_c = N_c - B_c.$$

$$N_{c\,\text{rad}} = 2\tau_r N'_\beta N'_\gamma.$$

# Radioactivity measurements and uncertainty evaluation

$$N'_{\beta} = N_0 \left[ \varepsilon_{\beta} + (1 - \varepsilon_{\beta}) \frac{\alpha \left[ \varepsilon_{ce} + (1 - \varepsilon_{ce}) \varepsilon_x \right] + \varepsilon_{\beta\gamma}}{1 + \alpha} \right]$$

$$N'_{\beta} = N_0 \varepsilon_{\beta}$$ **for $^{60}$Co**

$$N'_{\gamma} = N_0 \varepsilon_{\gamma}$$

$$N'_{c\,true} = \varepsilon_{\beta} \varepsilon_{\gamma} N_0 = N'_c - 2\tau_r N_{\beta} N_{\gamma},$$

$$N_0 = \frac{N'_{\beta} N'_{\gamma}}{N'_c - 2\tau_r N'_{\beta} N'_{\gamma}},$$

**which, in absence of corrections, reduces to**

$$N_0 = \frac{N'_{\beta} N'_{\gamma}}{N'_c}.$$

# Radioactivity measurements and uncertainty evaluation

**The activity of the source is then given by**

$$N_0 = \left\{ \frac{N_\beta}{(1 - N_\beta \tau_\beta)} - B_\beta \right\} \left\{ \frac{N_\gamma}{(1 - N_\gamma \tau_\gamma)} - B_\gamma \right\} \times \frac{1}{\left( N_c - B_c \right)} \times e^{-\frac{\mathrm{Ln}(2) \times t}{T_{1/2}}},$$

**and the activity concentration of the source by**

$$C_0 = \left\{ \frac{N_\beta}{(1 - N_\beta \tau_\beta)} - B_\beta \right\} \left\{ \frac{N_\gamma}{(1 - N_\gamma \tau_\gamma)} - B_\gamma \right\} \times \frac{1}{\left( N_c - B_c \right)} \times e^{-\frac{\mathrm{Ln}(2) \times t}{T_{1/2}}} \times \frac{1}{m}.$$

## *Evaluation of uncertainties*

**First evaluate the sensibility coefficients for the variable $N_\beta$:**

$$\frac{\partial C_0}{\partial N_\beta} = \left\{ \frac{N_\gamma}{(1 - N_\gamma \tau_\gamma)} - B_\gamma \right\} \times \frac{1}{(N_c - B_c)} \times e^{-\frac{\mathrm{Ln}(2) \times t}{T_{1/2}}} \times \frac{1}{m} \times \frac{\partial}{\partial N_\beta} \left\{ \frac{N_\beta}{(1 - N_\beta \tau_\beta)} - B_\beta \right\},$$

$$= \left\{ \frac{N_\gamma}{(1 - N_\gamma \tau_\gamma)} - B_\gamma \right\} \times \frac{1}{(N_c - B_c)} \times e^{-\frac{\mathrm{Ln}(2) \times t}{T_{1/2}}} \times \frac{1}{m} \times \frac{1}{(1 - N_\beta \tau_\beta)^2},$$

$$= C_0 \frac{1}{\left(N_\beta - B_\beta (1 - N_\beta \tau_\beta)\right)(1 - N_\beta \tau_\beta)}.$$

**A similar expression is found for $N\gamma$ :**

$$\frac{\partial C_0}{\partial N_\gamma} = C_0 \frac{1}{\left(N_\gamma - B_\gamma (1 - N_\gamma \tau_\gamma)\right)(1 - N_\gamma \tau_\gamma)}.$$

# Radioactivity measurements and uncertainty evaluation

**And so on for the variables $\tau_\beta$, $\tau_\gamma$, $N_c$, $T_{1/2}$, $m$**

$$\frac{\partial C_0}{\partial \tau_\beta} = C_0 \frac{N_\beta^2}{(1-N_\beta \tau_\beta)\left(N_\beta - B_\beta\left(1-N_\beta\tau_\beta\right)\right)},$$

$$\frac{\partial C_0}{\partial \tau_\beta} = C_0 \frac{N_\gamma^2}{(1-N_\gamma \tau_\gamma)\left(N_\gamma - B_\gamma\left(1-N_\gamma\tau_\gamma\right)\right)},$$

