The European Commission's science and knowledge service

Joint Research Centre

Low level instrumentation

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Course on gamma-ray spectrometry
CEA, Paris, June 12, 2018

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Literature

Product catalogues of manufacturers

- **Ortec**
- **Canberra (Mirion)**
- **(Eurisys)**
- **DSG**
- **(PGT)**
- **(Tennelec)**
- **(Oxford Instr.)**

K. Debertin and R.G. Helmer, "Gamma- and X-ray spectrometry with semiconductor detectors", North-Holland (Elsevier), 1988

G. Gilmore and J. Hemingway, "Practical gamma-ray spectrometry", Wiley, 1995

M.F. Annunziata Ed., Handbook of Radioactivity Analysis, Academic Press, 2003

R. Jenkins, R.W. Gould and D. Gedcke , Quantitative X-ray Spectrometry Dekker Inc., 1995

Kai Siegbahn,

Wikipedia – (surprisingly) good source of information for physics

The High Purity Germanium detector (HPGe-detector

The "Work-horse" of the modern gamma-ray spectrometry laboratory

“good” resolution
“good” efficiency
Easy to operate
Reasonable robust …….
Price...
......

Useful for: Radiopurity studies, investigate unknown samples, secondary standardisation etc.

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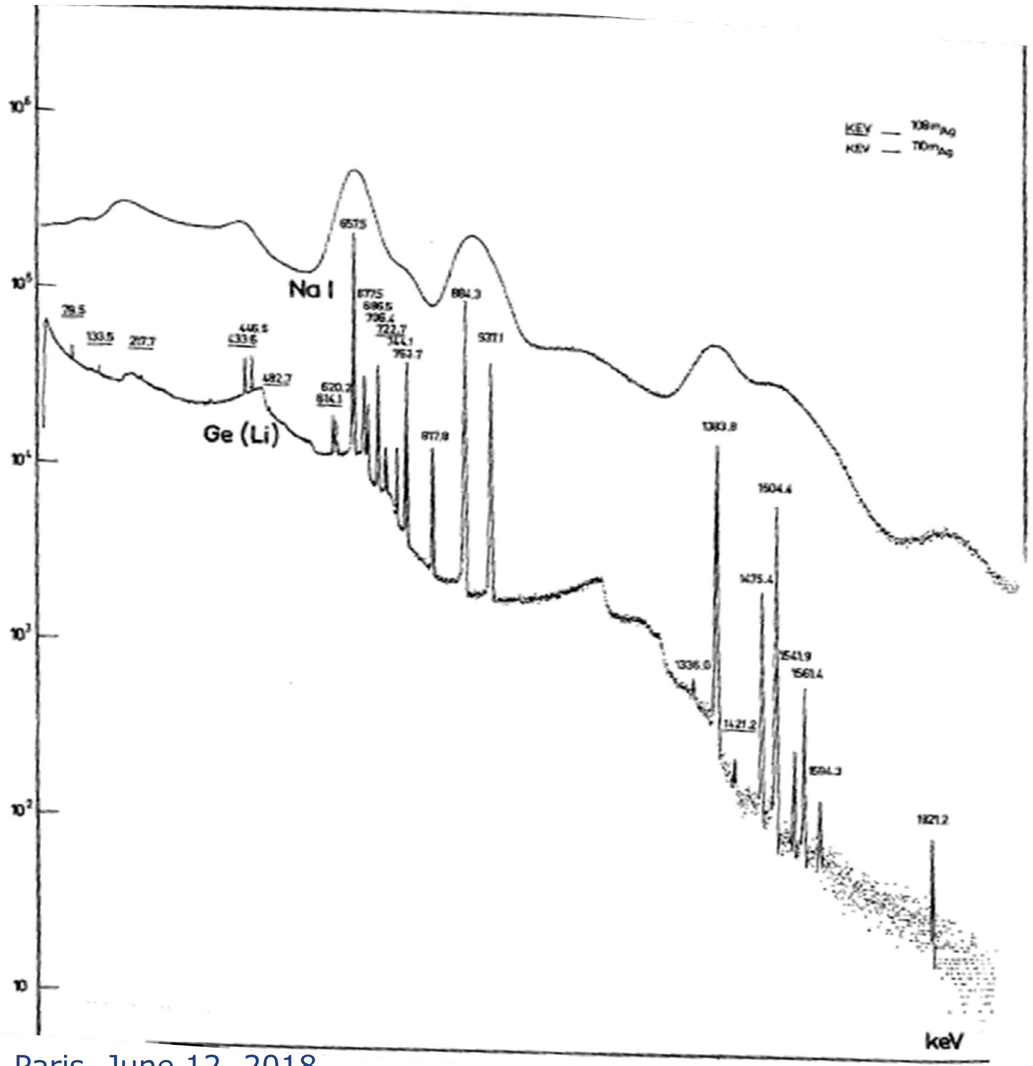
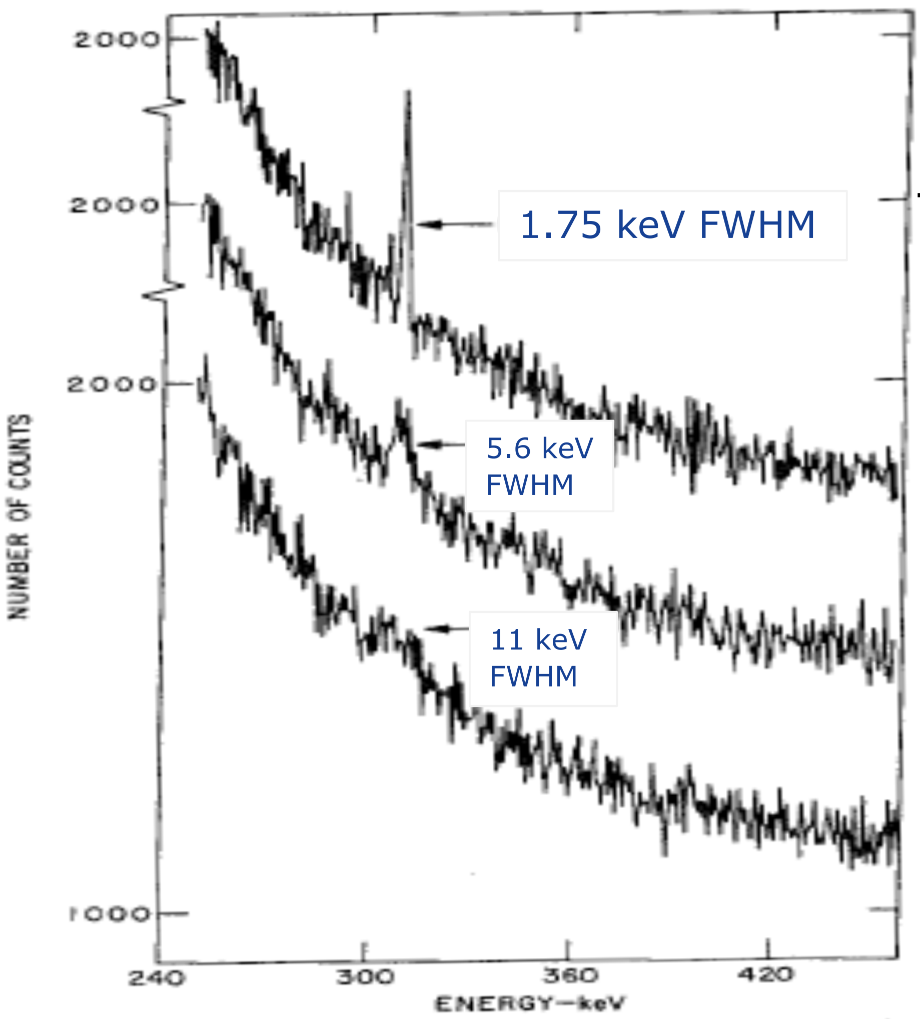
The High Purity Germanium detector (HPGe-detector)



$^{108m}Ag + ^{110m}Ag$

Extract from Knoll
"Radiation Detection and Measurements"

Ge-resolution: ~ 0.18%
NaI resolution: ~ 6%
@ 662 keV
~factor of 35 difference



The effect of detector resolution

Extract from Knoll
"Radiation Detection and Measurements"

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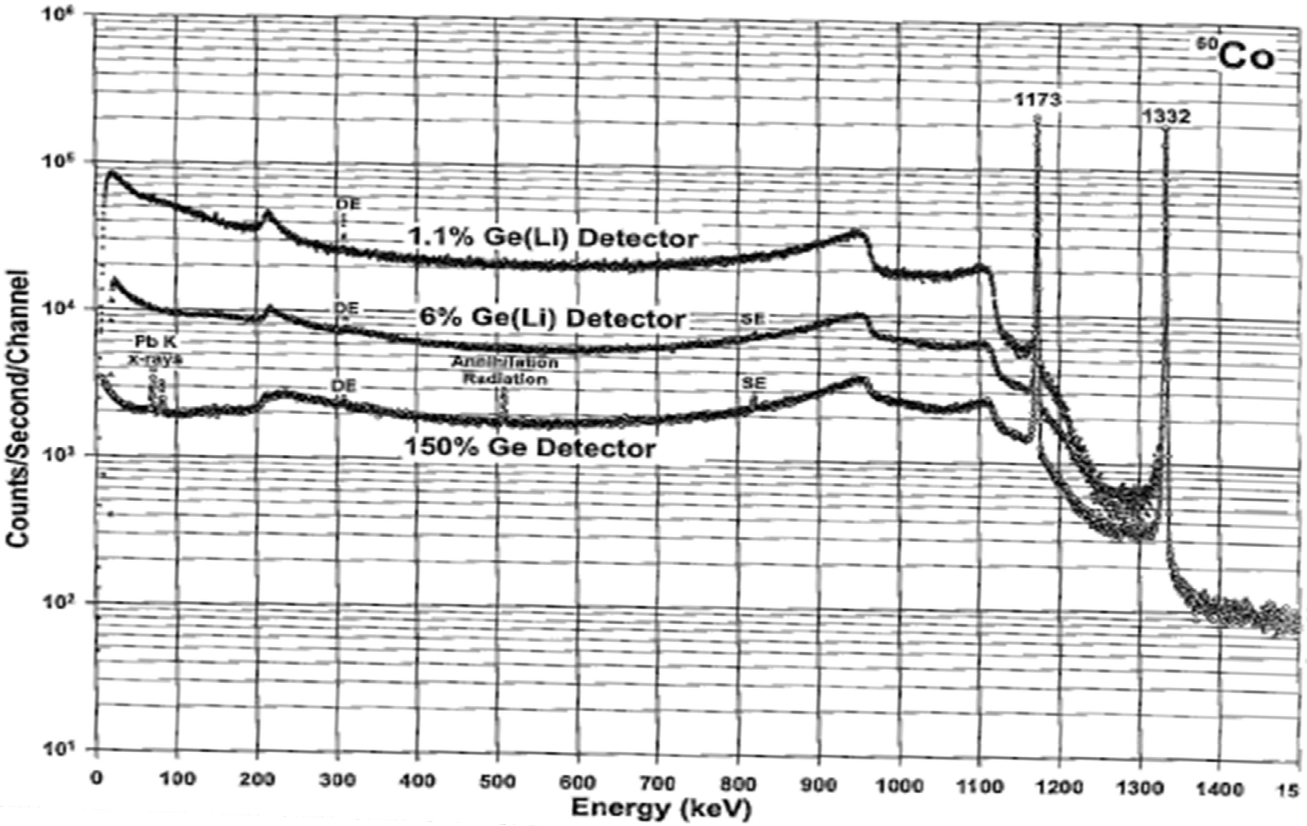
The effect of detector size



Extract from Knoll "Radiation Detection and Measurements"

