E15 Chair for Experimental Physics and Astroparticle Physics

MLL

Lehrstuhl E12

Determination of the ^{60}Fe Half-Life - A Successful Colaboration in ERAWAST

Georg Rugel

supported by DFG (EXC 153)

PAUL SCHERRER INSTITUT

ERAWAST II, 29.8.-2.9.2011, Villigen

HZDR HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

HELMHOLTZ GEMEINSCHAFT

Outline

- Past Dreams at ERAWAST
- Work done
- Future DREAMS (DREsden AMS)

Status:

better than 1%

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

Already better than 4%

In progress

New value for the half live of ^{60}Fe

$$\frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

ICP-MS (instrument: Nu Plasma)
FZ Karlsruhe

Still high background problems to measure iron isotopes

Adding carrier (5mg added at time of iron extraction)

Absolute AMS measurement

AMS relative to a standard

At this workshop:

Measurement at PSI

Status $T_{1/2}$ (^{60}Fe) 02/2009

KARLSRUHER NUKLIDKARTE 7. Auflage 2006

CHART OF THE NUCLIDES, 7th Edition 2006
CARTE DES NUCLÉIDES, 7ème Edition 2006
CARTA DE NUCLEIDOS, 7ª Edición 2006
Таблица радионуклидов, 7-е издание 2006
核素图，第7版

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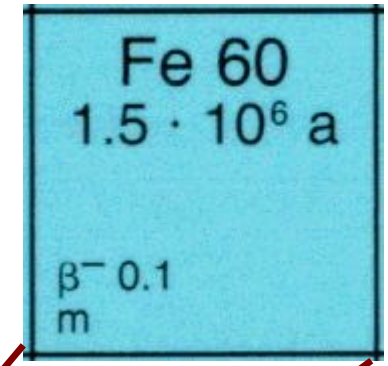
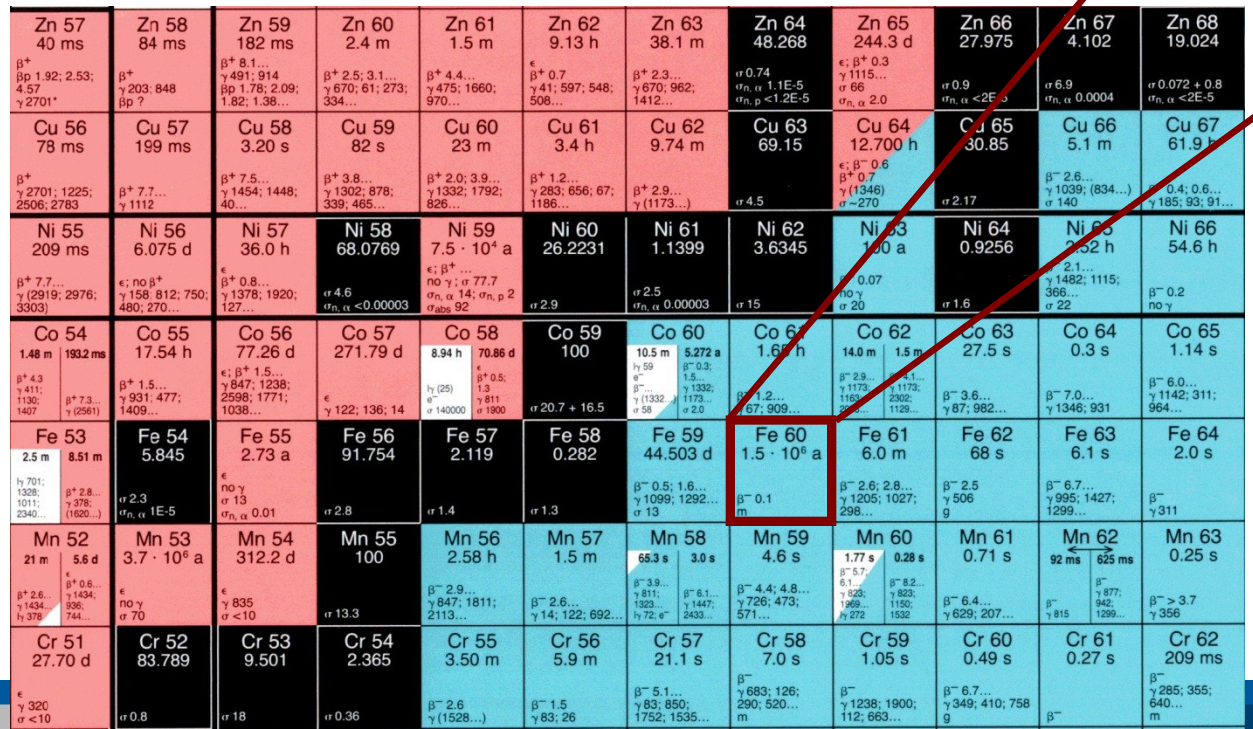
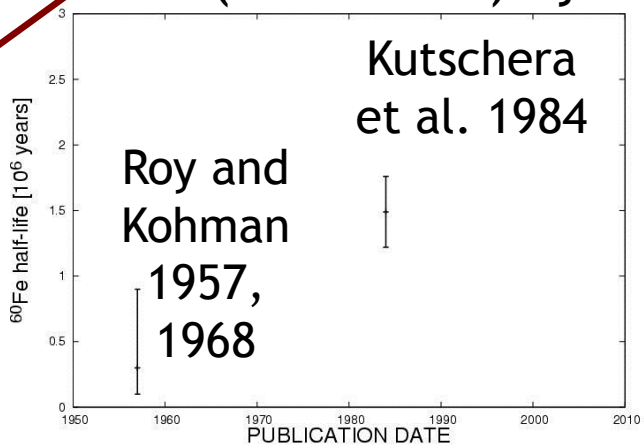
[2] formerly Forschungszentrum Karlsruhe in der Helmholtz-Gemeinschaft
P.O. Box 3640, 76021 Karlsruhe, Germany





(1.49 ± 0.27) Myr



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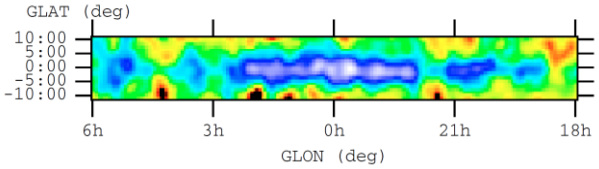
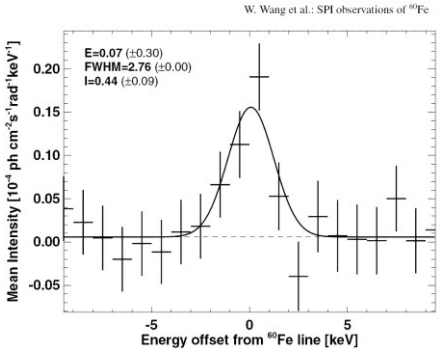
Motivation $T_{1/2}$ ^{60}Fe

Nucleosynthesis in the Galaxy

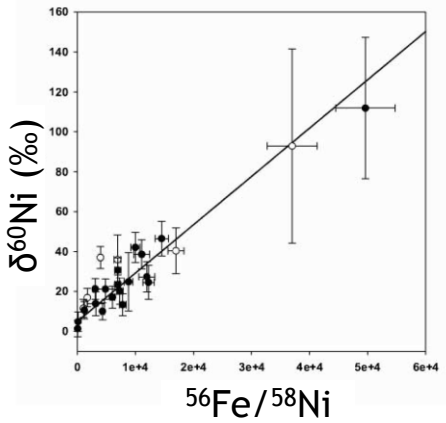
see e.g. R. Diehl, MPA



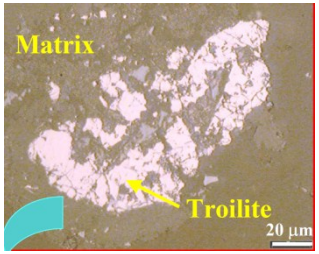


History of the Early Solar System

e.g. A. Shukolyukov and G.W. Lugmair, Science 1993



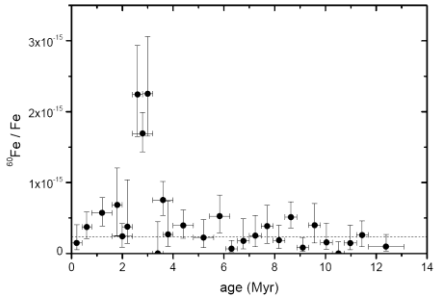
e.g: S. Mostefaoui et al., 2005



Deposits of supernova ejecta on Earth

e.g. K. Knie et al., PRL 2004;

C. Fitoussi et al., PRL 2008



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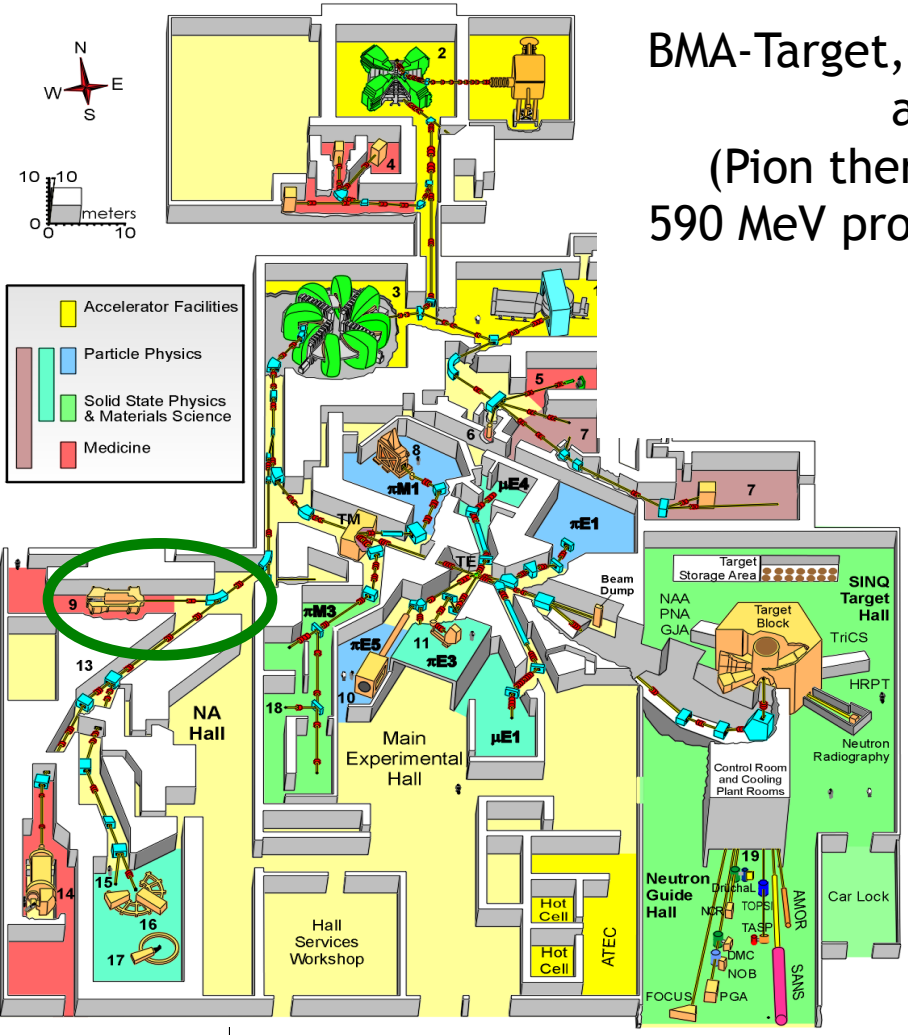
We need ^{60}Fe !