$$\frac{\partial C_0}{\partial N_c} = C_0\left(-\frac{1}{N_c - B_c}\right),$$

$$\frac{\partial C_0}{\partial T_{1/2}} = \left\{\frac{N_\beta}{(1-N_\beta\tau_\beta)} - B_\beta\right\} \times \left\{\frac{N_\gamma}{(1-N_\gamma\tau_\gamma)} - B_\gamma\right\} \times \frac{1}{\left(N_c - B_c\right)} \times \frac{1}{m} \times \frac{\partial}{\partial T_{1/2}} e^{-\frac{\mathrm{Ln}(2)\times t}{T_{1/2}}}$$

$$= \left\{\frac{N_\beta}{(1-N_\beta\tau_\beta)} - B_\beta\right\} \times \left\{\frac{N_\gamma}{(1-N_\gamma\tau_\gamma)} - B_\gamma\right\} \times \frac{1}{\left(N_c - B_c\right)} \times \frac{1}{m} \times \left(\frac{\mathrm{Ln}(2)\times t}{T_{1/2}^{\;2}}\right) \times e^{-\frac{\mathrm{Ln}(2)\times t}{T_{1/2}}}$$

$$= C_0\left(\frac{\mathrm{Ln}(2)\times t}{T_{1/2}^{\;2}}\right),$$

$$\frac{\partial C_0}{\partial m} = C_0\left(-\frac{1}{m}\right).$$

# Radioactivity measurements and uncertainty evaluation

**Finally the uncertainty of the activity concentration becomes**

$$u_{C_0}^2 = \left(\frac{\partial}{\partial N_\beta} C_0\right)^2 u_{N_\beta}^2 + \left(\frac{\partial}{\partial N_\gamma} C_0\right)^2 u_{N_\gamma}^2 + \left(\frac{\partial}{\partial N_c} C_0\right)^2 u_{N_c}^2 + \left(\frac{\partial}{\partial \tau_\beta} C_0\right)^2 u_{\tau_\beta}^2 +$$

$$\left(\frac{\partial}{\partial \tau_\gamma} C_0\right)^2 u_{\tau_\gamma}^2 + \left(\frac{\partial}{\partial T_{1/2}} C_0\right)^2 u_{T_{1/2}}^2 + \left(\frac{\partial}{\partial m} C_0\right)^2 u_m^2.$$

**Sometimes an expression for the relative uncertainty is also useful**

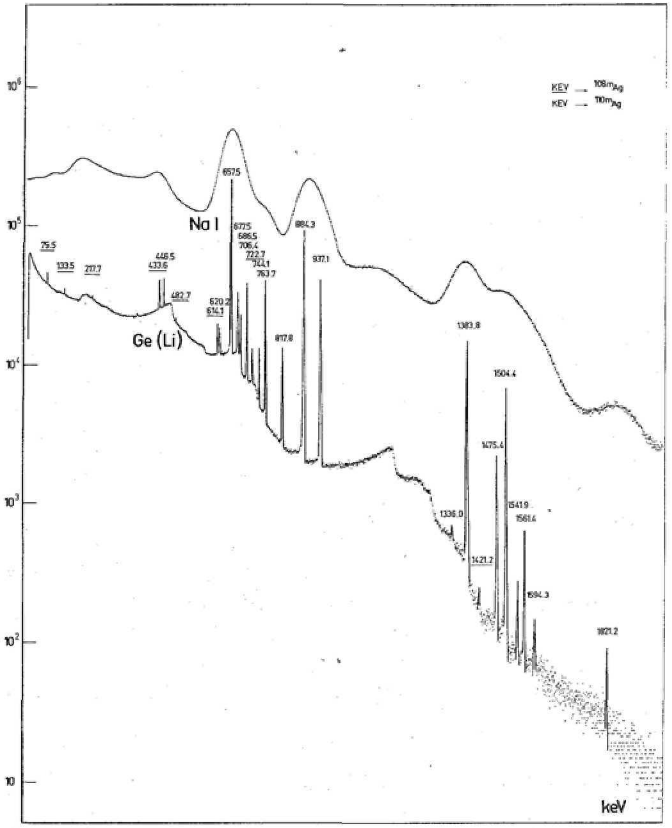
$$\frac{u_{C_0}^2}{C_0^2} = \left[\frac{1}{\left(N_\beta - B_\beta(1 - N_\beta\tau_\beta)\right)(1 - N_\beta\tau_\beta)}\right]^2 u_{N_\beta}^2 + \left[\frac{1}{\left(N_\gamma - B_\gamma(1 - N_\gamma\tau_\gamma)\right)(1 - N_\gamma\tau_\gamma)}\right]^2 u_{N_\gamma}^2 +$$