Ge-detectors

- **The workhorse of modern radiometric laboratories**

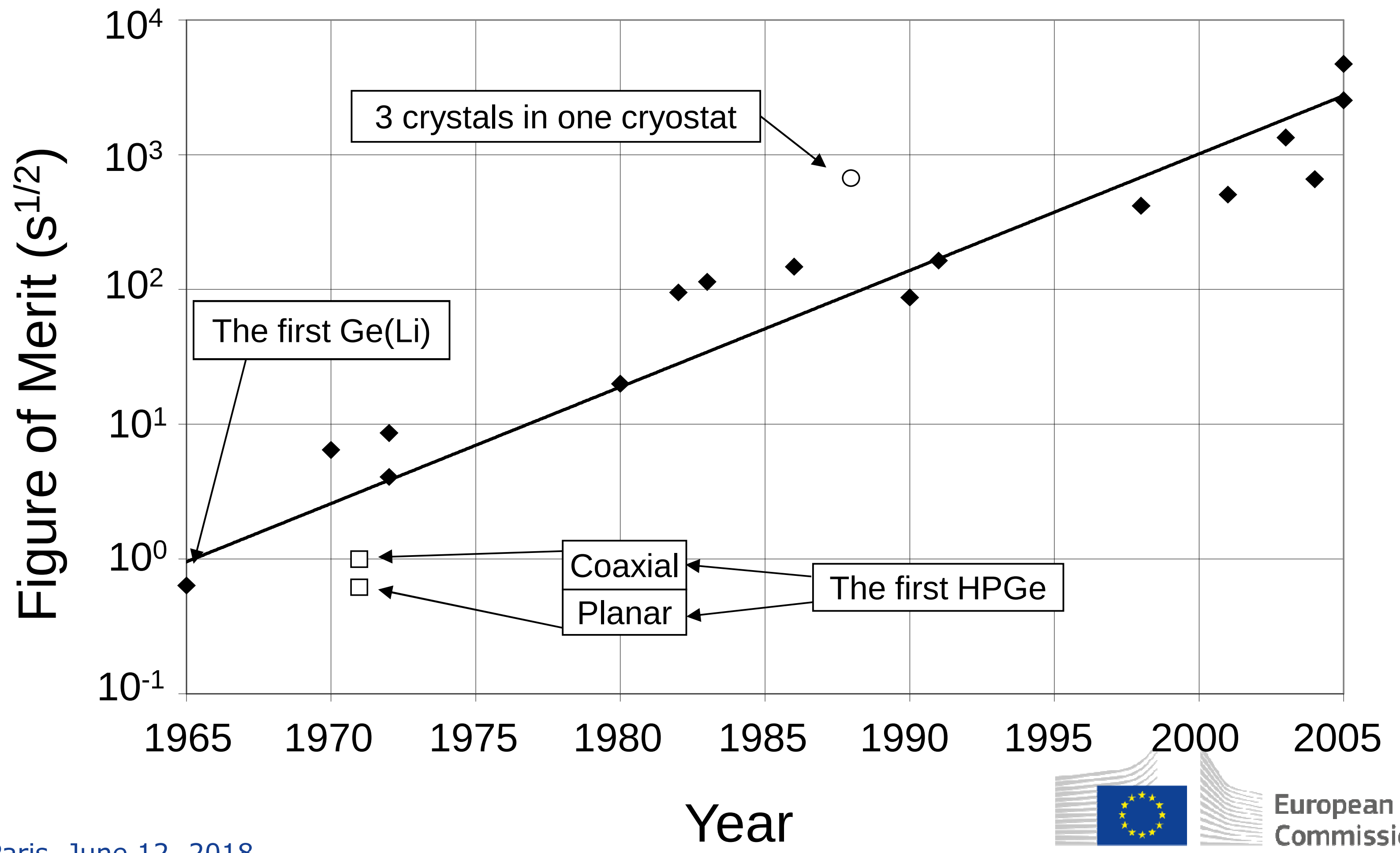
- **Li-drifting first described in 1960 (Pell)**

- **First Ge-detector in 1963 (Tavendale):**
  1 cm^3, same resolution as a NaI detector.

- **Improved detection limits if FoM is maximised (important for low-level measurements)**

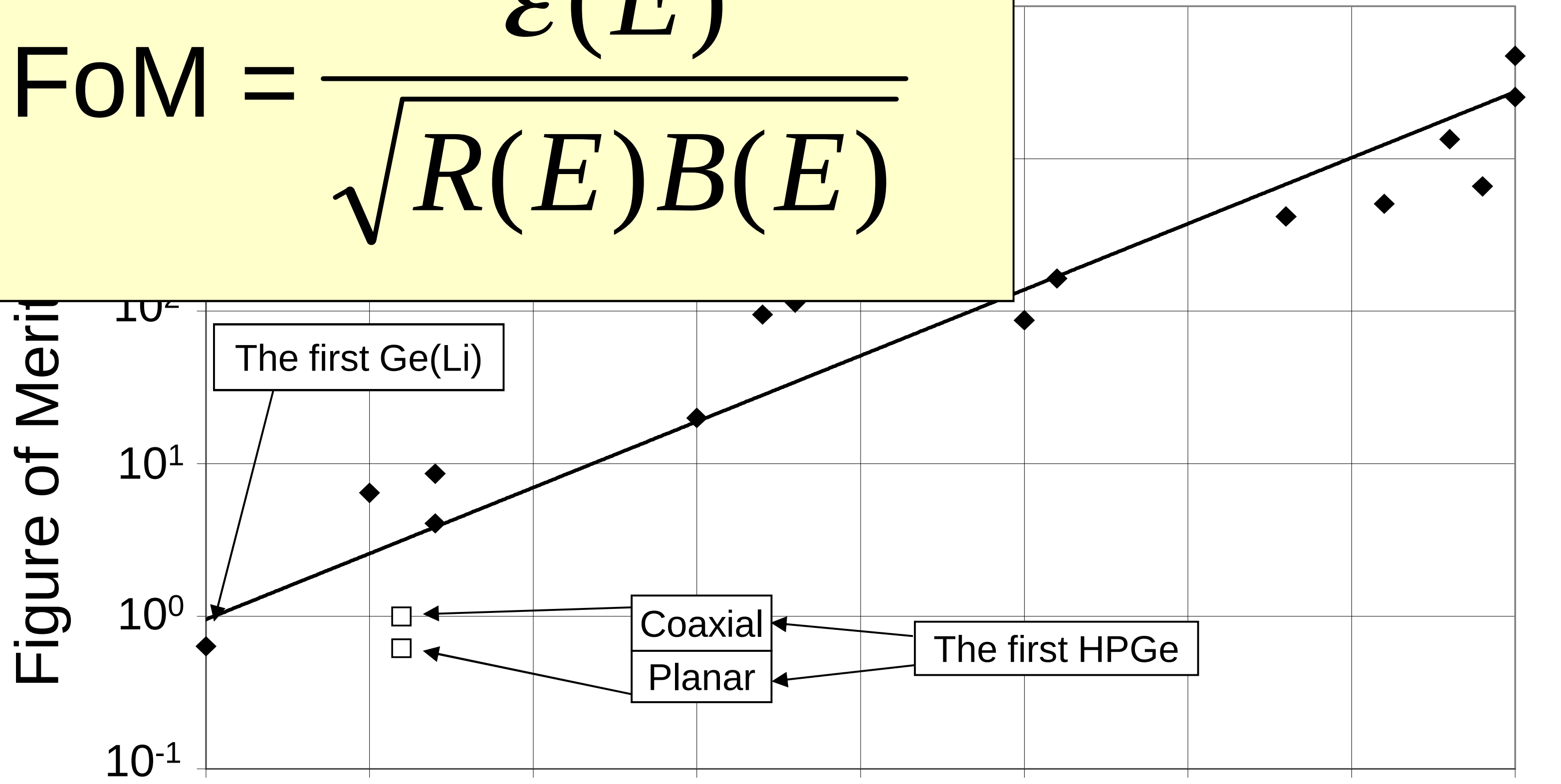
$$\mathrm{FoM} = \frac{\varepsilon(E)}{\sqrt{R(E)B(E)}} = \frac{\text{efficiency}}{(\text{FWHM} \times \text{background})^{1/2}}$$

Development in germanium detector technology



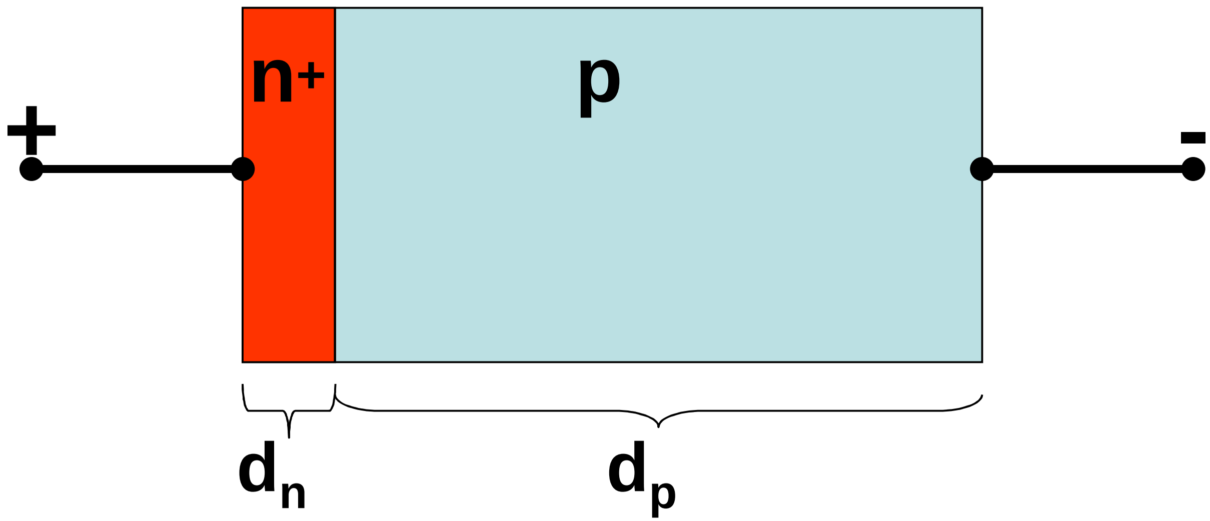
ector technology

$$\text{FoM} = \frac{\varepsilon(E)}{\sqrt{R(E)B(E)}}$$



M. Hult, Low-level gamma-ray spectrometry using Ge detectors, Metrologia 44 (2007) pp. S87-S94. And: Erratum, Metrologia 44 (2007) p. 425.

Basically a reversed biased diode



$$N_{electron} \cdot d_n + = N_{holes} \cdot d_p$$

$$d \sim sqrt(V)$$

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Li-contacts in HPGe

The n+ contacts are made by diffusing Li into the Ge for a short time (by heating and applying a voltage)

In germanium at room temperature Li diffuses about 0.1 mm in 1 year.

=> **A detector that was kept at room temperature for long has thicker deadlayers**

Today, manufacturers try to (succeed in) producing Li-free contacts.

Ge-production (i)

1) Raw material: residue from e.g. Zn-ore with 3-5% Ge or re-cycled electronics

2) Reduction of Ge-oxide

3) Zone-refinement => polycrystal

4) Czochralski growth => single crystal

Resistivity measurement

repeat

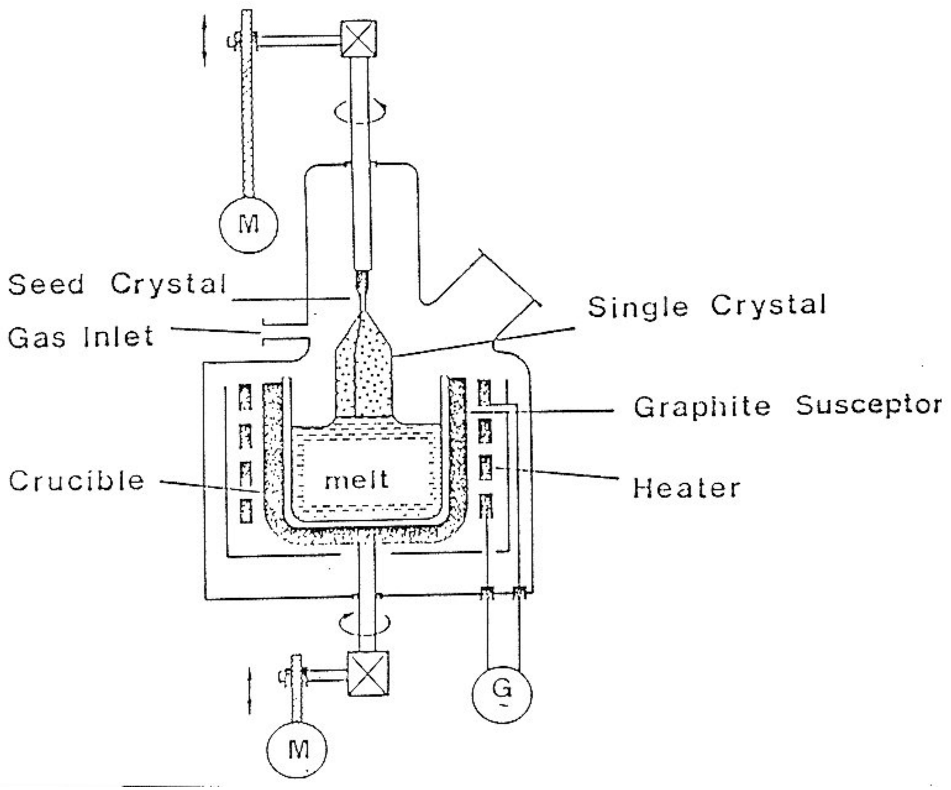
Measurements

- Resistivity
- Hall
- DLTS

Ge-production (ii)
Czochralski crucible pulling…

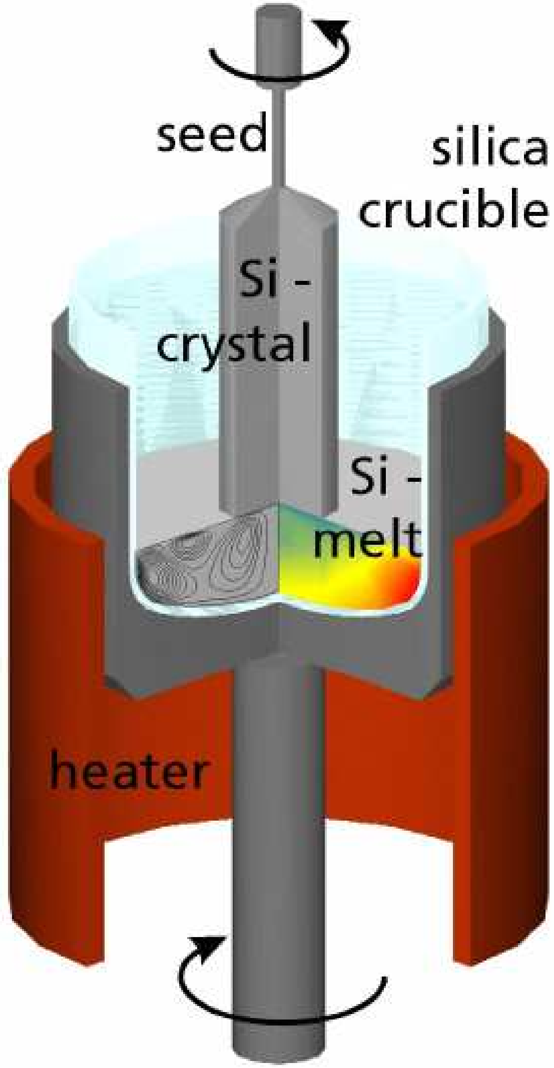
- **2-3 days**
- **Small "low power", some gas**
- **Needs clean room for large HPGe-detectors**
- **Many secret recipes**
- **Nowadays 4" for HPGe and 6" for other applications**