$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$



BMA-Target, Beam dump and shielding
(Pion therapy station, 590 MeV protons, 0.1Ah)

Irradiated from 1980 till Sept. 1992

Zn 58 84 ms β+ γ 203; 848 βp ?	Zn 59 182 ms β+ 8.1… γ 491; 914 βp 1.78; 2.09; 1.82; 1.38…	Zn 60 2.4 m β+ 2.5; 3.1… γ 670; 61; 273; 334…	Zn 61 1.5 m β+ 4.4… γ 475; 1660; 970…	Zn 62 9.13 h ε β+ 0.7 γ 41; 597; 548; 508…	Zn 63 38.1 m β+ 2.3… γ 670; 962; 1412…	Zn 64 48.268 σ 0.74 σn, α 1.1E-5 σn, p <1.2E-5	Zn 65 244.3 d ε; β+ 0.3 γ 1115… σ 66 σn, α 2.0	Zn 66 27.975 σ 0.9 σn, α <2E-5	Zn 67 4.102 σ 6.9 σn, α 0.0004	Zn 68 19.024 σ 0.072 + 0.8 σn, α <2E-5
Cu 57 199 ms β+ 7.7… γ 1112	Cu 58 3.20 s β+ 7.5… γ 1454; 1448; 40…	Cu 59 82 s β+ 3.8… γ 1302; 878; 339; 465…	Cu 60 23 m β+ 2.0; 3.9… γ 1332; 1792; 826…	Cu 61 3.4 h β+ 1.2… γ 283; 656; 67; 1186…	Cu 62 9.74 m β+ 2.9… γ (1173…)	Cu 63 69.15 σ 4.5	Cu 64 12.700 h ε; β− 0.6 β+ 0.7 γ (1346) σ ~270	Cu 65 30.85 σ 2.17	Cu 66 5.1 m β− 2.6… γ 1039; (834…) σ 140	Cu 67 61.9 h β− 0.4; 0.6… γ 185; 93; 91…
Ni 56 6.075 d ε; no β+ γ 158. 812; 750; 480; 270…	Ni 57 36.0 h ε β+ 0.8… γ 1378; 1920; 127…	Ni 58 68.0769 σ 4.6 σn, α <0.00003	Ni 59 7.5 · 10⁴ a ε; β+ … no γ; σ 77.7 σn, α 14; σn, p 2 σabs 92	Ni 60 26.2231 σ 2.9	Ni 61 1.1399 σ 2.5 σn, α 0.00003	Ni 62 3.6345 σ 15	Ni 63 100 a β− 0.07 no γ σ 20	Ni 64 0.9256 σ 1.6	Ni 65 2.52 h β− 2.1… γ 1482; 1115; 366… σ 22	Ni 66 54.6 h β− 0.2 no γ
Co 55 17.54 h β+ 1.5… γ 931; 477; 1409…	Co 56 77.26 d ε; β+ 1.5… γ 847; 1238; 2598; 1771; 1038…	Co 57 271.79 d ε γ 122; 136; 14	Co 58 8.94 h / 70.86 d ε β+ 0.5; 1.3 γ 811 σ 1900 hγ (25) e− σ 140000	Co 59 100 σ 20.7 + 16.5	Co 60 10.5 m / 5.272 a hγ 59 e− β− 0.3; 1.5… γ 1332; 1173… σ 2.0 β−… γ (1332… σ 58	Co 61 1.65 h β− 1.2… γ 67; 909…	Co 62 14.0 m / 1.5 m β− 2.9… γ 1173; 1163; 2003… β− 4.1… γ 1173; 2302; 1129…	Co 63 27.5 s β− 3.6… γ 87; 982…	Co 64 0.3 s β− 7.0… γ 1346; 931	Co 65 1.14 s β− 6.0… γ 1142; 311; 964…
Fe 54 5.845 σ 2.3 σn, α 1E-5	Fe 55 2.73 a ε no γ σ 13 σn, α 0.01	Fe 56 91.754 σ 2.8	Fe 57 2.119 σ 1.4	Fe 58 0.282 σ 1.3	Fe 59 44.503 d β− 0.5; 1.6… γ 1099; 1292… σ 13	Fe 60 1.5 · 10⁶ a β− 0.1 m	Fe 61 6.0 m β− 2.6; 2.8… γ 1205; 1027; 298…	Fe 62 68 s β− 2.5 g	Fe 63 6.1 s β− 6.7… γ 995; 1427; 1299…	Fe 64 2.0 s β− γ 311
Mn 53 3.7 · 10⁶ a ε no γ σ 70	Mn 54 312.2 d ε γ 835 σ <10	Mn 55 100 σ 13.3	Mn 56 2.58 h β− 2.9… γ 847; 1811; 2113…	Mn 57 1.5 m β− 2.6… γ 14; 122; 692…	Mn 58 65.3 s / 3.0 s β− 3.9… γ 811; 1323… hγ 72; e− β− 6.1… γ 1447; 2433…	Mn 59 4.6 s β− 4.4; 4.8… γ 726; 473; 571…	Mn 60 1.77 s / 0.28 s β− 5.7; 6.1… γ 823; 1969… hγ 272 β− 8.2… γ 823; 1150; 1532	Mn 61 0.71 s β− 6.4… γ 629; 207…	Mn 62 92 ms / 625 ms β− γ 815 β− γ 877; 942; 1299…	Mn 63 0.25 s β− > 3.7 γ 356

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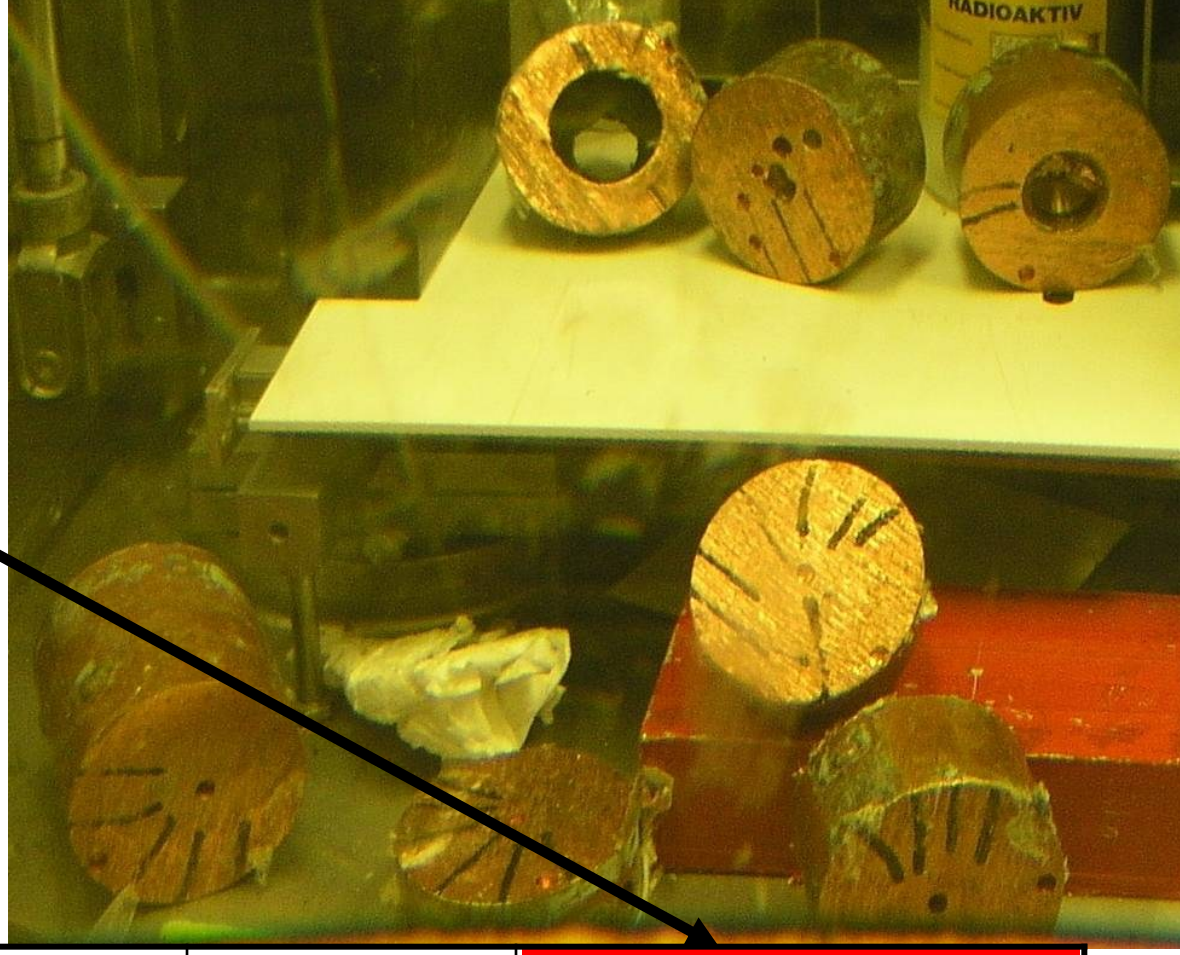
Drilling a hole into central part:

3.86g copper



Activation measurement of 2kg of the beamdump:

Date	Activity ^{60}Co [Bq]	^{60}Co atoms	Initial sample ^{60}Co [Bq]	Initial sample ^{60}Co atoms
01.09.1992	7×10^{9}	1.8×10^{18}	1.4×10^{7}	**3.5×10^{15}**
08.07.2005	1.4×10^{9}	3.3×10^{17}	2.6×10^{6}	6.4×10^{14}

Iron sample after the first chemical separation steps (Okt 2004)

$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$

Volume: 4000 µl
from master solution (~ 1nHCl)
Weight: 4.036 g

+ 1000 µl H_2O

Germanium detector

Same material used for ICP-MS
(March 2008)

$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$

Efficiency calibration of the detector:

Avoiding geometrical corrections etc.