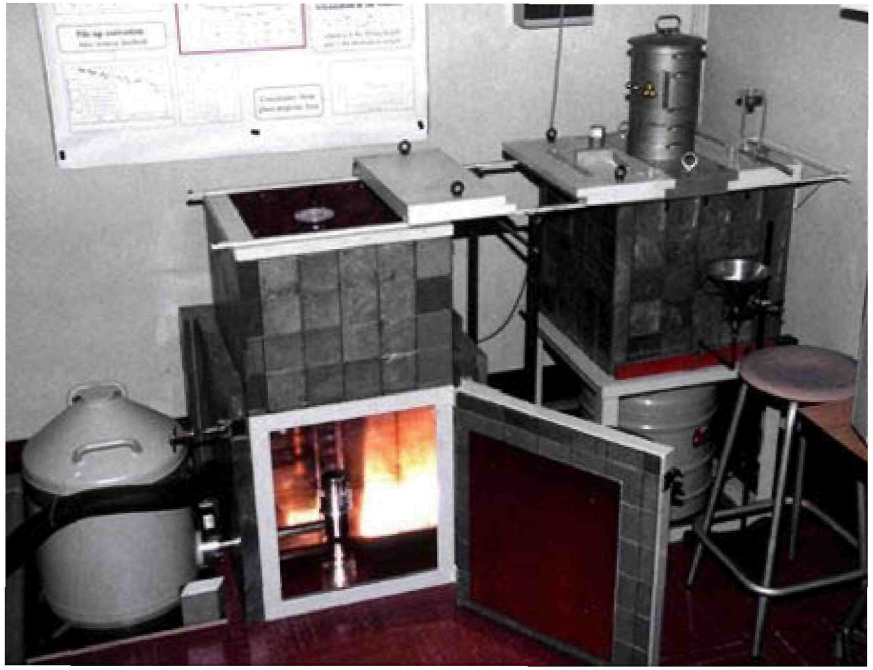
$$\left[\frac{1}{\left(N_c - B_c\right)}\right]^2 u_{N_c}^2 + \left[\frac{N_\beta}{\left(1 - N_\beta\tau_\beta\right)} - B_\beta\right]^2 u_{\tau_\beta}^2 + \left[\frac{N_\gamma}{\left(1 - N_\gamma\tau_\gamma\right)} - B_\gamma\right]^2 u_{\tau_\gamma}^2 +$$

$$\left[\frac{\mathrm{Ln}(2)t}{T_{1/2}^2}\right]^2 u_{T_{1/2}}^2 + \left[\frac{1}{m}\right]^2 u_m^2.$$