czochraliski cricible pulling



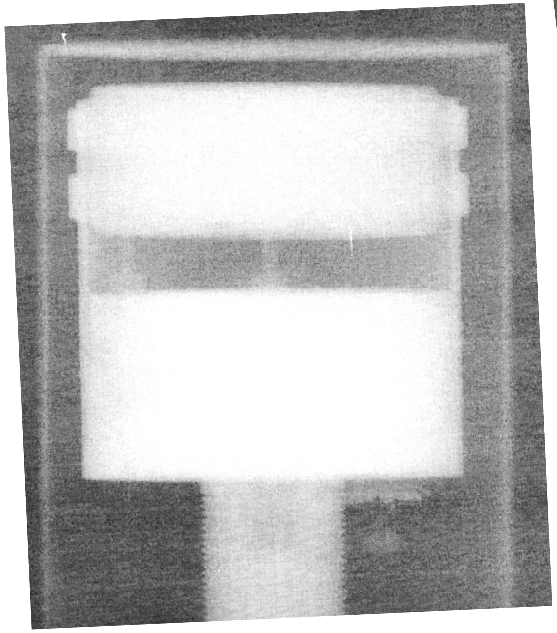
Ge-production (iii)

Czochralski crucible pulling…

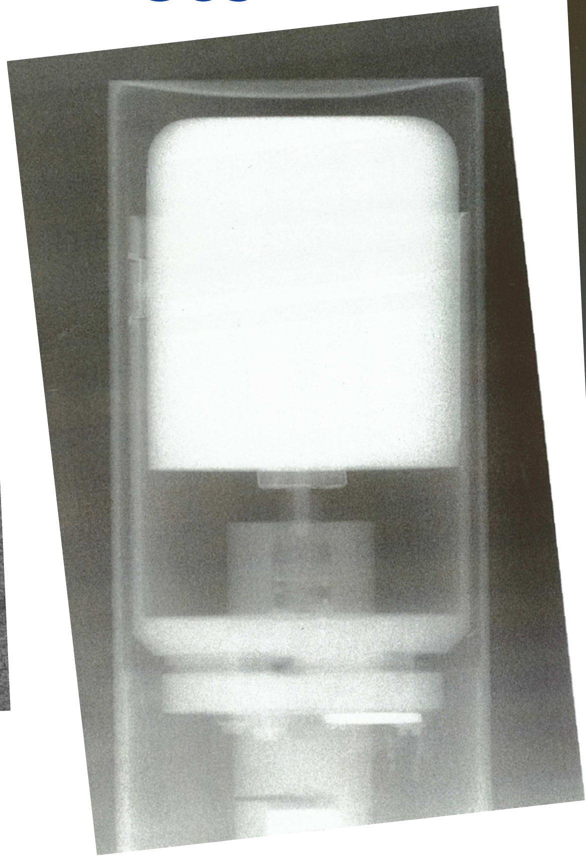


Radiography

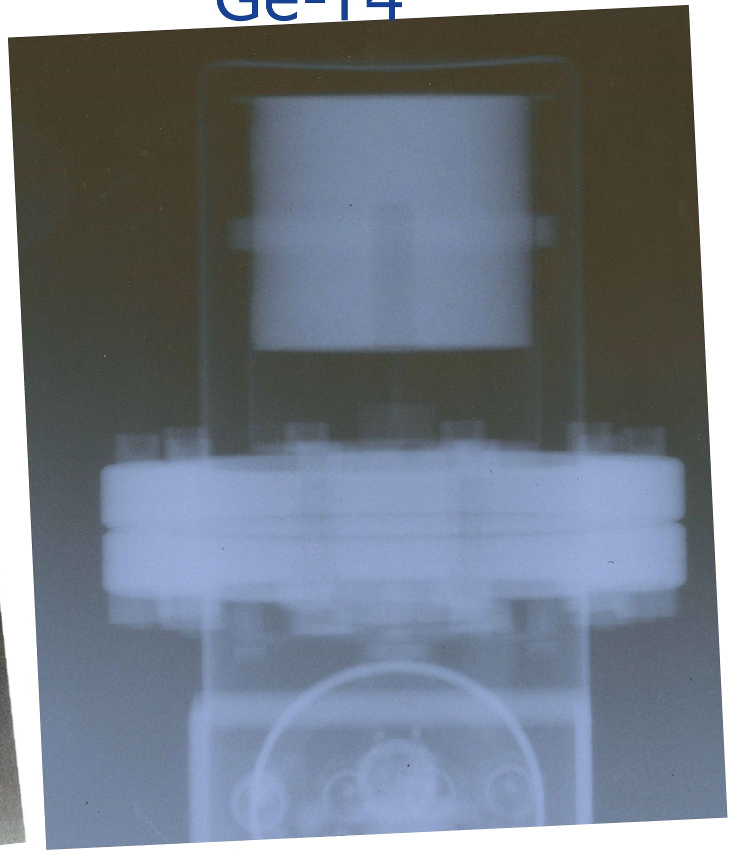
Ge8



Ge3



Ge-T4

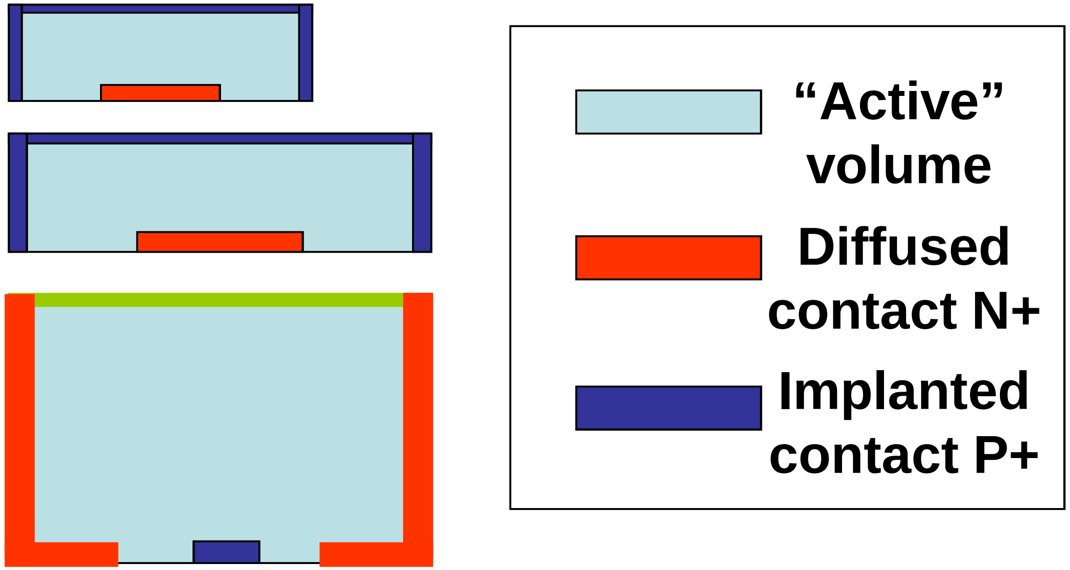


Planar detector
(Canberra notation – similar detector available from others)

Ultra LEGe
n-type

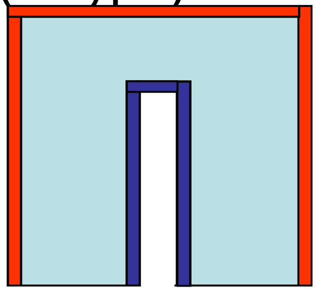
LEGe – Low Energy Ge
n-type

BEGe – Broad Energy Ge
p-type (special process)

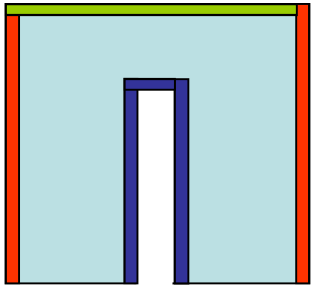


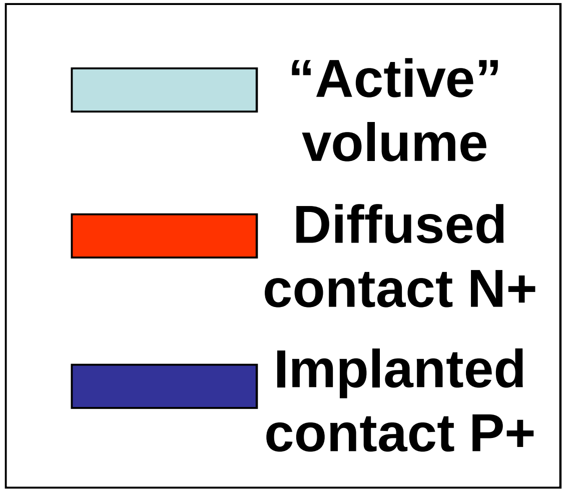
Coaxial detector
(Canberra notation – similar detector available from others)

Coaxial
(P-type):

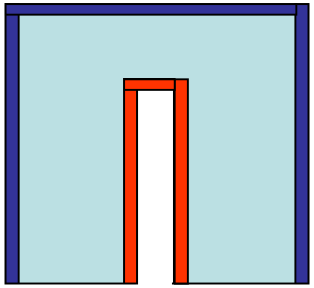


XtRa
(Extended range)
(p-type):

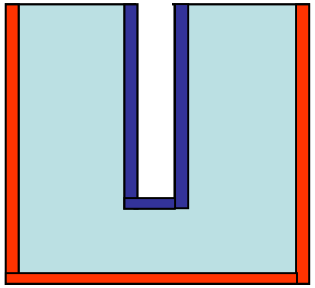




REGe
Reversed Electrode Ge
(n-type):



Well-detector:



Ge-production (iii)
Crystal treatment

(i) Mechanical “shaping”

(ii) Grinding

(iii) Contact structures (etching, diffusion, implantation)

Bulletization

Weak electric field in corners => long rise time => not completely collected within reasonable integration time => rounding of edges =bulletization

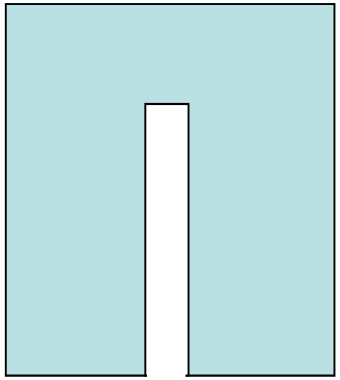
Important to include in computer model!!!

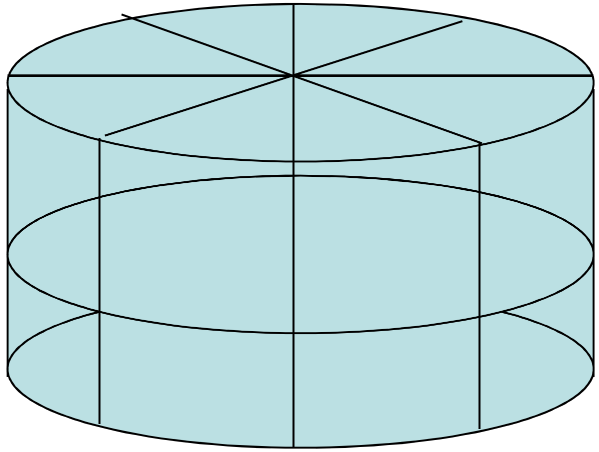
New crystals with sharp edges may have poor charge collection in corners => difficult with Monte Carlo simulations

Different crystal configurations

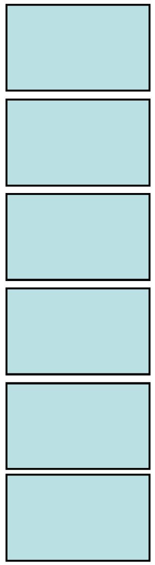
Multi-segmented

Coaxial

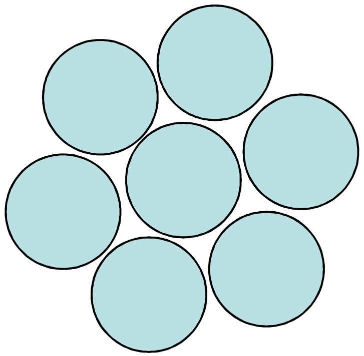




Planar



Array detector



1. Primordial (Here since the formation of the earth)

2. Anthropogenic (man-made)

3. Cosmogenic (Induced by cosmic rays)

1. Primordial radionuclides

(natural, existing since the formation of the earth)

Earth is about $4.5 \cdot 10^9$ years

^{238}U, $T_{1/2}$ = 4.5×10^9 years
^{235}U, $T_{1/2}$ = 0.7×10^9 years
^{232}Th, $T_{1/2}$ = 14×10^9 years
^{40}K, $T_{1/2}$ = 1.3×10^9 years

1 Bq $^{238}U \Rightarrow$ 14 Bq
in the whole decay chain

Decays to
radium-226, radon-222,
polonium-210, lead-210 etc.