Calibration source (^{60}Co) with the same geometry:

5ml 0.1 nHCl

102.0 (± 1.5) Bq ^{60}Co

(all uncertainties 1 sigma)

Germanium detector

$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$

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5ml 0.1 nHCl

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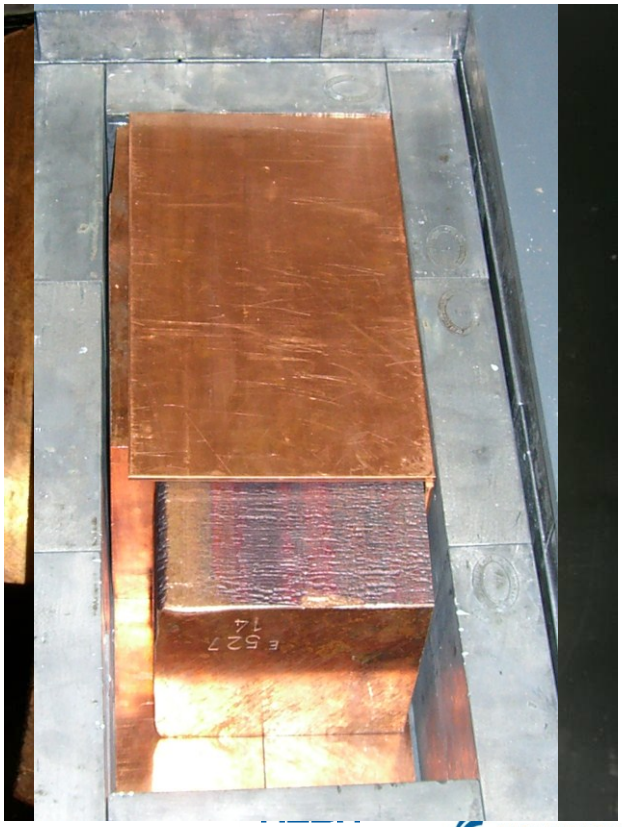
Germanium detector



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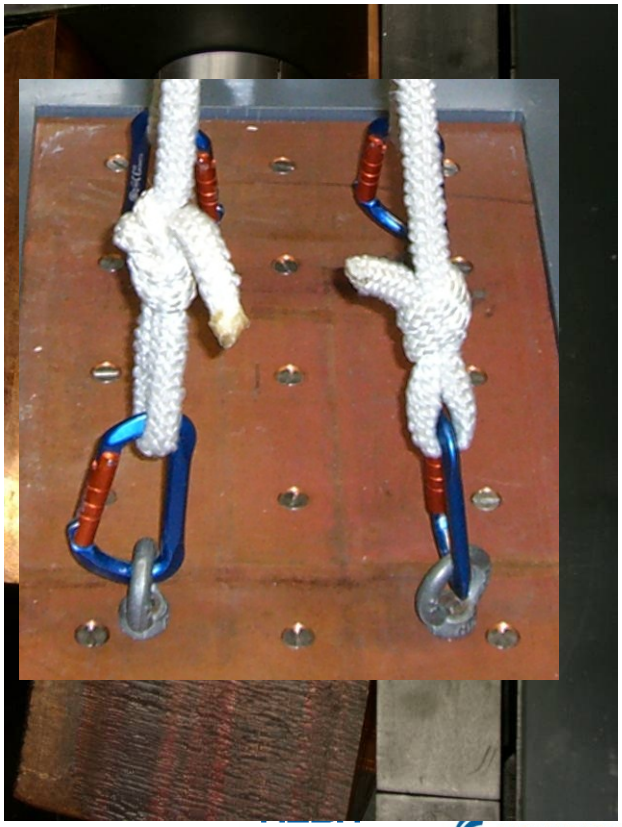

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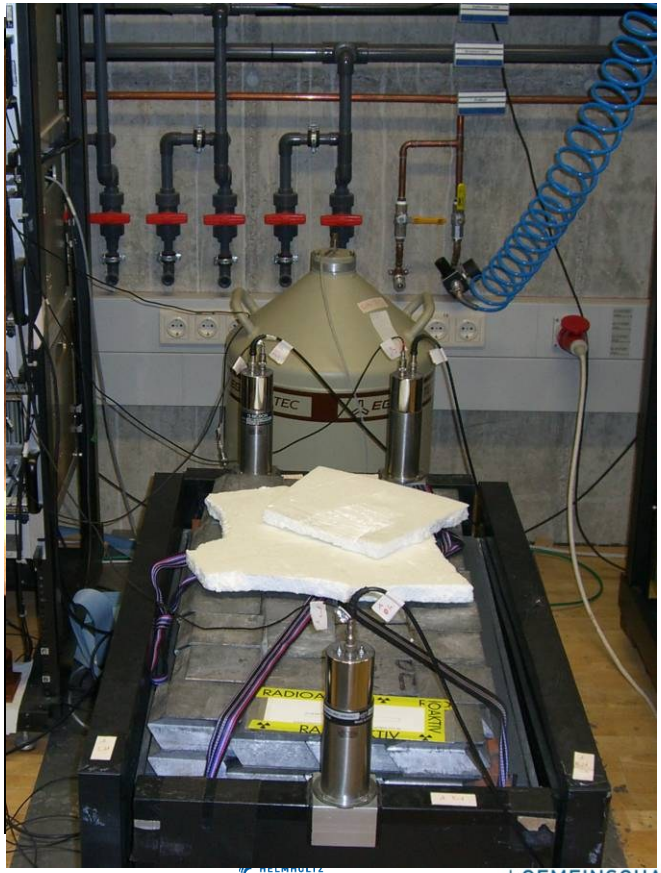
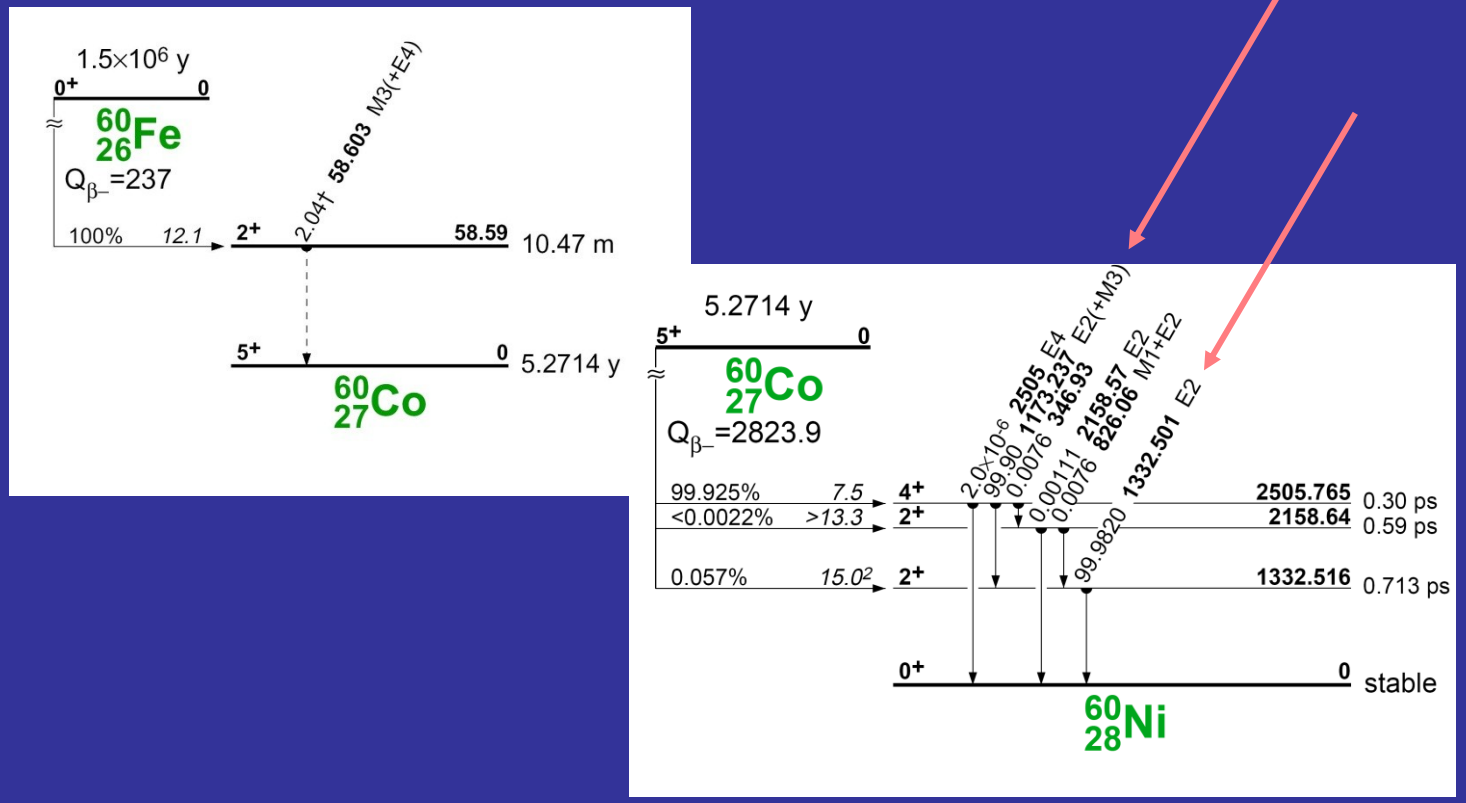
→ Germanium detector