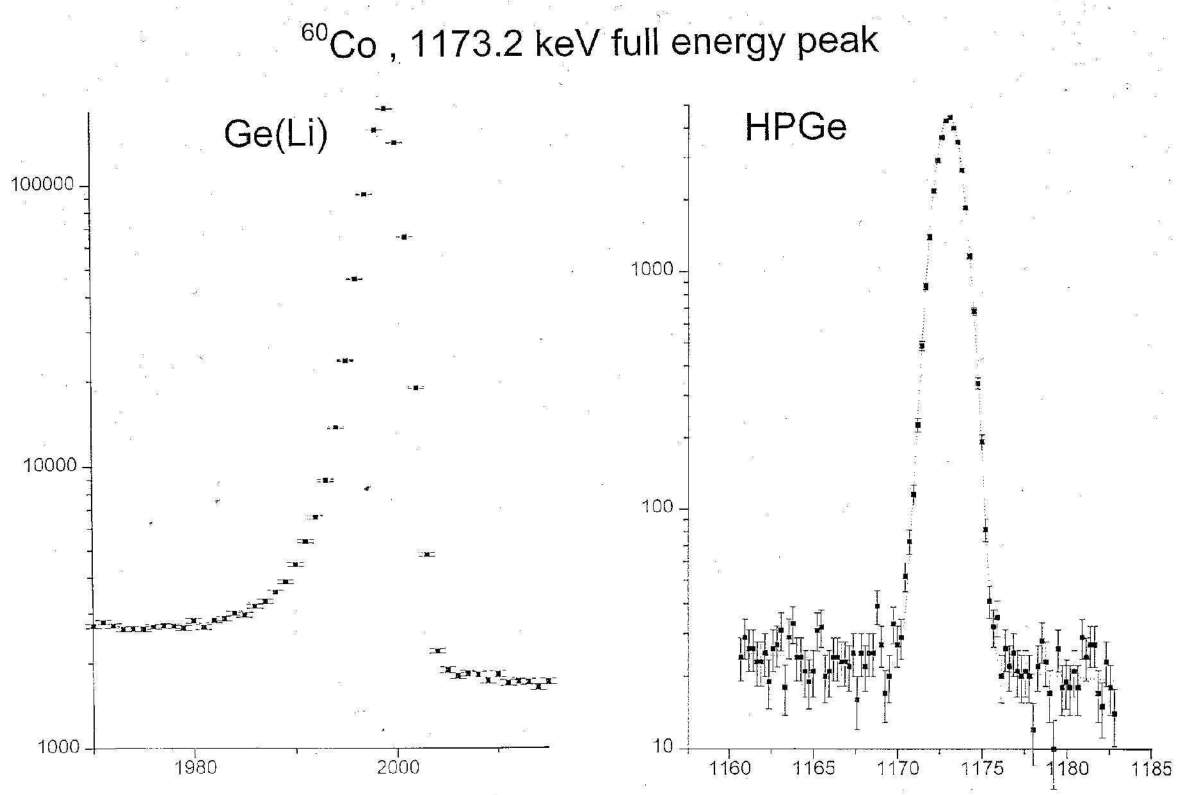
# Radioactivity measurements and uncertainty evaluation

## b) Determination of the activity of a pure γ emitter



# Radioactivity measurements and uncertainty evaluation



# Radioactivity measurements and uncertainty evaluation

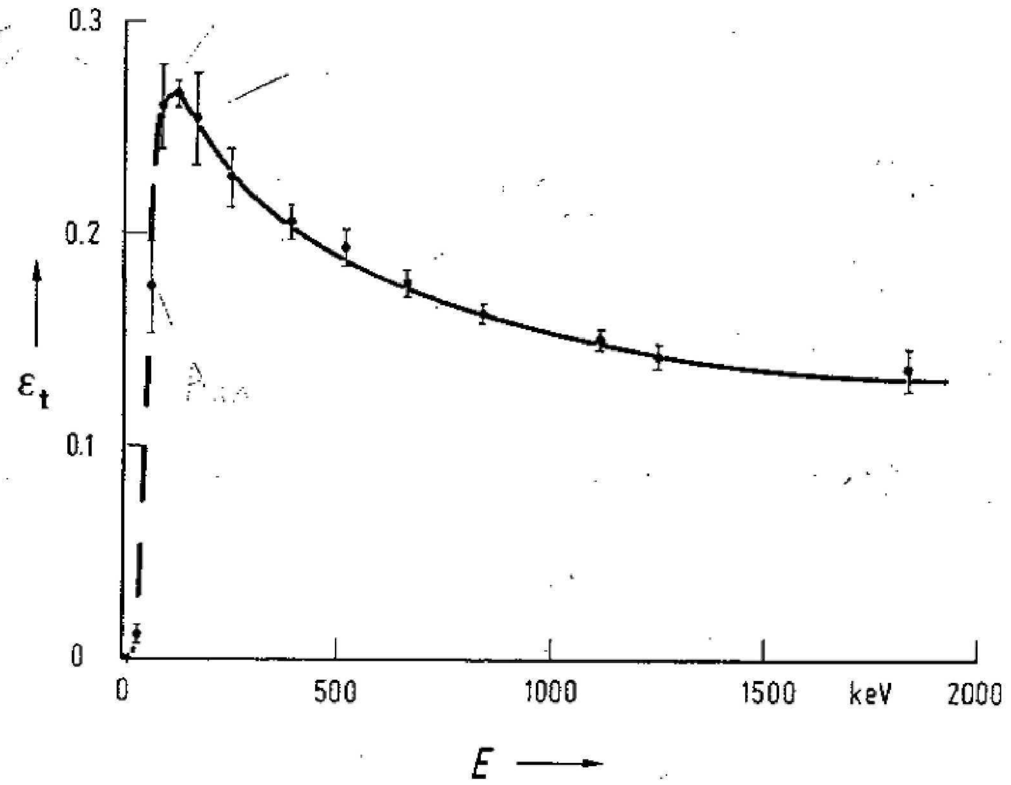



Fig. 4.18. Total efficiency for a Ge(Li) detector ($\epsilon_r = 12.5\%$) with the source mounted on top of the detector window.