Less common ones
La-138, Rb-87, Sm-147, Lu-176, Re-187

More in my next presentation

2. Anthropogenic (man-made)

Fission products: ^{137}Cs, ^{134}Cs, ^{85}Kr, ^{125}Sb, ^{131}I, ^{129}I, …..

Activation products: ^{60}Co, ^{41}Ar, ….

Normally no problem for background, but after Chernobyl many (also new) detectors were contaminated

Note: always difficult with pure beta emitters; ^{90}Sr, ^{3}H, ^{115}In - bremsstrahlung

3. Cosmogenic (some examples… also spallation reactions)

#	target	reaction	Produced radionuclide	γ-ray energy	$T_{1/2}$	Side reaction
9	^{59}Co	(n,p)	^{59}Fe	1099; 1291	44.53 d	(μ^-,0n)
10	^{60}Ni	(n,p)	^{60}Co	1173.2; 1332.5	5.271 y	
13	^{63}Cu	(n,4p6n)	^{54}Mn	834.84; 840.8[c]	312.3 d	(μ^-,3p5n)
14	^{63}Cu	(n,2p5n)	^{57}Co	122.1; 136.5; 143.6[c]	271.79 d	(μ^-,p4n)
15	^{63}Cu	(n,2p4n)	^{58}Co	810.8; 817.9[c]	70.86 d	(μ^-,p3n)
16	^{63}Cu	(n,α)	^{60}Co	1173.2; 1332.5	5.271 y	
17	^{65}Cu	(n,γ)	^{66*}Cu	186.0		
18	^{65}Cu	(n,n’)	^{65*}Cu	1115.5; 1481.7		
19	^{70}Ge	(n,γ)	^{71m}Ge	23.5;174.9; 198.3	22 ms	72Ge(n,2n)
20	^{70}Ge	(n,γ)	^{71}Ge	10.37	11.34 d	
21	^{70}Ge	(n,3n)	^{68}Ge	10.37	271 d	
22	^{70}Ge	(n,2p4n)	^{65}Zn	1125.2	244.3 d	(μ^-,p4n)
23	^{72}Ge	(n,γ)	^{73m}Ge	13.3; 53.4; 66.7	0.5 s	74Ge(n,2n)
24	^{72}Ge	(n,n’)	^{72*}Ge	691.0[b]; 834[b]		

Many reactions in Cu ! (and Ge)

In Ge: ^{68}Ge, ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn, ^{54}Mn, ^{63}Ni, ^{55}Fe

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Main background sources – a practical classification scheme

⇒ Radon

⇒ Laboratory environment except radon (i.e. radioactivity and neutrons from fission and (α,n) reactions in surrounding materials)

⇒ Directly induced by cosmic rays

⇒ Indirectly induced by cosmic rays (Activation of Ge-crystal, cryostat and shield)

⇒ Radioimpurities in detector and shield

Important to identify location of background sources

$\Rightarrow$ Enables improvement (=reduction) of background

$\Rightarrow$ Enables improved design for future detector systems

Ways to quantify importance of background sources

1) **Radon** – monitor Rn-concentration and the background simultaneously. Extrapolate to zero Rn concentration to see count rate from other sources

2) **Radioimpurities in shield and detector** – Cover endcap with a very pure material (Hg) and compare the difference in count rate

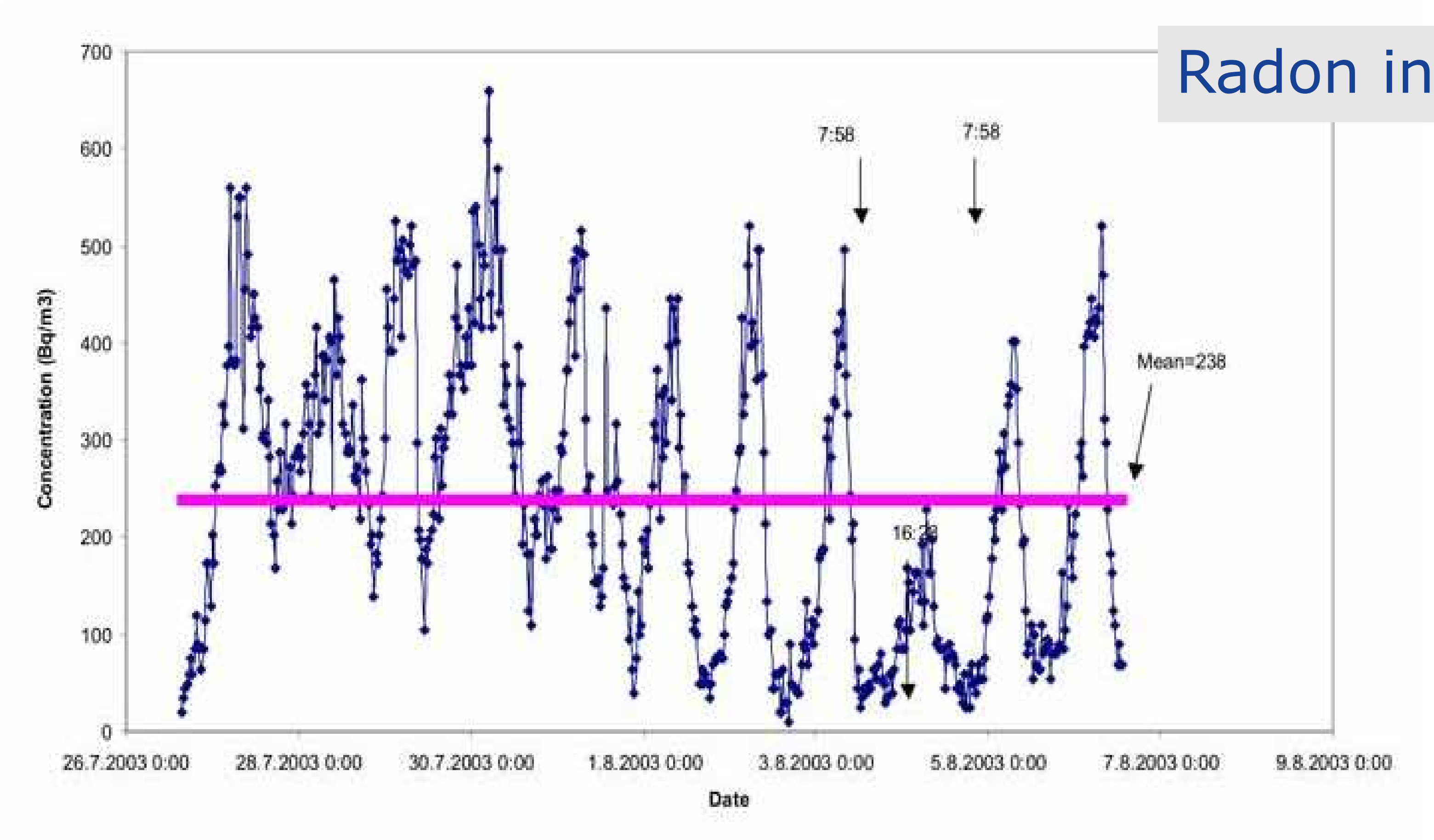
3) **Environment** – Vary the shield thickness (both lead and borated paraffin)- very cumbersome!!

4) **Activation** – Note count rate of activation peaks from crystal (e.g. Zn-65 at 1115+8.3 keV) and make simulations to find total count rate.

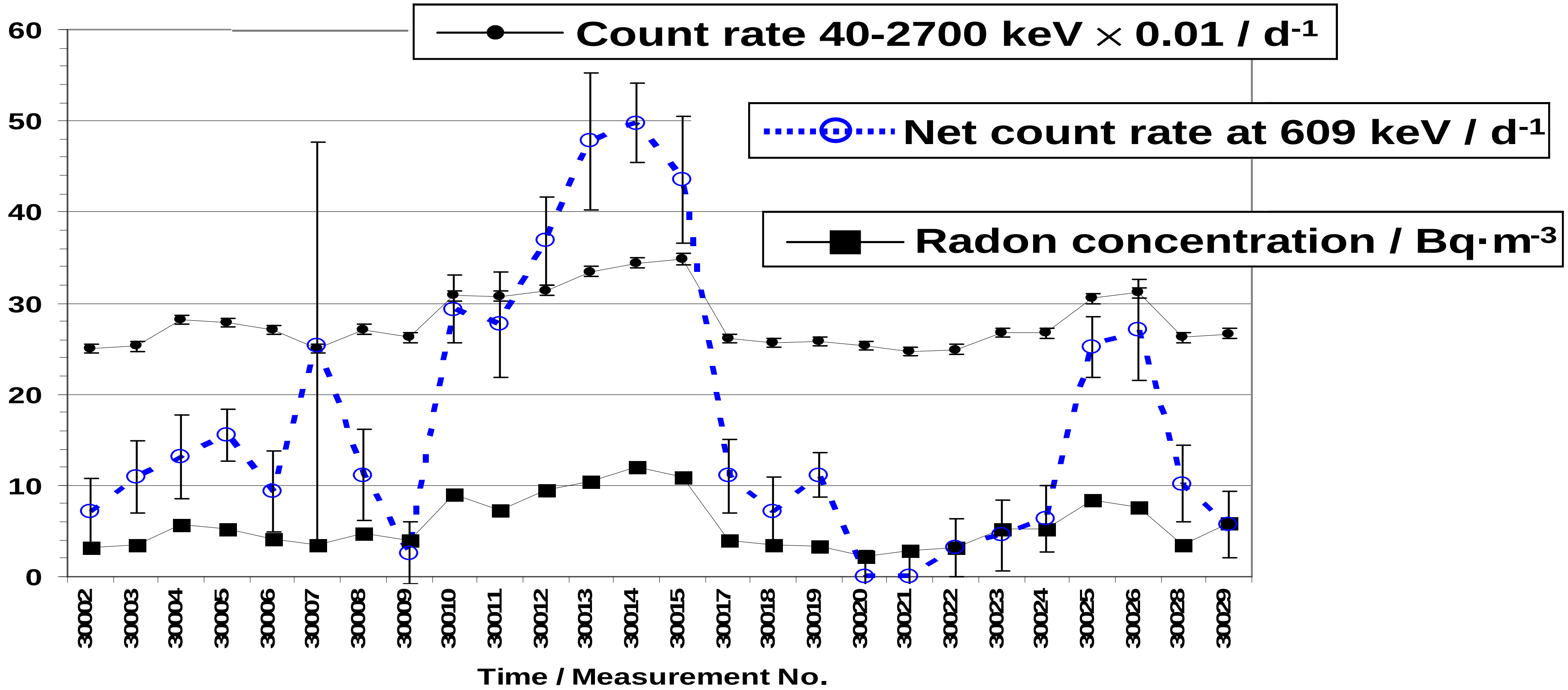
5) **Muons** - Correlate the background changes with changes in cosmic ray flux (see e.g. the Kiel neutron monitor on internet) or use a muon detector *(Extrapolate to zero or have your detector system tested underground)*

……..