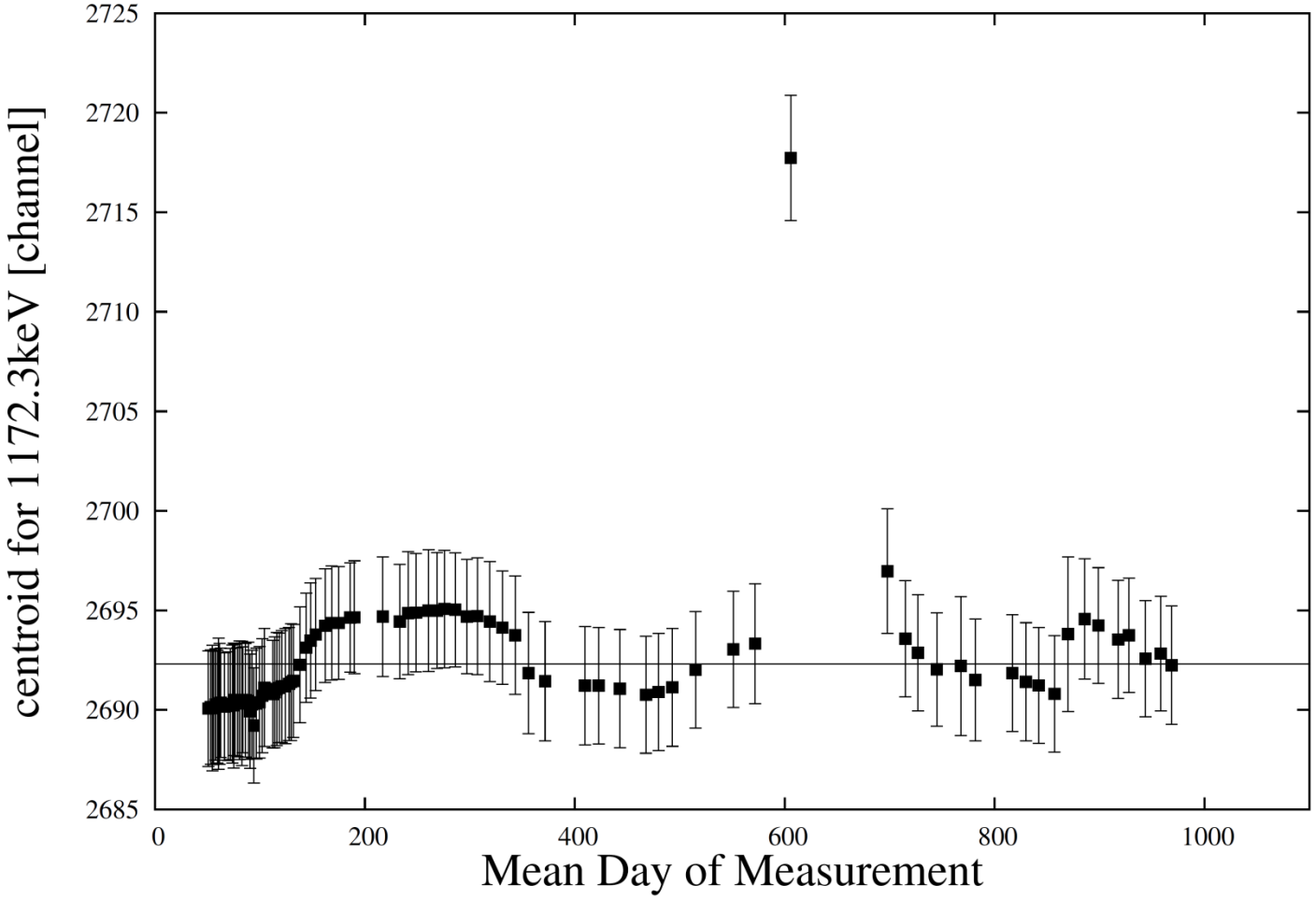
Build-up of the ^{60}Co activity



$$A_{^{60}Co} = N_{^{60}Fe}\lambda_{^{60}Fe} \cdot (1 - e^{-\lambda_{^{60}Co} \cdot t})$$

$$\approx N_{^{60}Fe} \cdot \lambda_{^{60}Fe} \cdot \lambda_{^{60}Co} \cdot t$$ for $t << T_{1/2,^{60}\mathrm{Co}}$

$$\approx N_{^{60}\mathrm{Fe}} \cdot \lambda_{^{60}\mathrm{Fe}}$$ for $t >> T_{1/2,^{60}\mathrm{Co}}$


centroid for 1172.3keV [channel]
2725
2720
2715
2710
2705
2700
2695
2690
2685
0
200
400
600
800
1000
Mean Day of Measurement

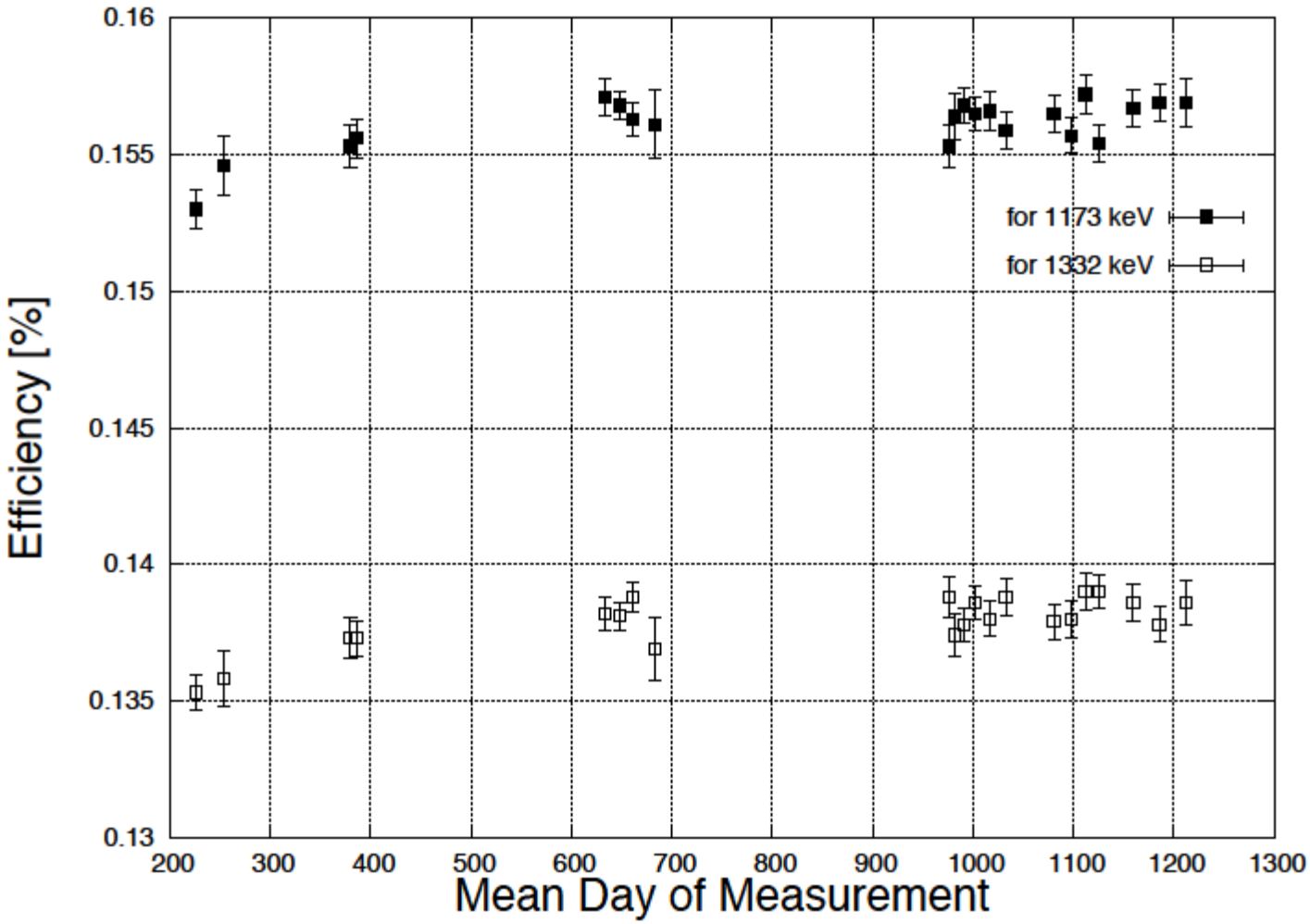

0.16
0.155
0.15
0.145
0.14
0.135
0.13
Efficiency [%]
200
300
400
500
600
700
800
900
1000
1100
1200
1300
Mean Day of Measurement
for 1173 keV
for 1332 keV

TABLE I. Efficieny used for the data analysis. The column days refers always to the mean of the days after the chemical extraction. The uncertainty value given is only statistics.

days	efficiency [%] for 1.17 MeV line	1.33 MeV line	distance comment
50–80	1.157(10)	1.000(10)	close geometry
83–190	1.207(6)	1.065(5)	close geometry mod.
217	0.229(7)	0.199(6)	7.55 cm; 2mm up
233–371	0.1546(4)	0.1365(4)	10 cm
410–606	0.1563(3)	0.1380(3)	10 cm
698–969	0.1565(2)	0.1383(2)	10 cm
976–1212	0.1564(2)	0.1384(2)	10 cm