# Radioactivity measurements and uncertainty evaluation

**In this case the activity is given by the expression**

$$A_0 = \frac{\left(N_i - B_{\gamma i}\right)}{p_i \varepsilon_i \left(E_i\right)} \prod_{j=1}^{l} K_j \times e^{-\frac{\mathrm{Ln}(2)t}{T_{1/2}}},$$

**where the coefficients $K_j$ represent the possible correction factors to take into account phenomena such as pile-up, dead-time effects and K-x-ray escape. The expression for the activity concentration becomes**

$$C_0 = \frac{A_0}{m} = \frac{\left(N_i - B_{\gamma i}\right)}{p_i \varepsilon_i \left(E_i\right)} \prod_{j=1}^{l} K_j \times e^{-\frac{\mathrm{Ln}(2)t}{T_{1/2}}} \times \frac{1}{m}.$$

# Radioactivity measurements and uncertainty evaluation

**As for 4πβγ measurements the sensitivity coefficients are required to evaluate the uncertainty.**

$$\frac{\partial A_0}{\partial p_i}=\left\{\frac{\left(N_i-B_{\gamma i}\right)}{\varepsilon_i\left(E_i\right)}\prod_{j=1}^{l}K_j\times e^{-\frac{\mathrm{Ln}(2)t}{T_{1/2}}}\right\}\times-\frac{1}{\left(p_i\right)^2}=-\frac{A_0}{p_i},$$

$$\frac{\partial A_0}{\partial N_i}=\left\{\frac{1}{p_i\varepsilon_i\left(E_i\right)}\prod_{j=1}^{l}K_j\times e^{-\frac{\mathrm{Ln}(2)t}{T_{1/2}}}\right\}=\frac{A_0}{\left(N_i-B_{\gamma i}\right)},$$

$$\frac{\partial A_0}{\partial \varepsilon_i\left(E_i\right)}=\left\{\frac{\left(N_i-B_{\gamma i}\right)}{p_i}\prod_{j=1}^{l}K_j\times e^{-\frac{\mathrm{Ln}(2)t}{T_{1/2}}}\right\}\times-\frac{1}{\left[\varepsilon_i\left(E_i\right)\right]^2}=-\frac{A_0}{\varepsilon_i\left(E_i\right)},$$

$$\frac{\partial A_0}{\partial K_k}=\left\{\frac{\left(N_i-B_{\gamma i}\right)}{p_i\varepsilon_i\left(E_i\right)}\prod_{j\neq k=1}^{l}K_j\times e^{-\frac{\mathrm{Ln}(2)t}{T_{1/2}}}\right\}=\frac{A_0}{K_k},\ \forall k,\ \text{and}$$

$$\frac{\partial A_0}{\partial T_{1/2}}=\left\{\frac{\left(N_i-B_{\gamma i}\right)}{p_i\varepsilon_i\left(E_i\right)}\prod_{j=1}^{l}K_j\times e^{-\frac{\mathrm{Ln}(2)t}{T_{1/2}}}\right\}\times\left(-\frac{\mathrm{Ln}(2)t}{T_{1/2}^2}\right)=A_0\times\left(-\frac{\mathrm{Ln}(2)t}{T_{1/2}^2}\right).$$

# Radioactivity measurements and uncertainty evaluation

**For the activity concentration an additional term has to be evaluated**

$$\frac{\partial C_0}{\partial m} = \left\{ \frac{\left(N_i - B_{\gamma i}\right)}{p_i \varepsilon_i \left(E_i\right)} \prod_{j=1}^{l} K_j \times e^{-\frac{\mathrm{Ln}(2)t}{T_{1/2}}} \right\} \times -\frac{1}{m^2} = -\frac{C_0}{m}.$$