Radon in air



⇒ **Due to variation with time, it is necessary to use the standard-deviation of the background in the uncertainty budget**



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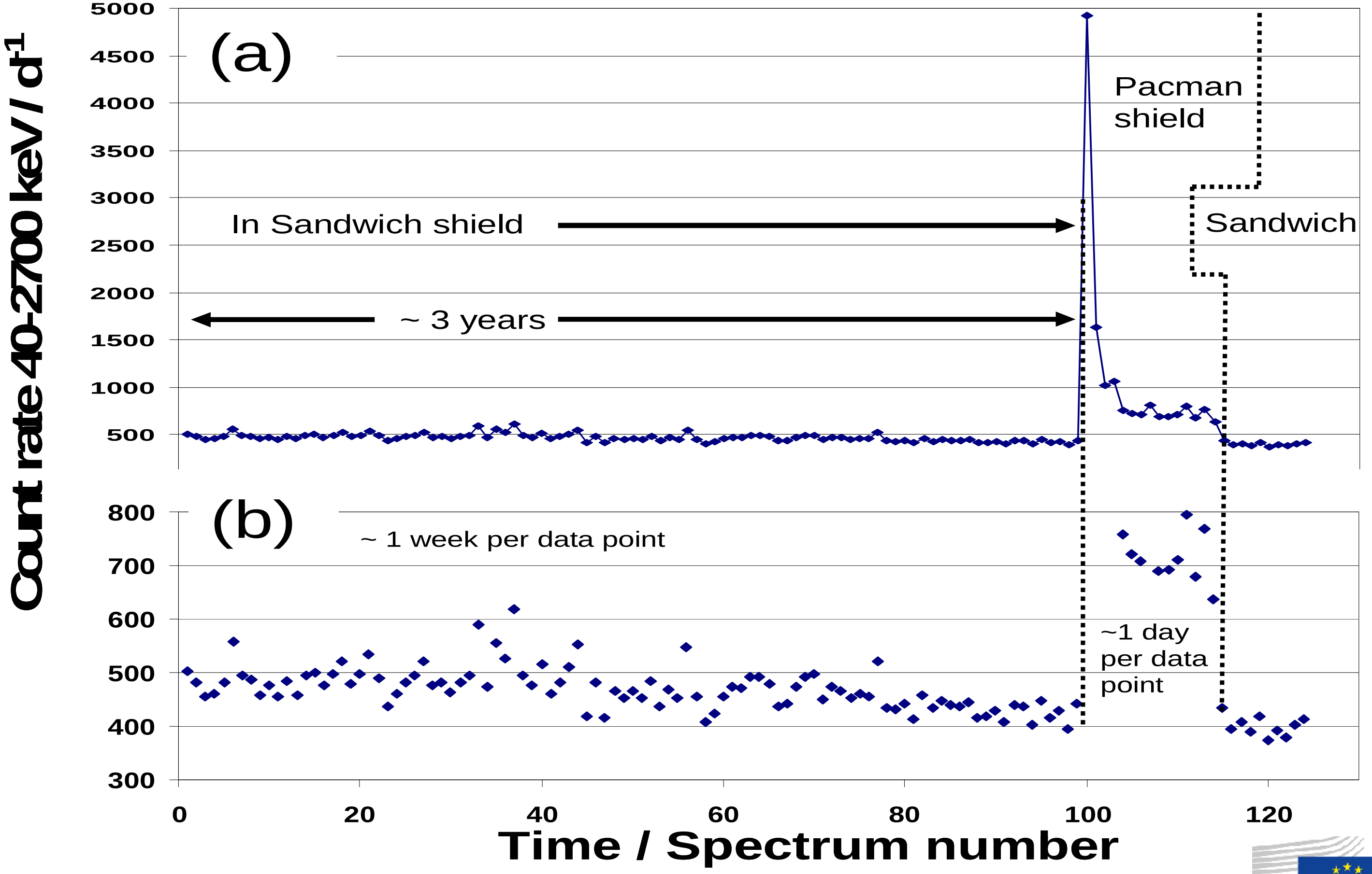
Detector Ge-7 – XtRa (p-type w thin deadlayer), 90% rel eff. Inverted cryostat



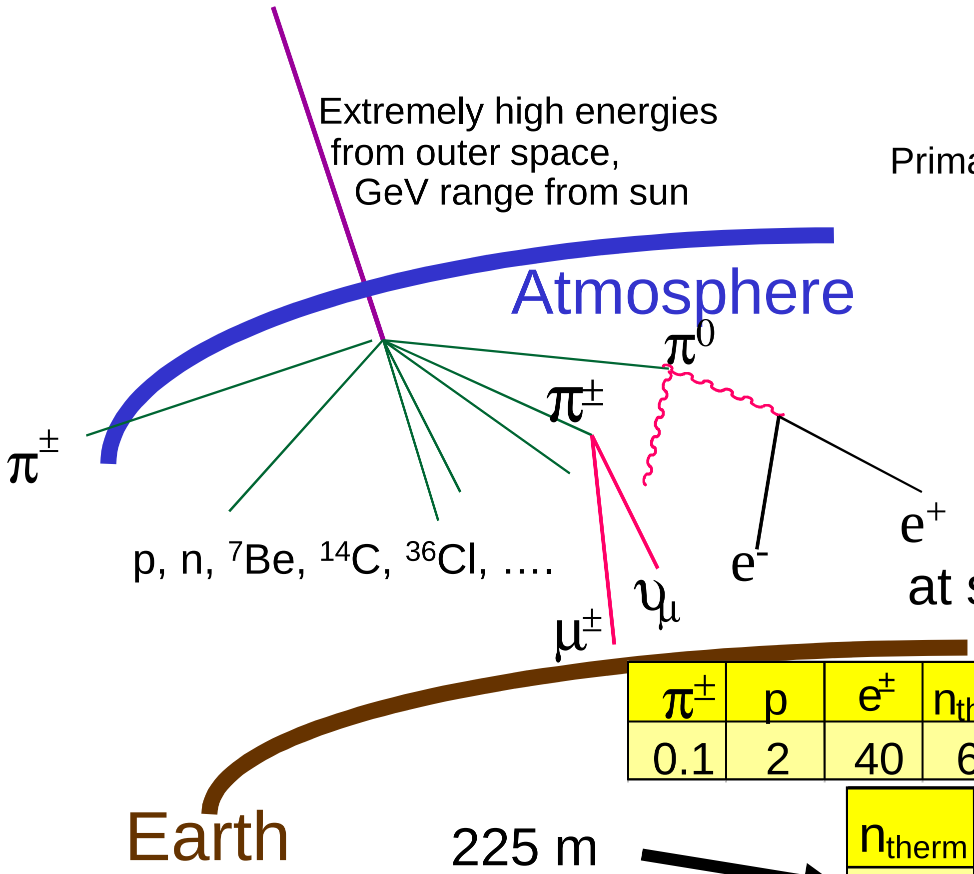
Other ways to understand background

- Keep a log of the background – both background peaks and certain intervals
- Measure background in different shields – a good way to understand if a shield is bad or the detector is bad. (if background is good, then both must be good)

  Quite cumbersome and time consuming. Moving lead and detectors, re-doing calibrations,….

…..Unless there are special lead shields that allow easy removal of detectors ………

$\Rightarrow$ Possibility for generating interesting articles



Primary cosmic ray: $10^3\ m^{-2}\ s^{-1}$

9 % α

90 % p

1 % heavier nuclei (up to Fe)

at surface ($m^{-2}s^{-1}$)

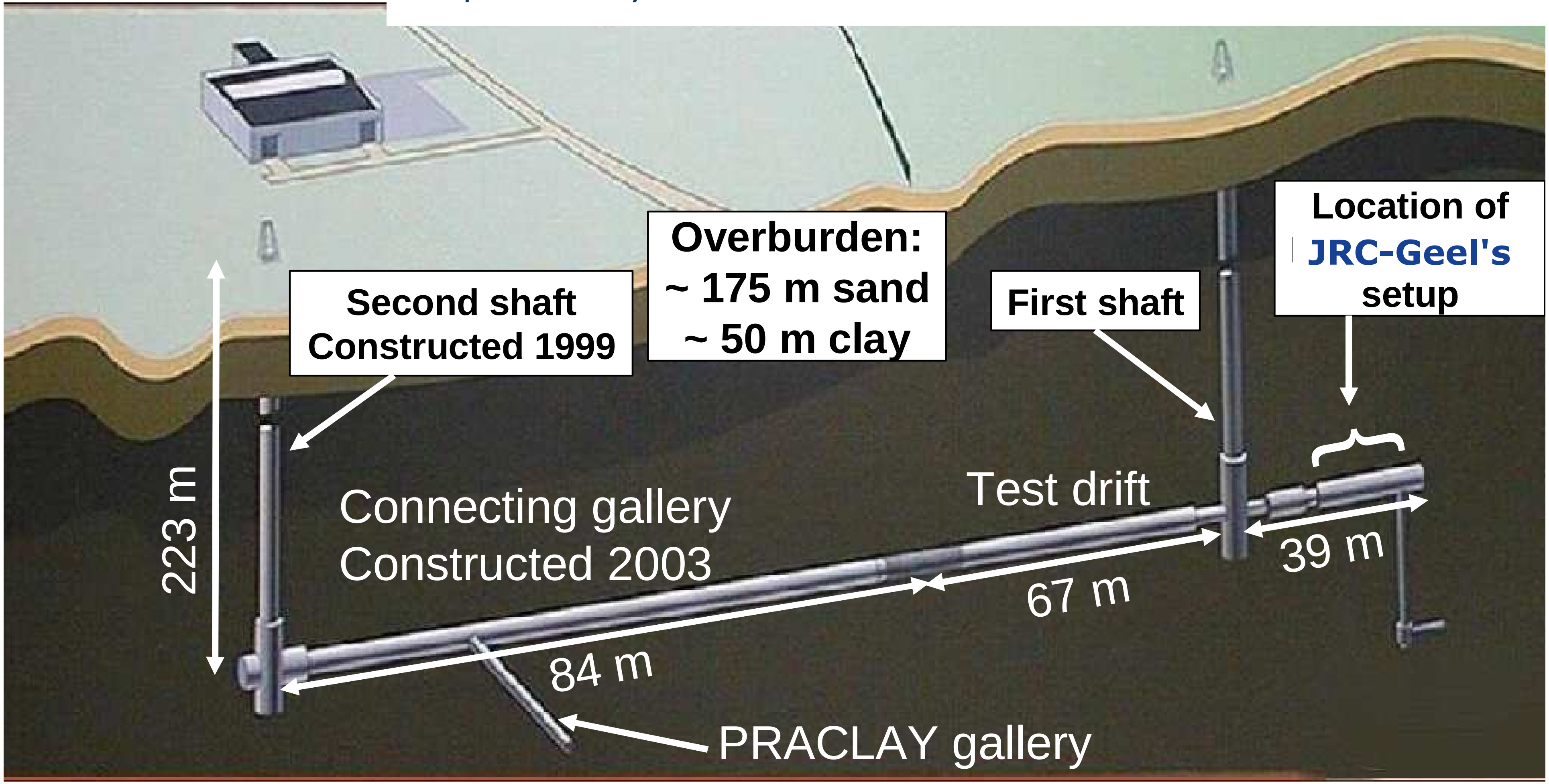
$\pi^{\pm}$	p	$e^{\pm}$	n_{therm}	n_{fast}	$\mu^{\pm}$	υ
0.1	2	40	60	200	200	10^{11}

225 m underground ($m^{-2}s^{-1}$)

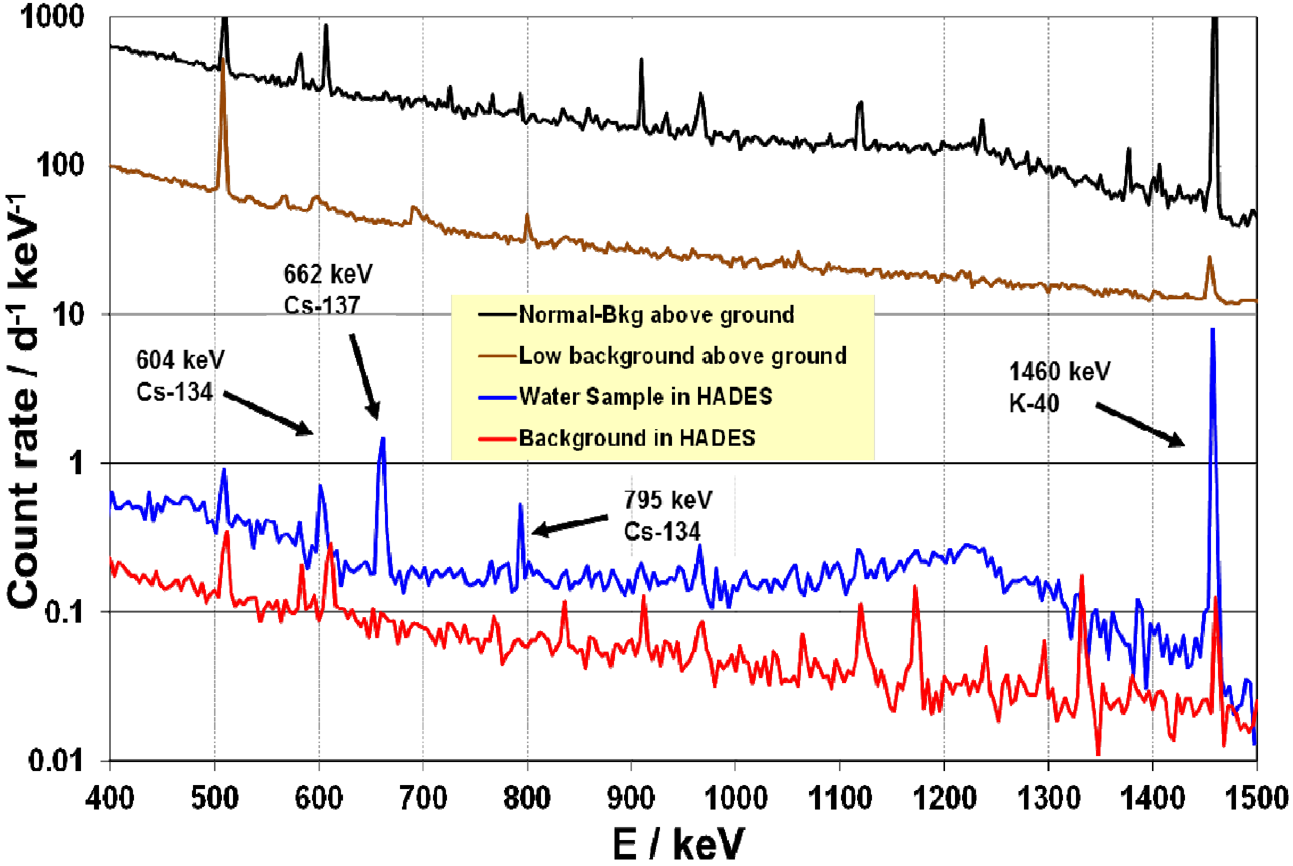
n_{therm}	n_{fast}	$\mu^{\pm}$	υ
<2	<2	0.1	10^{11}