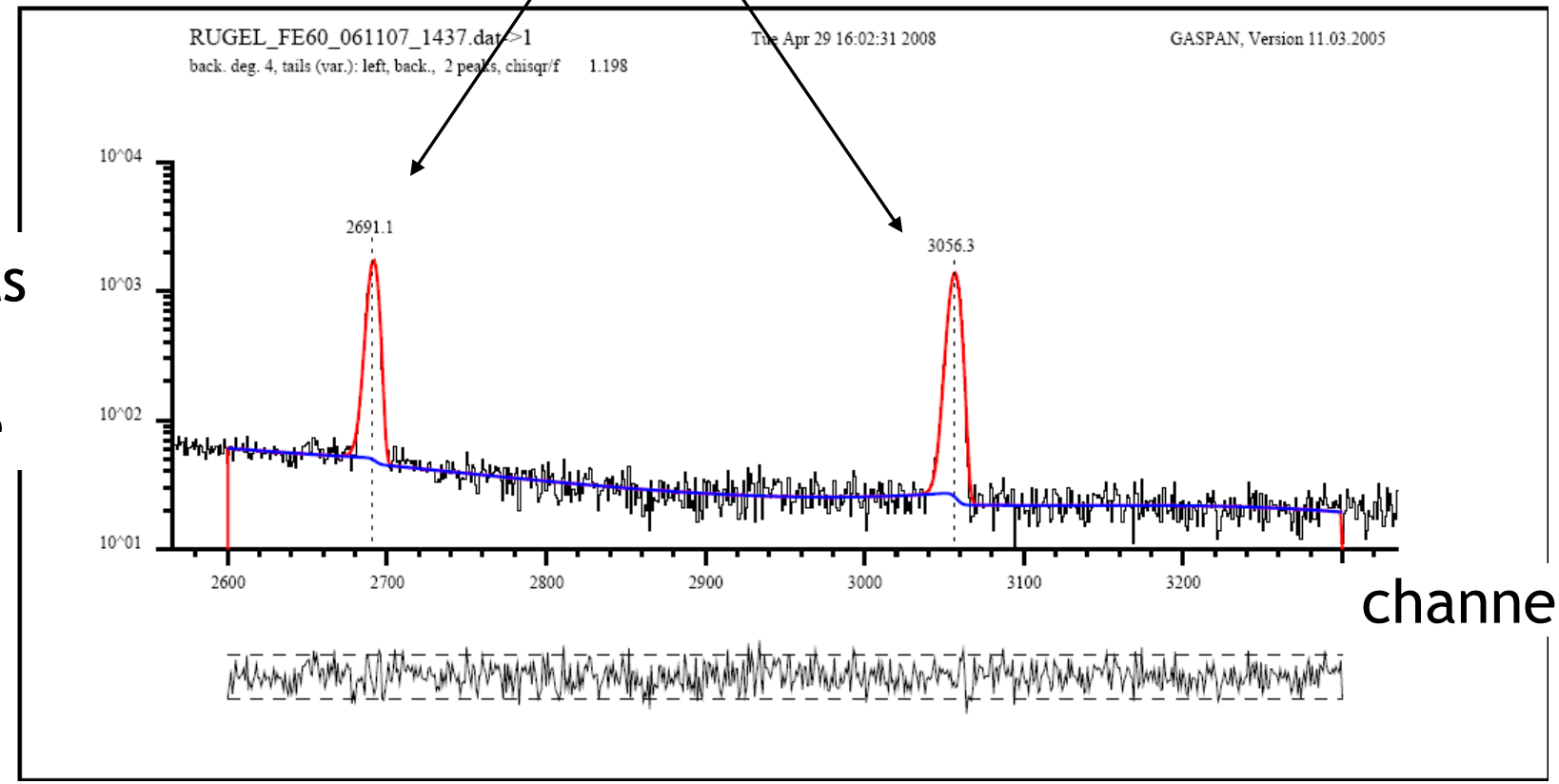
$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$

^{60}Co

12710±130 cts

11450±120 cts

Counts log scale



channel

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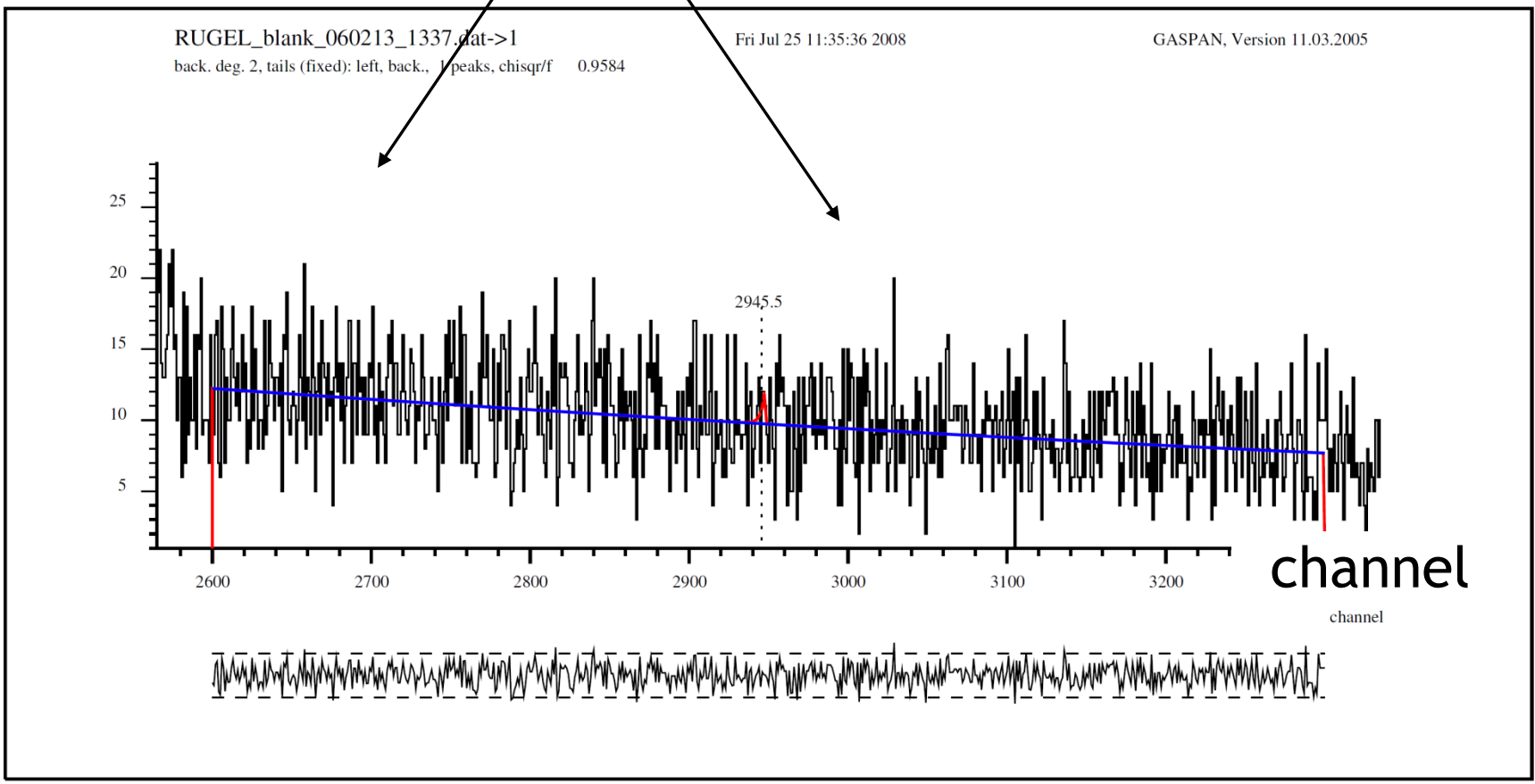
Example of a Background spectrum

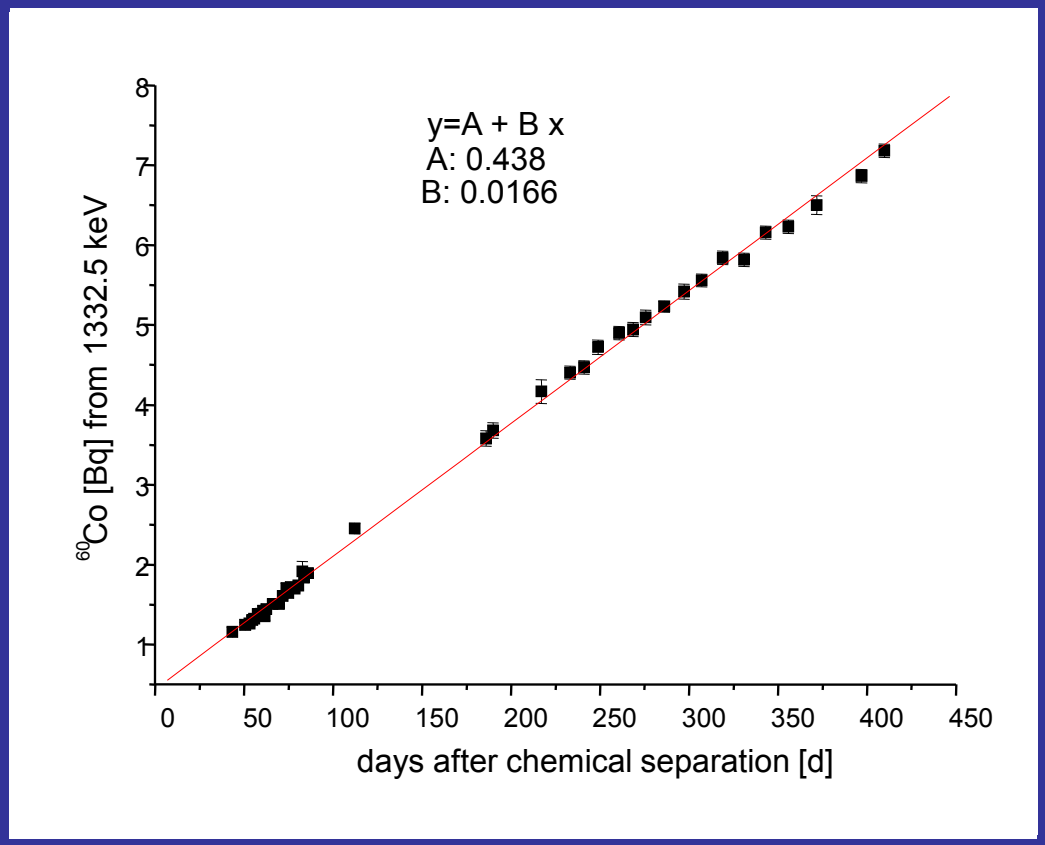
$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$

^{60}Co

< 0.5 mBq



Build-up of the ^{60}Co activity



T= 0

^{60}Co ~ 0.3 Bq

Chemical reduction at least 10^7 !