**Thus the uncertainty of the activity concentration becomes**

$$u_{C_0}^2 = \left(\frac{\partial}{\partial N_i} C_0\right)^2 u_{N_i}^2 + \left(\frac{\partial}{\partial \varepsilon_i (E_i)} C_0\right)^2 u_{\varepsilon_i(E_i)}^2 + \left(\frac{\partial}{\partial p_i} C_0\right)^2 u_{p_i}^2$$

$$+ \sum_{j=1}^{l} \left(\frac{\partial}{\partial K_j} C_0\right)^2 u_{K_j}^2 + \left(\frac{\partial}{\partial T_{1/2}} C_0\right)^2 u_{T_{1/2}}^2 + \left(\frac{\partial}{\partial m} C_0\right)^2 u_m^2.$$

**and immediately its relative uncertainty**

$$\frac{u_{C_0}^2}{C_0^2} = \frac{1}{p_i^2} u_{p_i}^2 + \frac{1}{\varepsilon_i^2} u_{\varepsilon_i}^2 + \left(\frac{1}{N_i - B_{\gamma i}}\right)^2 u_{N_i}^2 + \sum_{j=1}^{l} \frac{1}{K_j^2} u_{K_j}^2 + \left(\frac{\mathrm{Ln}(2)t}{T_{1/2}^2}\right)^2 u_{T_{1/2}}^2 + \frac{1}{m^2} u_m^2.$$

## Radioactivity measurements and uncertainty evaluation

**If the relative uncertainties of all variables are used the preceding expression can be written in a more condensed way :**

$$\frac{r^2_{C_0}}{C_0^2} = r^2_{p_i} + r^2_{\varepsilon_i} + \frac{1}{N_i} + \sum_{j=1}^{l} r^2_{K_j} + \left[\frac{\mathrm{Ln}(2)t}{T_{1/2}}\right]^2 r^2_{T_{1/2}} + r^2_m.$$

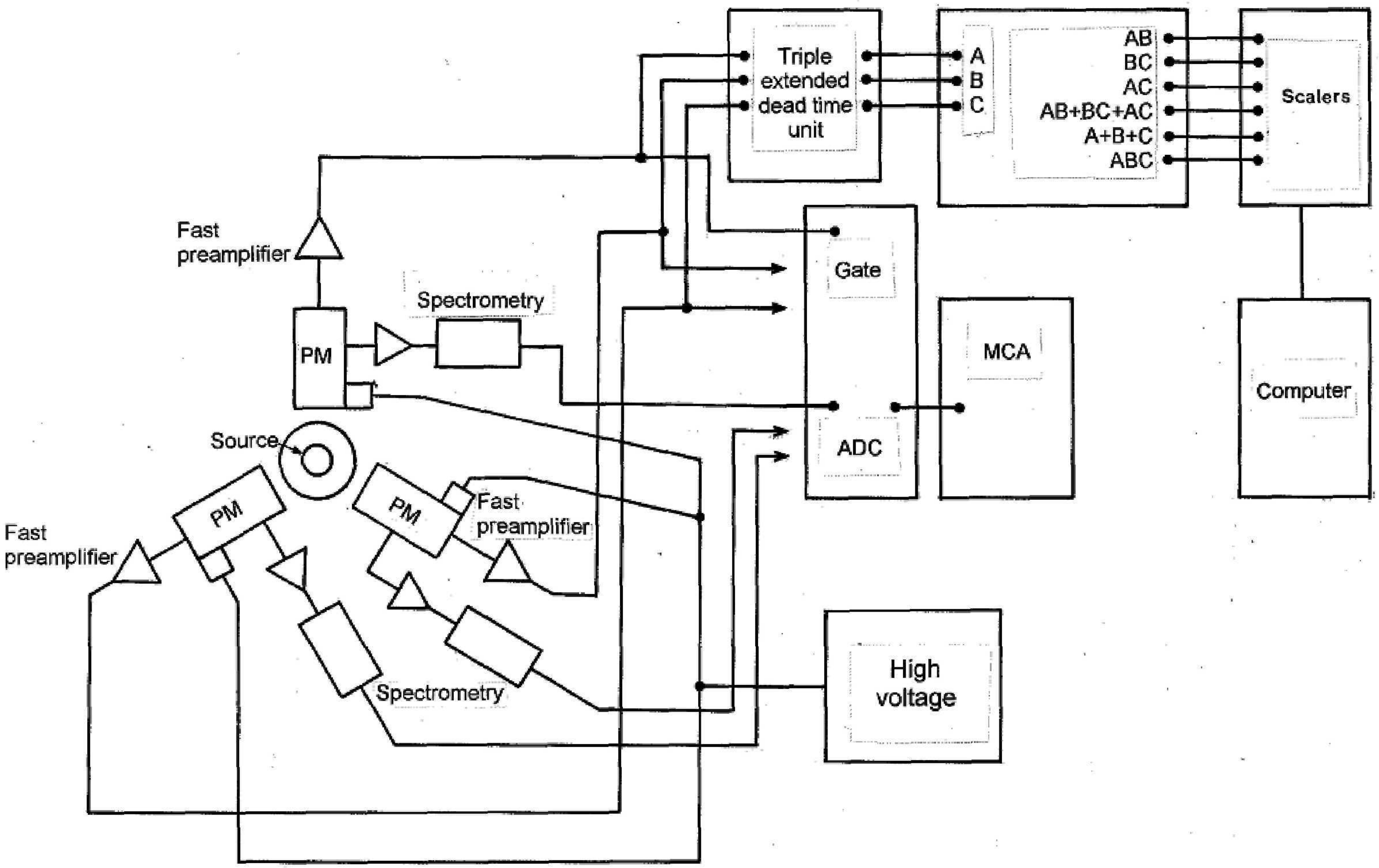
**where it was assumed that the background was negligible** $\left(N_i - B_{\gamma i}\right)^2 \approx N_i^2$,