HADES

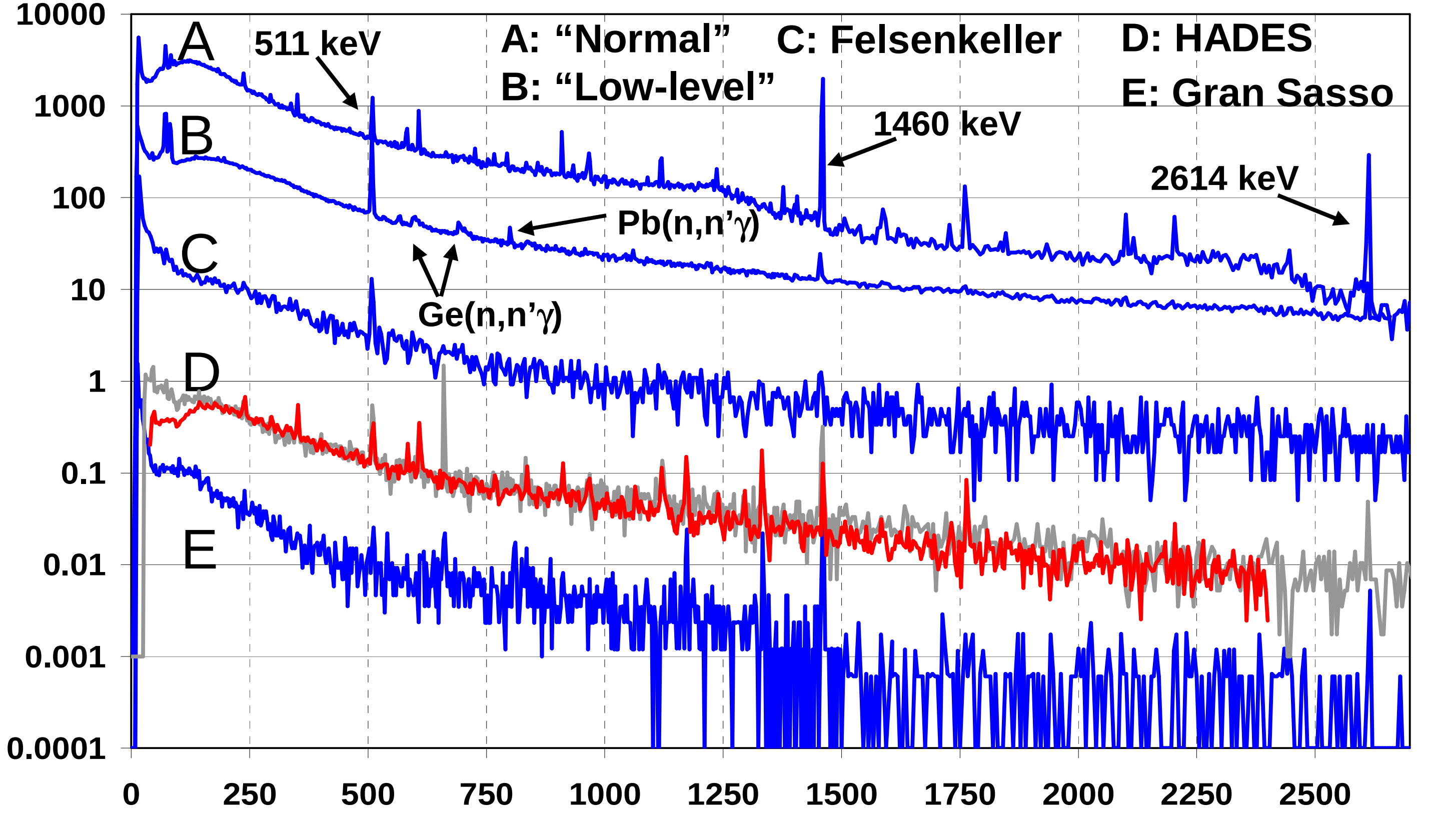
HADES = High Activity Disposal Experimental Site
– Operated by EURIDICE and located at SCK•CEN in Mol



HADES - The flux of muons is reduced a factor ~10^4



Background Comparison – Gamma-ray spectrometry



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Low background

Low level Gamma-ray Spectrometry (LGS) = Gamma-ray spectrometry using a detector and shield built from selected radiopure materials and a shield of

Ultra Low level Gamma-ray Spectrometry (ULGS) = LGS with additional measures such as placement in an underground laboratory or use of a muon shield.

Low background

Advice:

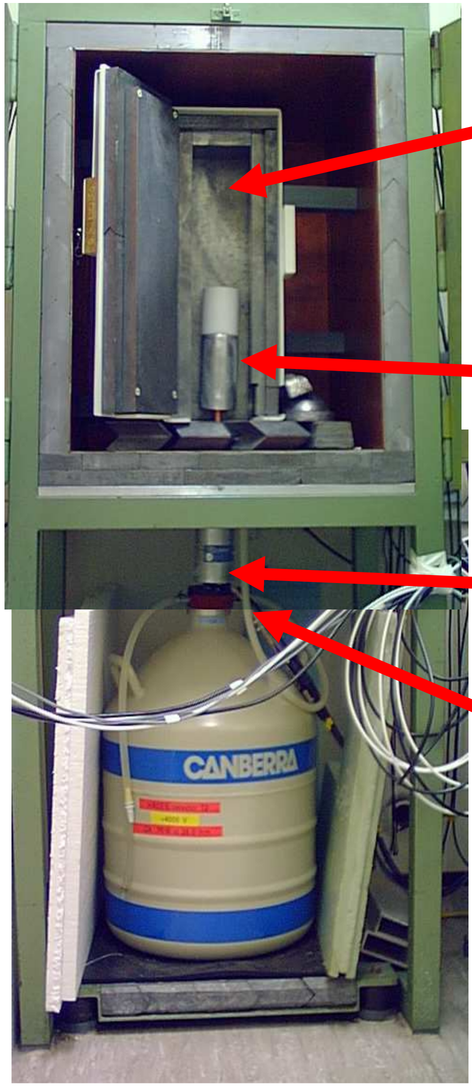
A) Don’t buy a Low background detector unless you really need to!

(i) more fragile (ii) more expensive

B) Don’t place a detector for underground use, above ground

activation of crucial parts

Low Background Detector



Inner 2.5 cm from ULB lead (2 Bq/kg)

Endcap, cryostat and front-end-electronics from selected radiopure materials

Pre-amp outside lead shield

Tube for nitrogen “flushing”

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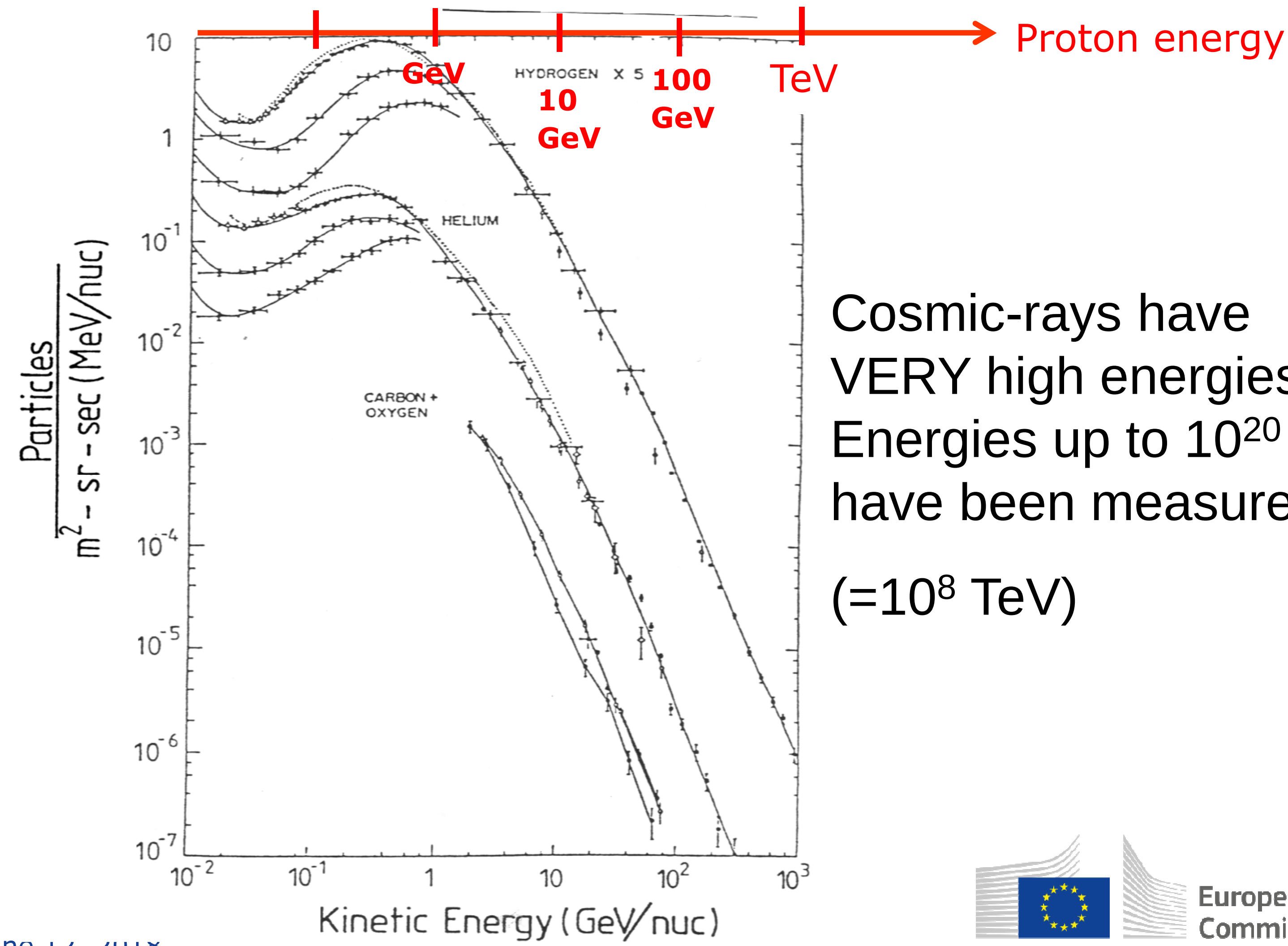
Extra benefits underground

- **Possibility to use thick lead shield (no increase of background after 15 cm)**
- **Possibility to use thick Cu shield as lining => not necessary with the best (most expensive) lead**
- **No activation of Ge**

Depth (m w.e.)	Idiom	Characteristics
< 10	Not underground or above ground	• The soft component (e, e+, photons) is strongly reduced but still plays a part. • Very little reduction of muon flux and neutron induced by muons. • Muon shields are useful.
10 – 100	Shallow underground	• The soft component of the cosmic ray has vanished. • The muon flux is reduced a factor of 5-50, but Muon shields are useful. • There is still a significant flux of neutrons produced by muons (reduction of factor 2-10). • The activation of crystal and shield are still important factors.
100 – 1000	Semi deep underground	• Cosmogenic activation can be neglected. • A slight improvement can be obtained by discriminating against muons. • The neutron flux is dominated by (α,n) sources
> 1000	Deep underground	• The influence of the cosmic rays can be neglected. • The only source for neutrons are (α,n) reactions.

Source: Hult et al. Acta Chimica Slovenica, 2006.