Saturation activity

46.1 Bq

With $T_{1/2}$ (TOI)

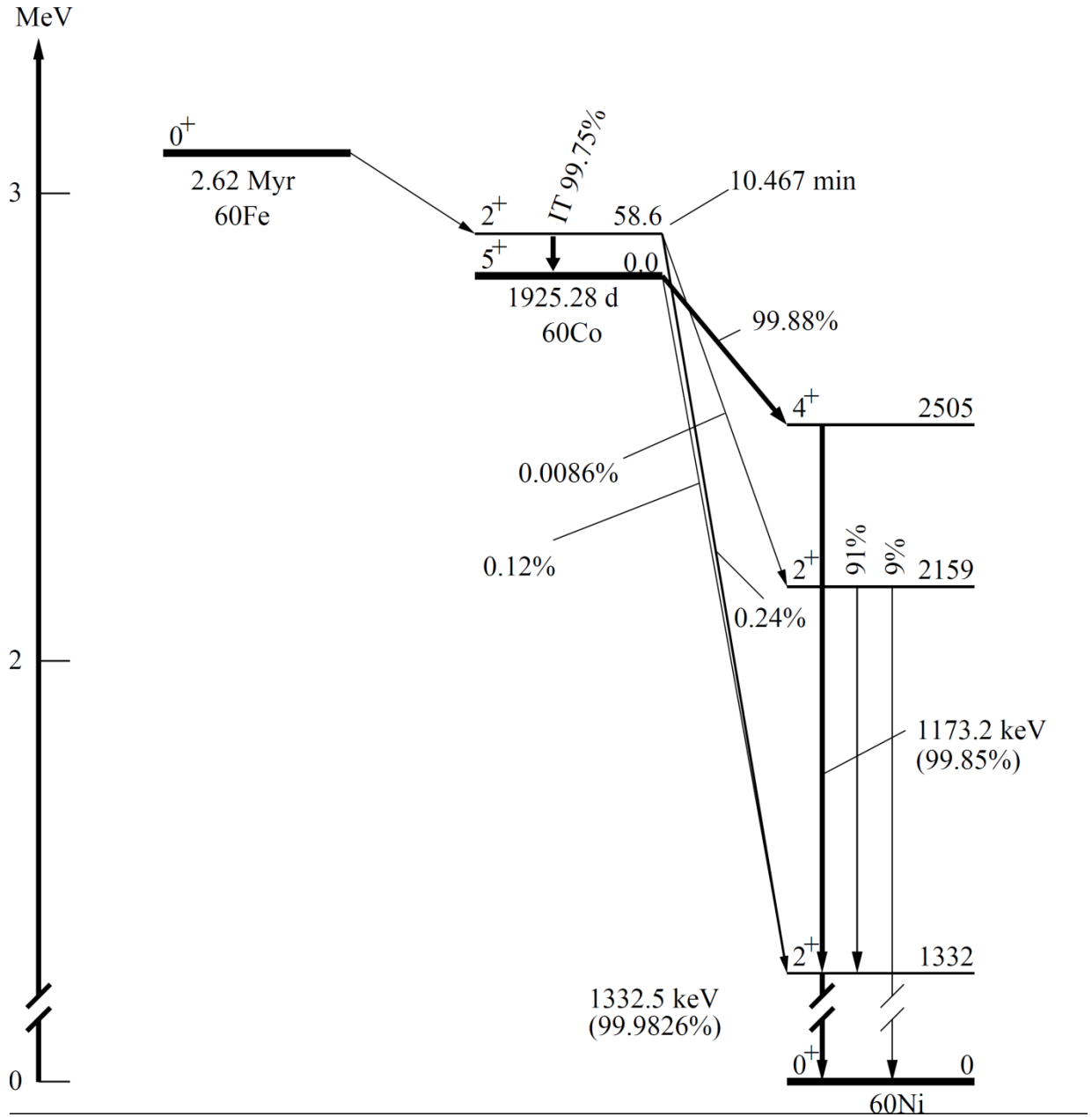
3.1 x 10^{15} ^{60}Fe

$$A_{^{60}Co} = N_{^{60}Fe}\lambda_{^{60}Fe} \cdot (1 - e^{-\lambda_{^{60}Co} \cdot t})$$

$$\approx N_{^{60}Fe} \cdot \lambda_{^{60}Fe} \cdot \lambda_{^{60}Co} \cdot t$$

for $t << T_{1/2,^{60}\mathrm{Co}}$

Closer look to the decay of ^{60}Fe



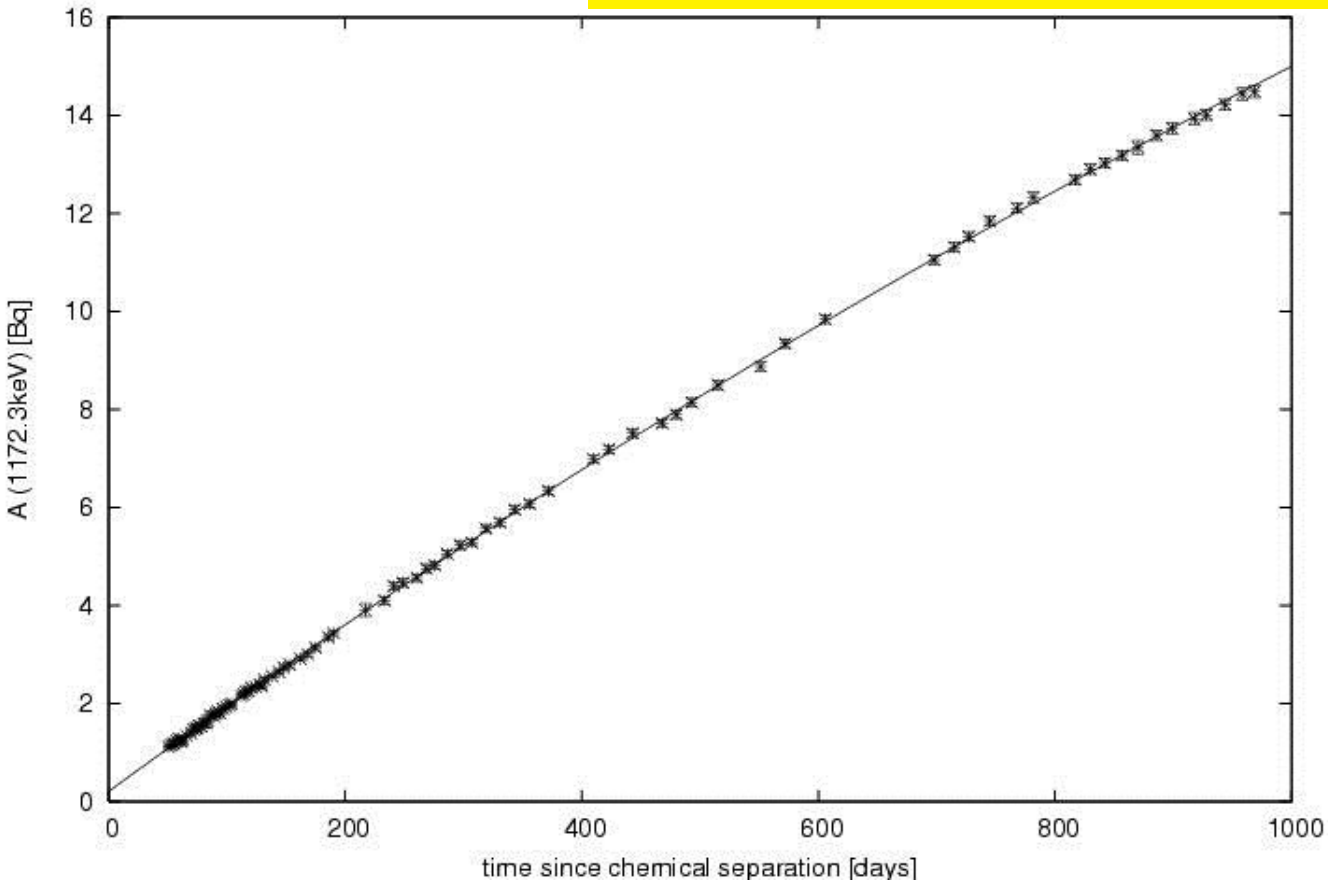
Data from:

Tuli NDS, 2003:

Schmidt-Ott, 1963

Build-up of ^{60}Co

$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$



T= 0

^{60}Co ~ 0.207 Bq

Chemical reduction at least 10^7 !

Saturation activity

49.14±0.08 Bq

$$A_{^{60}Co} = N_{^{60}Fe}\lambda_{^{60}Fe} \cdot (1 - e^{-\lambda_{^{60}Co} \cdot t})$$

for the 1172.3 keV line

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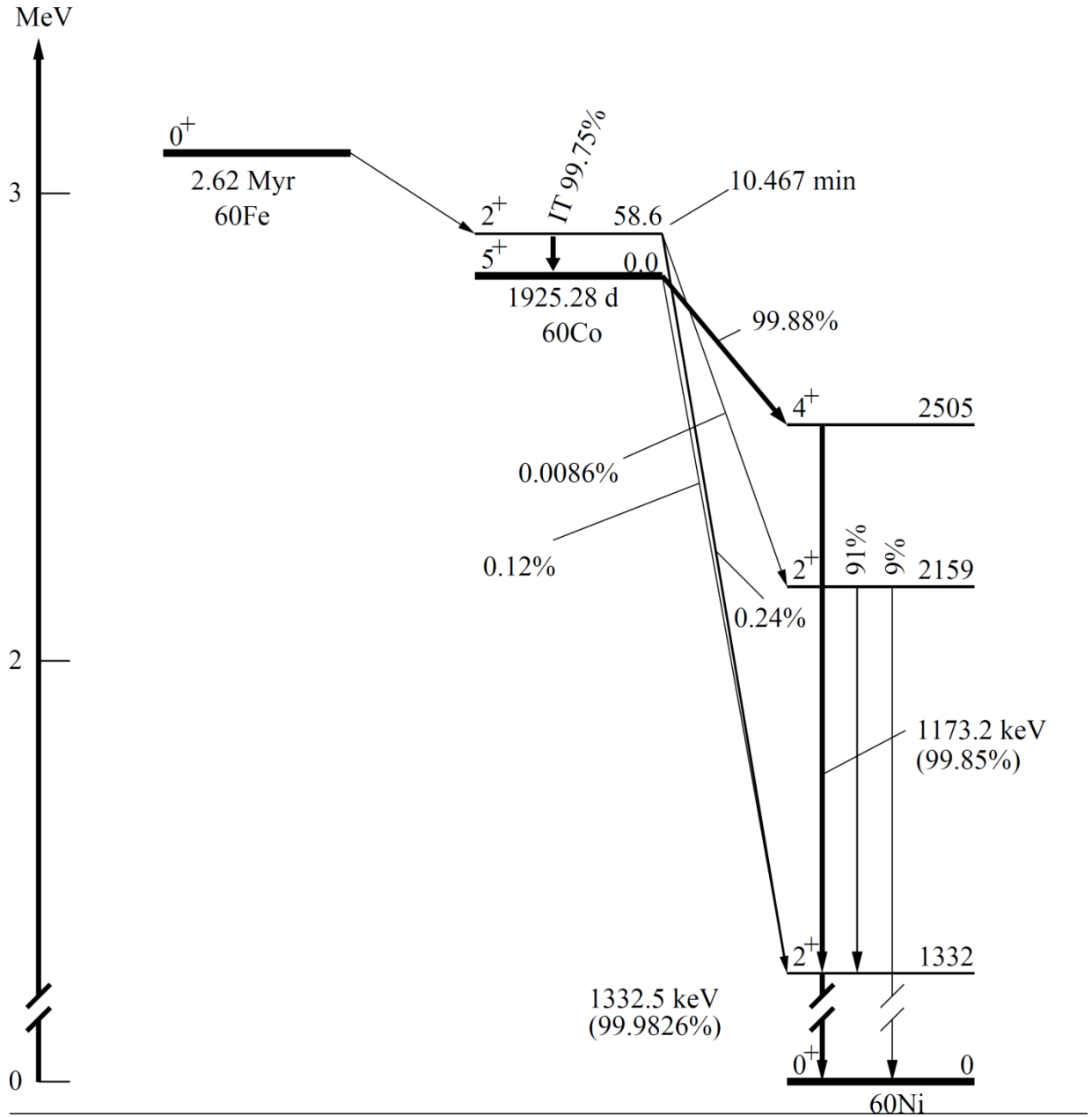
Closer look to the decay of ^{60}Fe