**and using the property that in the case of a Poisson distribution the standard deviation of a quantity is equal to the square root of this quantity.**

# Radioactivity measurements and uncertainty evaluation

c) Evaluation of the uncertainty on the Triple-to-Double Ratio (*TDCR*) in the TDCR-method

# Radioactivity measurements and uncertainty evaluation



# Radioactivity measurements and uncertainty evaluation

The total number of logical double coincidences is given by

$$N_{\mathrm{D}}^{\mathrm{logical}} = N_0 \varepsilon_{\mathrm{D}},$$

and the number of triple coincidences by

$$N_{123} = N_0 \varepsilon_{123}.$$

The number of logical coincidences can also be expressed in function of the triple coincidences and of the three registered double coincidences as

$$N_{\mathrm{D}}^{\mathrm{logical}} = N_{12} + N_{13} + N_{23} - 2N_{123}.$$

# Radioactivity measurements and uncertainty evaluation

**The TDCR ratio can be then defined by**

$$TDCR = \frac{\varepsilon_{123}}{\varepsilon_{\mathrm{D}}} = \frac{N_{123}}{N_{\mathrm{D}}^{\mathrm{logical}}}.$$

$$TDCR = \frac{N_{123} - N_{123}^{\mathrm{b}}}{\left(N_{\mathrm{D}}^{\mathrm{logical}} - N_{\mathrm{D}}^{\mathrm{b,logical}}\right)} \quad \text{or} \quad TDCR = \frac{N'_{123}}{\frac{1}{2}\sum\limits_{\substack{i,j=1,\\ i\neq j}}^{3} N'_{i,j} - 2N'_{123}},$$

**after the numbers of counts have been corrected for the background.**

**Evaluate now the different sensitivity coefficients**

$$\frac{\partial TDCR}{\partial N'_{123}} = \frac{1}{\left(N'_{12} + N'_{13} + N'_{23} - 2N'_{123}\right)} + \frac{2N'_{123}}{\left(N'_{12} + N'_{13} + N'_{23} - 2N'_{123}\right)^2}$$

$$= \frac{N'_{12} + N'_{13} + N'_{23}}{\left(N'_{12} + N'_{13} + N'_{23} - 2N'_{123}\right)^2}.$$

## Radioactivity measurements and uncertainty evaluation

**Because of the symmetry in the three phototubes the other required partial derivatives have similar expressions**

$$\frac{\partial TDCR}{\partial N'_{12}} = \frac{\partial TDCR}{\partial N'_{13}} = \frac{\partial TDCR}{\partial N'_{23}}$$

$$\frac{\partial TDCR}{\partial N'_{\substack{i,j=1,3\\ i\neq j}}} = -\frac{N'_{123}}{\left(N'_{12}+N'_{13}+N'_{23}-2N'_{123}\right)^2}.$$