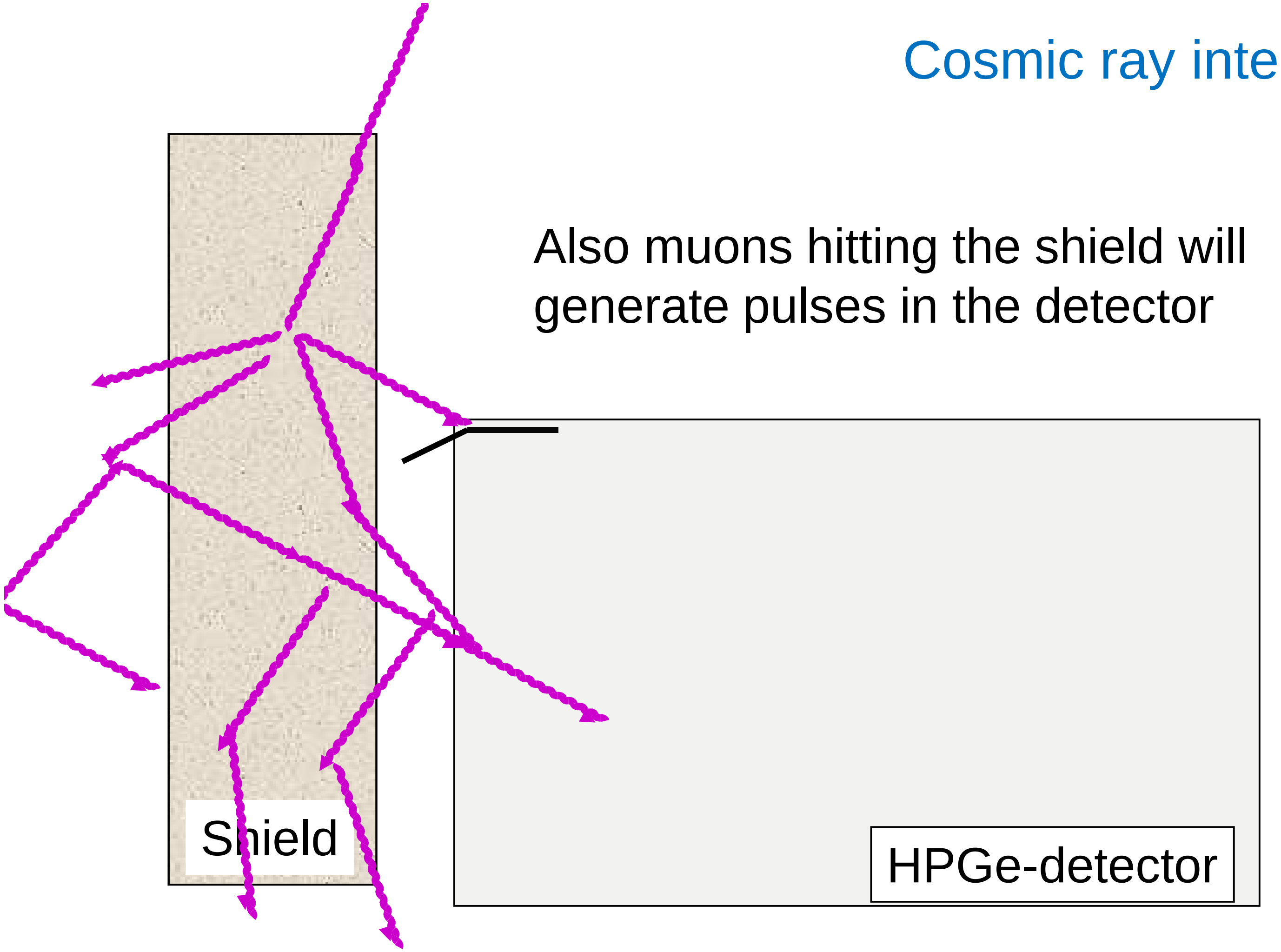


Cosmic-rays have VERY high energies. Energies up to 10^{20} eV have been measured

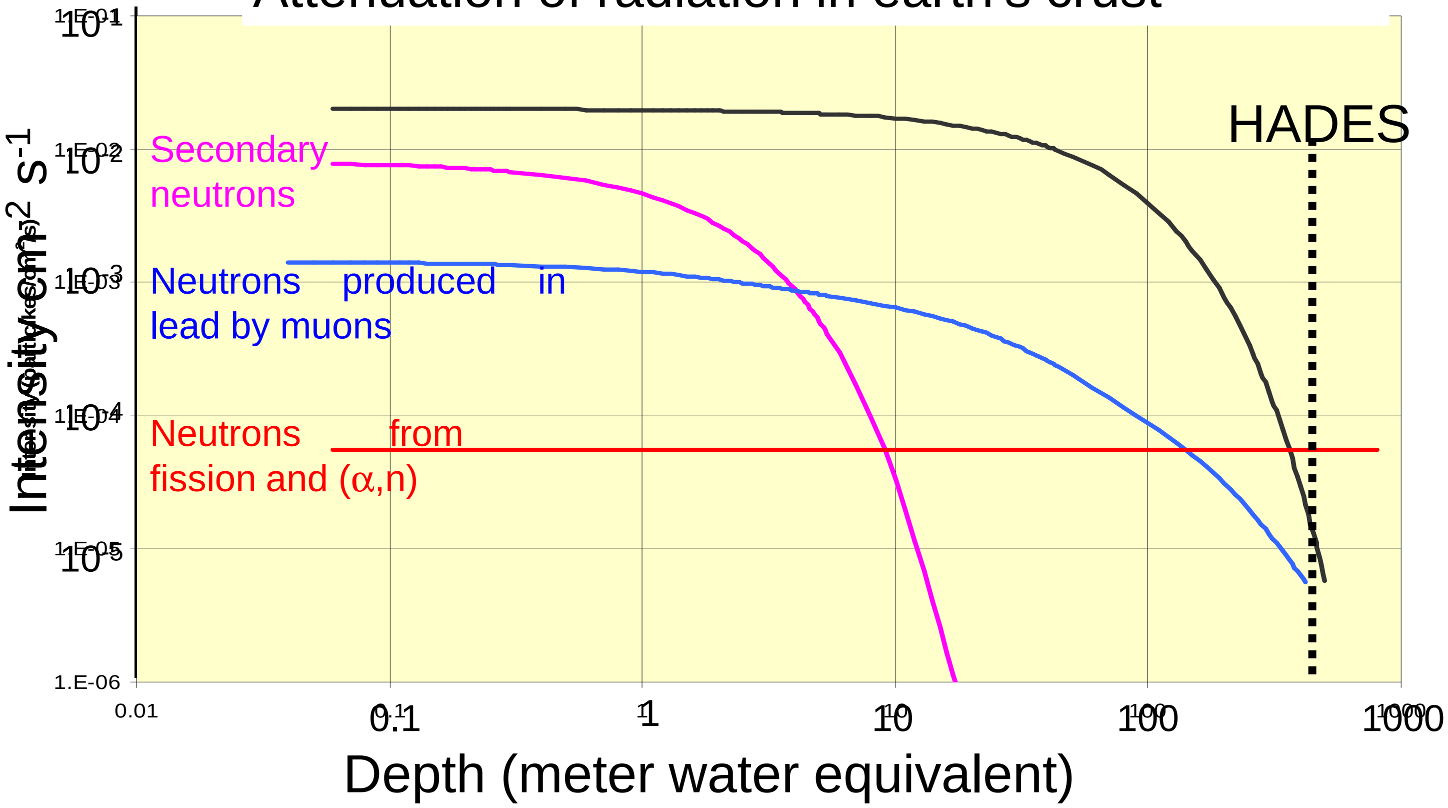
($=10^8$ TeV)

Cosmic ray interactions

Also muons hitting the shield will generate pulses in the detector



Attenuation of radiation in earth's crust



<u>No</u> air transport of a low background detector!!

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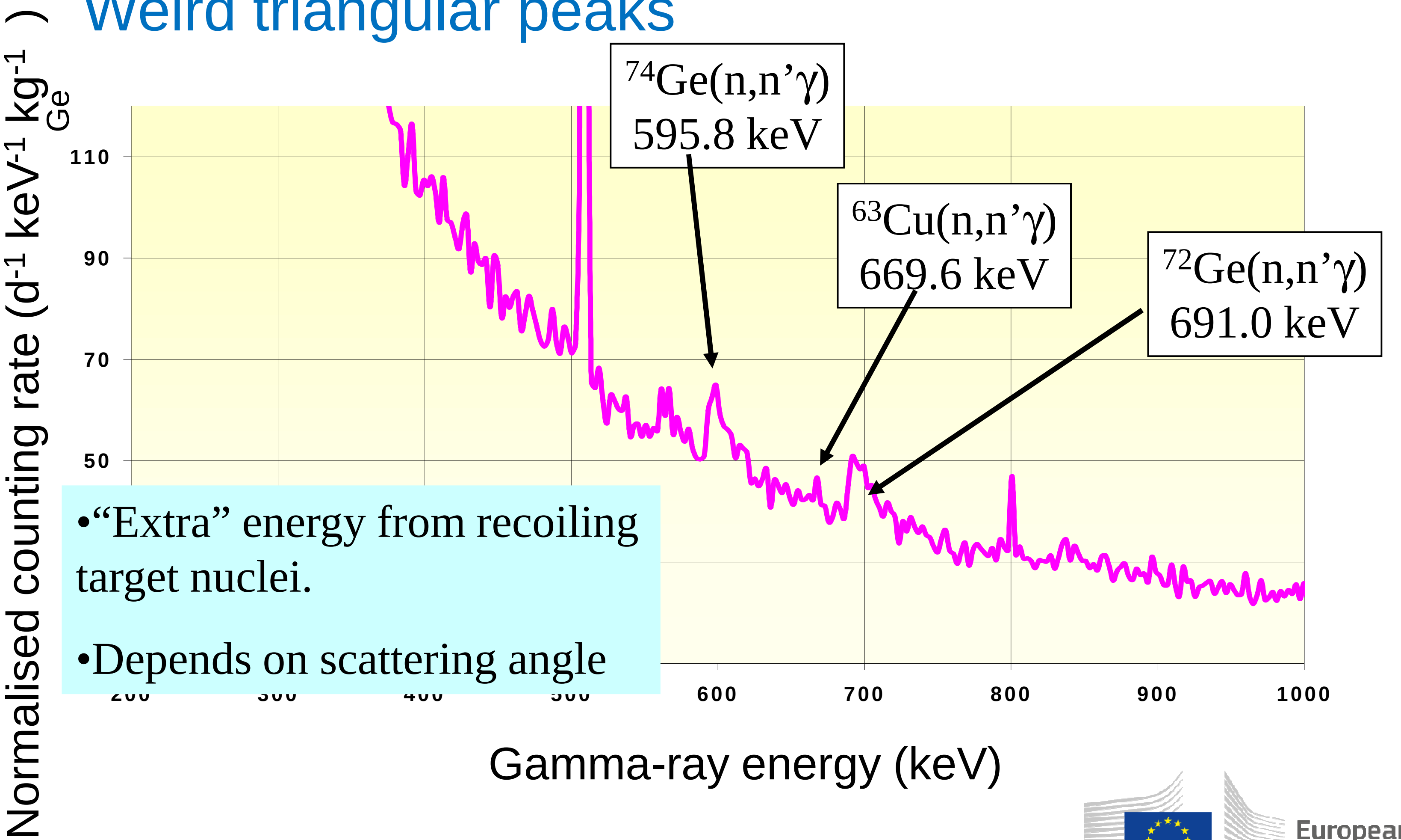
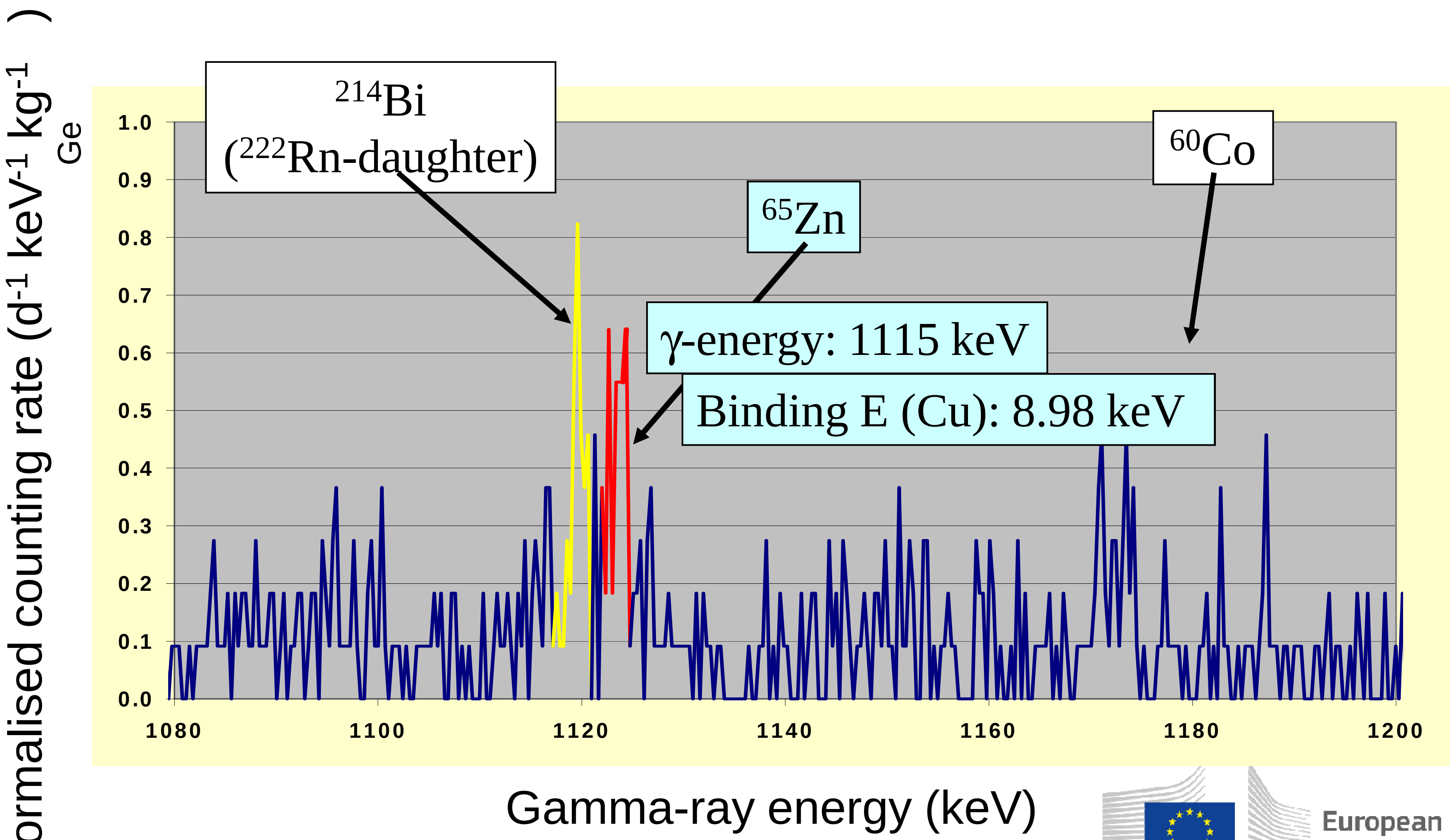
Weird triangular peaks



- "Extra" energy from recoiling target nuclei.
- Depends on scattering angle



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Other tricky peaks

^{57}Co (E.C.)