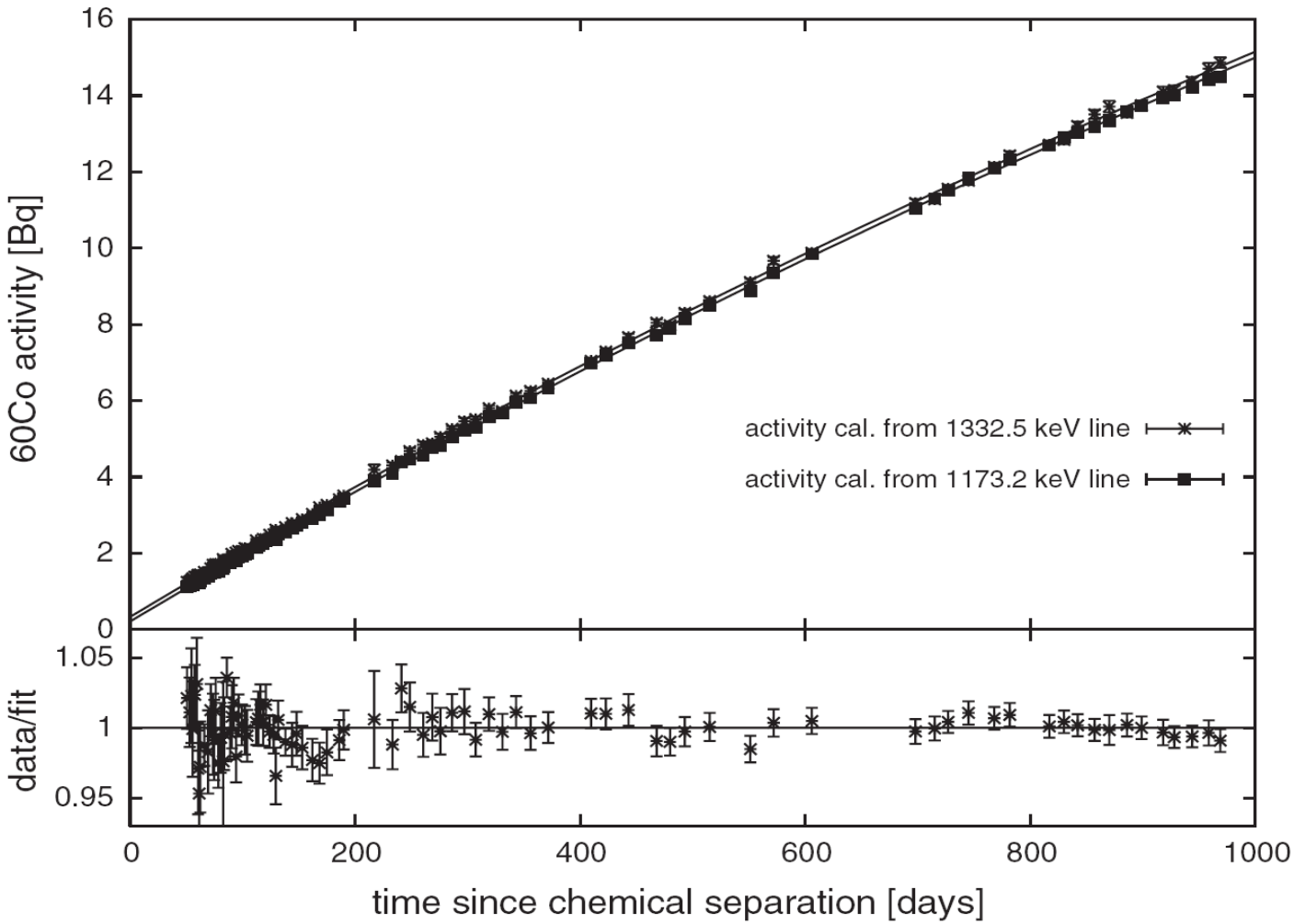


Data from:

Tuli NDS, 2003:

Schmidt-Ott, 1963

Grow in of the ^{60}Co activity



Combined result: 49.26±0.07 Bq

Master Sample (TUM) transferred to PSI

$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$

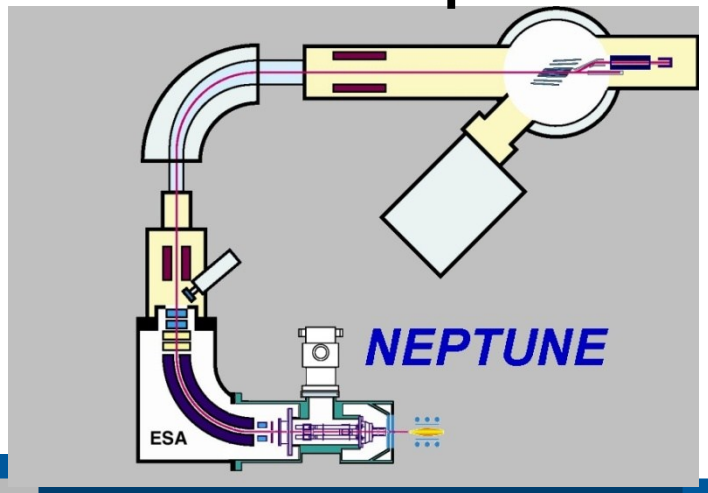
Determination of N

Subsamples taken gravimetrically after opening

$$\cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}}$$

$$\cdot N_{\mathrm{Fe}}$$

Multicollector – Inductively Coupled Plasma Mass Spectrometry MC-ICP-MS



PAUL SCHERRER INSTITUT PSI

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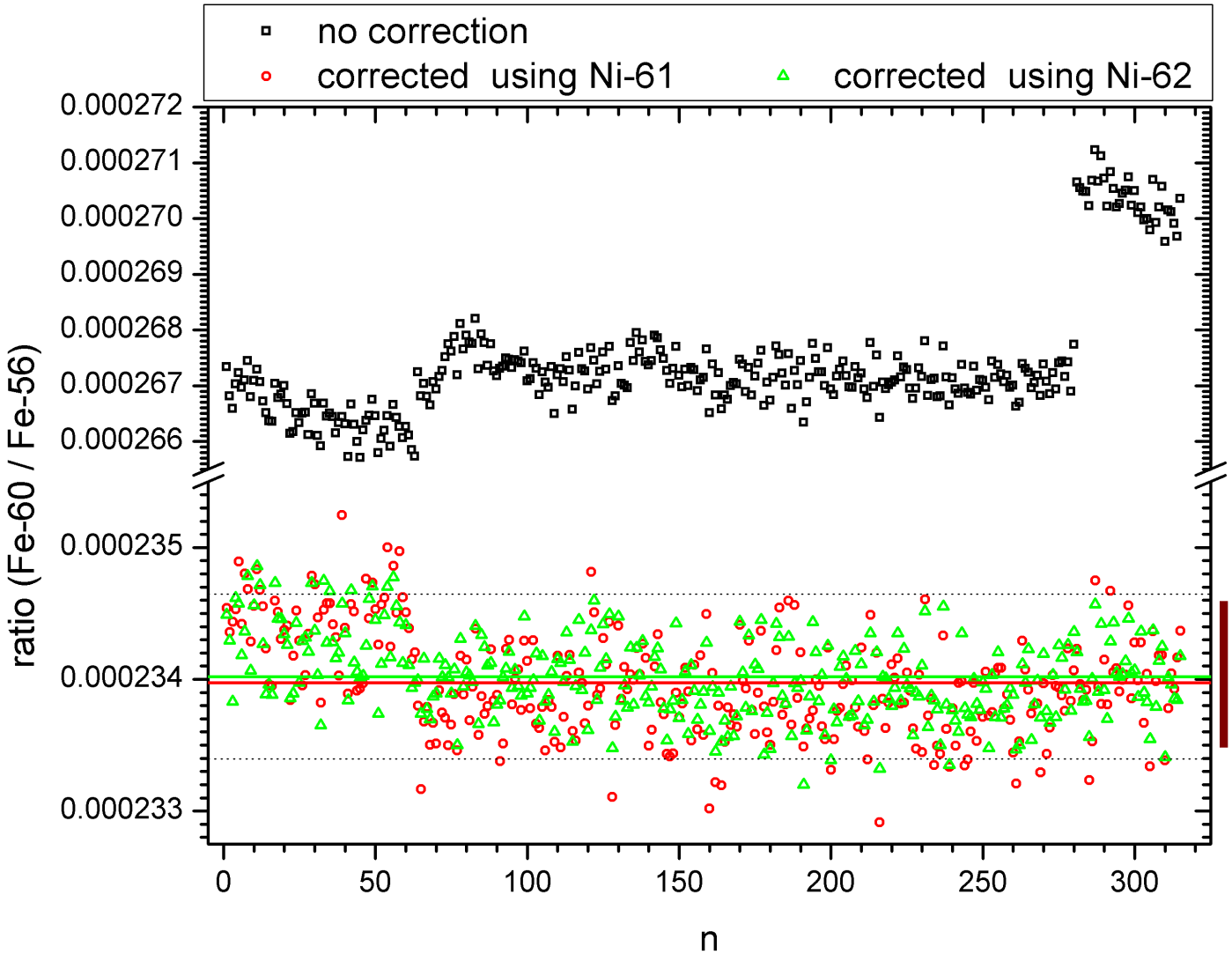
From N. Kivel