**The uncertainty of the *TDCR* ratio becomes then**

$$u^2_{TDCR} = \left(\frac{\partial TDCR}{\partial N'_{123}}\right)^2 u^2_{N'_{123}} + \left(\frac{\partial TDCR}{\partial N'_{12}}\right)^2 u^2_{N'_{12}} + \left(\frac{\partial TDCR}{\partial N'_{13}}\right)^2 u^2_{N'_{13}} + \left(\frac{\partial TDCR}{\partial N'_{23}}\right)^2 u^2_{N'_{23}},$$

# Radioactivity measurements and uncertainty evaluation

or $$u_{TDCR} = \sqrt{\left(\frac{\partial TDCR}{\partial N'_{123}}\right)^2 N'_{123} + \left(\frac{\partial TDCR}{\partial N'_{12}}\right)^2 N'_{12} + \left(\frac{\partial TDCR}{\partial N'_{13}}\right)^2 N'_{13} + \left(\frac{\partial TDCR}{\partial N'_{23}}\right)^2 N'_{23}},$$

**when the property of a Poisson distribution is used.**

**Ideally the three phototubes are matched and give identical responses and $u_{TDCR}$ *becomes***

$$u_{TDCR} = \sqrt{\left(\frac{\partial TDCR}{\partial N'_{123}}\right)^2 N'_{123} + 3\left(\frac{\partial TDCR}{\partial N'}\right)^2 N'}$$

$$= \frac{\sqrt{3N'_{123}N'\left(3N' + N'_{123}\right)}}{\left(3N' - 2N'_{123}\right)^2}.$$

# Radioactivity measurements and uncertainty evaluation

**If the three phototubes are not exactly matched but differ slightly in efficiency so that**

$$N'_{13} < N'_{12} < N'_{23}$$

**with**

$$N'_{12} = N', \; N'_{13} = N'(1-\varepsilon) \text{ and } N'_{23} = N'(1+\varepsilon').$$

$$u_{TDCR} = \frac{\sqrt{\left(3N' + (\varepsilon - \varepsilon')N'\right)N'_{123}\left(3N' + (\varepsilon - \varepsilon')N' + N'_{123}\right)}}{\left(3N' + (\varepsilon - \varepsilon')N' - 2N'_{123}\right)^2}.$$

**This expression reduces to that obtained for three identical phototubes when** $(\varepsilon - \varepsilon') = 0.$

## 6. Treatment of correlations occurring when combining results of two measurement methods

**The expressions obtained for the 4$\pi\beta\gamma$ coincidence method and the $\gamma$ counting depend both on the half-life and the mass of the sources used for the measurements. The two evaluations are therefore correlated through these two values.**

**The covariance associated with these two activity concentrations can be expressed as**

$$u\left(C_0^{\beta\text{-}\gamma}, C_0^{\text{spect.}}\right) = \sum_{i=1}^{L} \frac{\partial C_0^{\beta\text{-}\gamma}}{\partial q_i} \frac{\partial C_0^{\text{spect.}}}{\partial q_i} u^2(q_i), \text{ with } L = 2.$$

# Radioactivity measurements and uncertainty evaluation

**In a more explicit way this can be written as**

$$u\left(C_0^{\beta\text{-}\gamma}, C_0^{\text{spect.}}\right) = \frac{\partial C_0^{\beta\text{-}\gamma}}{\partial T_{1/2}} \frac{\partial C_0^{\text{spect.}}}{\partial T_{1/2}} u^2(T_{1/2}) + \frac{\partial C_0^{\beta\text{-}\gamma}}{\partial m} \frac{\partial C_0^{\text{spect.}}}{\partial m} u^2(m).$$

**Evaluating the derivatives in function of *m* and $T_{1/2}$ and replacing their respective expressions in the above equations one then obtains**

$$u\left(C_0^{\beta\text{-}\gamma}, C_0^{\text{spect.}}\right) = C_0^{\beta\text{-}\gamma} C_0^{\text{spect.}} \left\{ \left( -\frac{\text{Ln}(2)t}{T_{1/2}^2} \right)^2 u^2(T_{1/2}) + \left( \frac{1}{m^2} \right)^2 u^2(m) \right\}.$$

**Thank you for your attention!**