122.1+14.4 keV (γ-rays) + 7.1 keV (bind.E) = 143.6 keV

^{58}Co (E.C.)

811 keV (γ-ray) + 7 keV (bind.E) = 818 keV

1460.8 keV

From ^{40}K, BUT also a peak at 1459.2 keV from ^{228}Ac (P_γ = 1.06% or 0.83%) => take care to quantify ^{40}K if there is a relatively high amount of ^{232}Th/^{228}Ra in the sample.

661.7 keV

^{137}Cs BUT also 1173-511 keV.

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Other tricky peaks:

^{222}Rn-daughters

Background subtraction tricky if the Rn-concentration changes with time. Advice: Flush with N_2 and wait some time to start measurement.

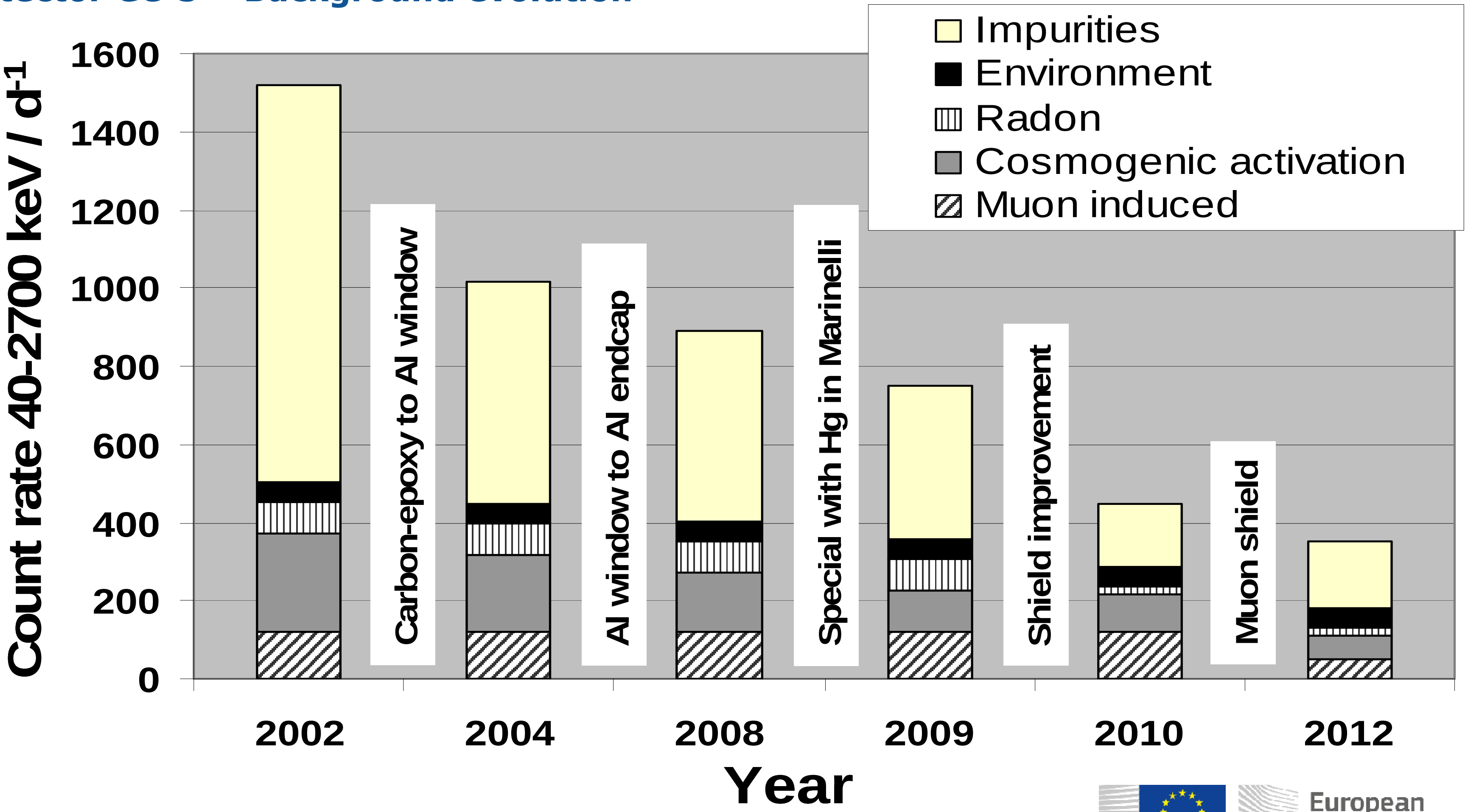
1293.6 keV

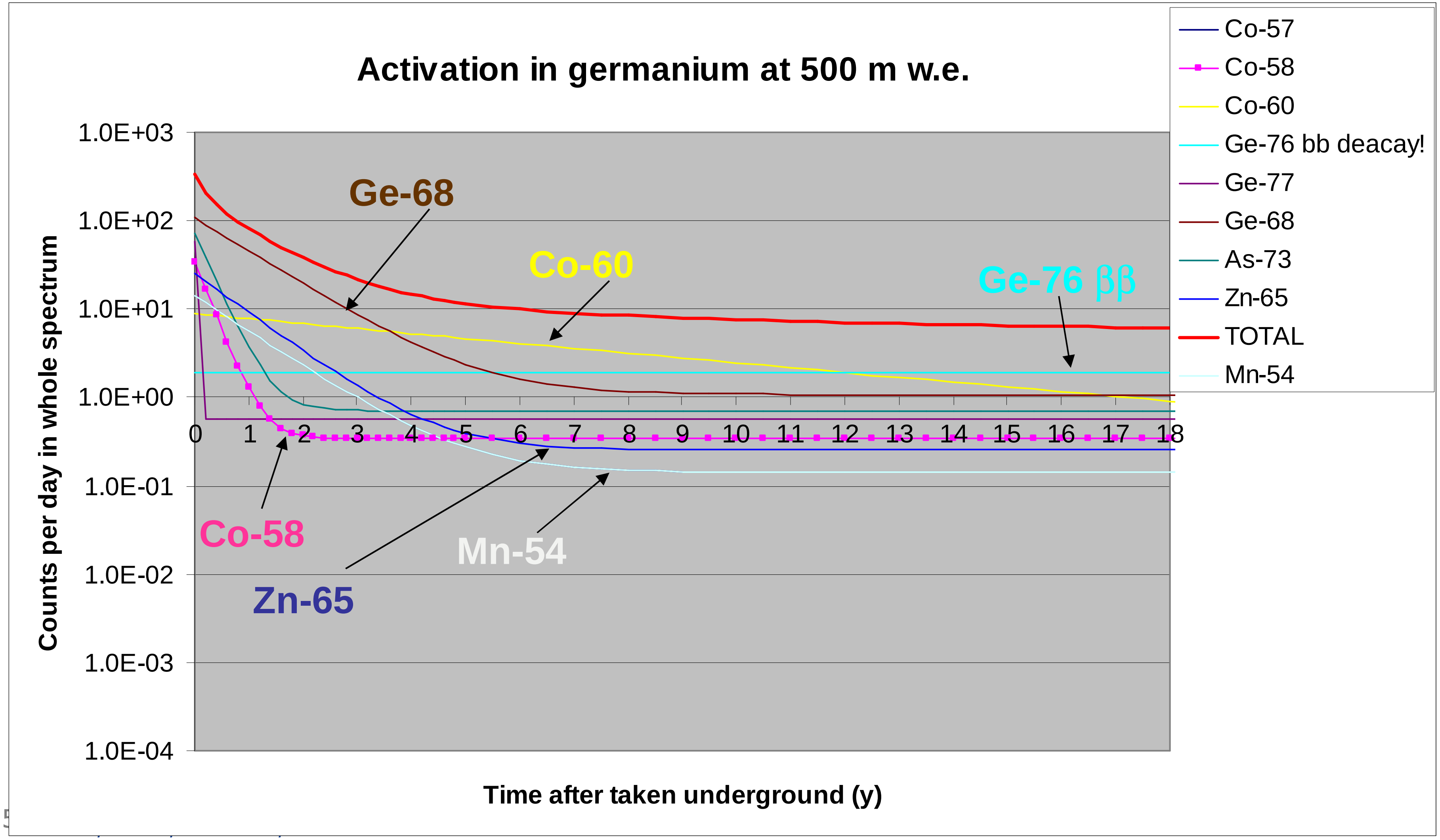
From ^{41}Ar produced in nuclear reactors.

^{40}K in big massive samples

The background subtraction could be incorrect since the sample will shield the detector from radioactivity in the shield

Detector Ge-5 – Background evolution





Activation in germanium at 500 m w.e.
Counts per day in whole spectrum
1.0E+03
1.0E+02
1.0E+01
1.0E+00
1.0E-01
1.0E-02
1.0E-03
1.0E-04
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
Time after taken underground (y)
Ge-68
Co-60
Ge-76 ββ
Co-58
Mn-54
Zn-65
Co-57
Co-58
Co-60
Ge-76 bb deacay!
Ge-77
Ge-68
As-73
Zn-65
TOTAL
Mn-54

QC & maintenance HPGe

- *Regular QC checks (daily/weekly/when changing sample) with reference source*
- *Log E-cal., FWHM, and possibly efficiency*
- *Measure background regularly and keep a log of it, both peak count rates as well as integrated count rate.*
- *Log leakage current and LN_2 consumption (weekly)*
- *Thermal cycle once per year – or more seldom (and clean Dewar)*

QC & maintenance HPGe

- *Keep cool at all times* (to avoid diffusion and increased deadlayers)
- *No H.V. unless cool – longer time to cool/heat for a big crystal*
- *Pump the cryostat when necessary !! (every XX year)*
  can vary from one detector to another depending on how good the vacuum seal is.
- *Remove ice from Dewar*
- *It is worth repairing an old (underground) detector!!*
  – a "big bad" detector is still valuable

Operation of HPGe

No electrical contact between detector and shield.
(not generally the case as we have examples were the detector work equally well with electrical connection)

Certain detectors can be sensitive to pressure to the detctor arm or window or endcap.

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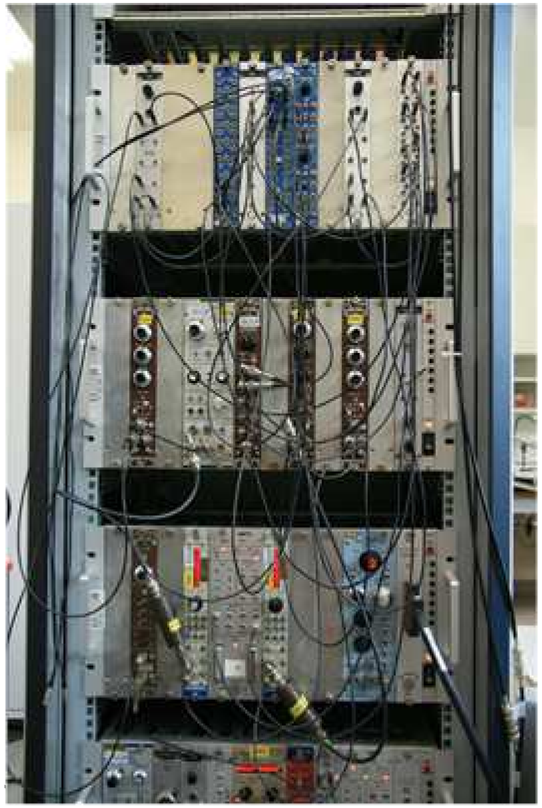
Coincidence techniques - Hardware

- *Analogue vs. digital*



Fast digitizers enable new instrumentations for safeguards work => Need for standards

(Anti-) Coincidence techniques - Hardware

- *Compton suppression*
- *Muon veto*
- *Finding the needle in a haystack (small peak in high background)*

Thank you
for your attention!

Stay in touch

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