^{60}Ni interference correction

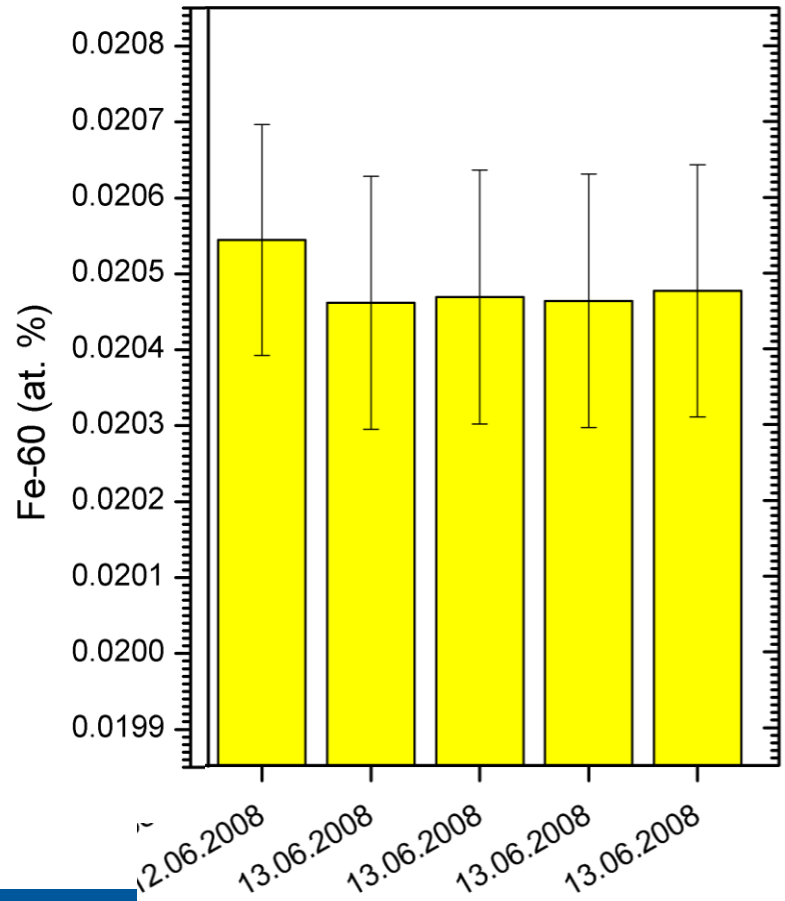


Uncertainty: 0.17% !

From N. Kivel

Isotopic composition

$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$



$(2.0483 \pm 0.0035) \times 10^{-4}$

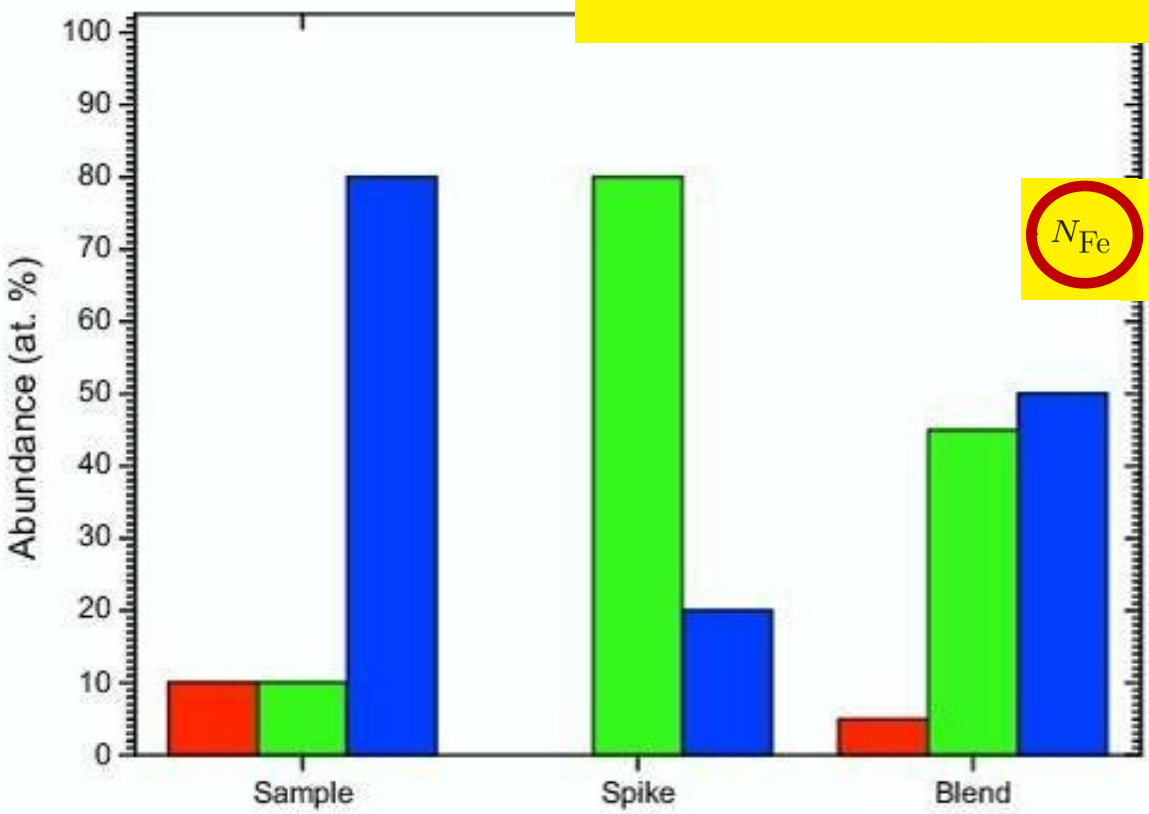
From N. Kivel

Isotope Dilution MS

isotope	^{54}Fe	^{56}Fe	^{57}Fe	^{58}Fe
percentage [%]	-	3.11	95.10	1.79

$$A_{^{60}Fe} = \lambda_{^{60}Fe} \cdot N_{^{60}Fe} = \frac{ln(2)}{T_{1/2}(^{60}Fe)} \cdot \frac{N_{^{60}Fe}}{N_{Fe}} \cdot N_{Fe}$$

N_{Fe} (2.662±0.009)mg



$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$

TABLE II: The various contributions to the uncertainty (1 σ) of the three measurements are listed.

	Rel. Uncertainty [%]	
	stat.	syst.
$A_{^{60}\mathrm{Fe}}$ (*master sample*)		
^{60}Co standard		1.5%
fit	0.23%	
N_{Fe} (*ID sample*)		
weighing		0.18%
ID-ICP-MS	0.28%	
$N_{^{60}\mathrm{Fe}}/N_{\mathrm{Fe}}$ (*N sample*)		
ICPMS	0.18%	
total	0.4%	1.51%

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Combination

$$A_{^{60}\mathrm{Fe}} = \lambda_{^{60}\mathrm{Fe}} \cdot N_{^{60}\mathrm{Fe}} = \frac{ln(2)}{T_{1/2}(^{60}\mathrm{Fe})} \cdot \frac{N_{^{60}\mathrm{Fe}}}{N_{\mathrm{Fe}}} \cdot N_{\mathrm{Fe}}$$

(2.62 ± 0.04) x 10^6 yr

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New Measurement of the ^{60}Fe Half-Life

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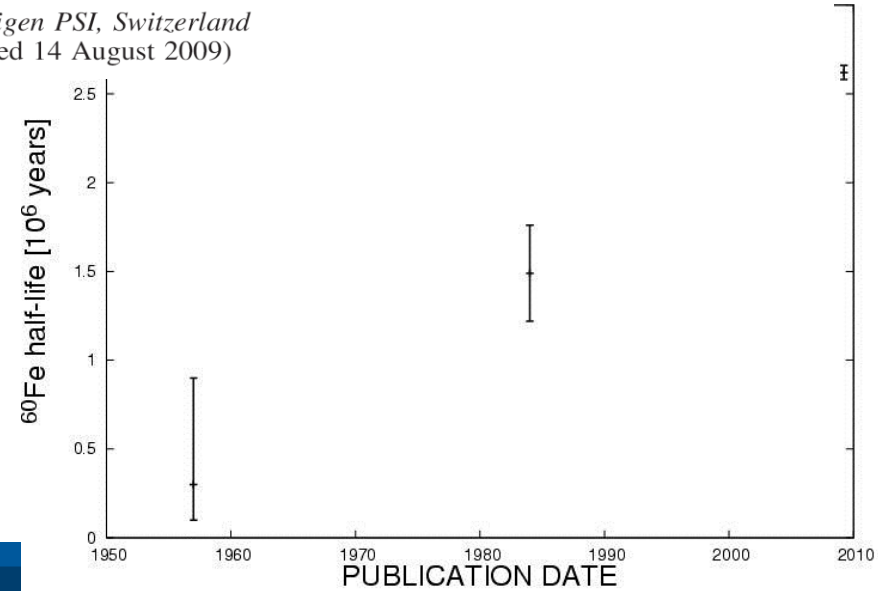
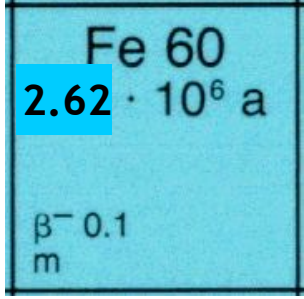
(Received 25 March 2009; published 14 August 2009)

More details in: N. Kivel et al.



TOI: $T_{1/2}(^{60}\mathrm{Fe})$: (1.49 ± 0.27) Myr

Thanks to my colleagues



Georg Rugel, *Thomas Faestermann, Klaus Knie, Gunther Korschinek, Mikhail Poutivtsev*
Technische Universität München

Dorothea Schumann, Regin Weinreich, Ines Günther-Leopold, Niko Kivel, Michael Wohlmuther
Paul Scherrer Institut, Villigen, Switzerland

AMS (Accelerator Mass Spectrometry)

Facility DREAMS (DREsden AMS)

Shavkat Akhmadaliev, Silke Merchel,
Stefan Pavetich, Georg Rugel (FWIA)

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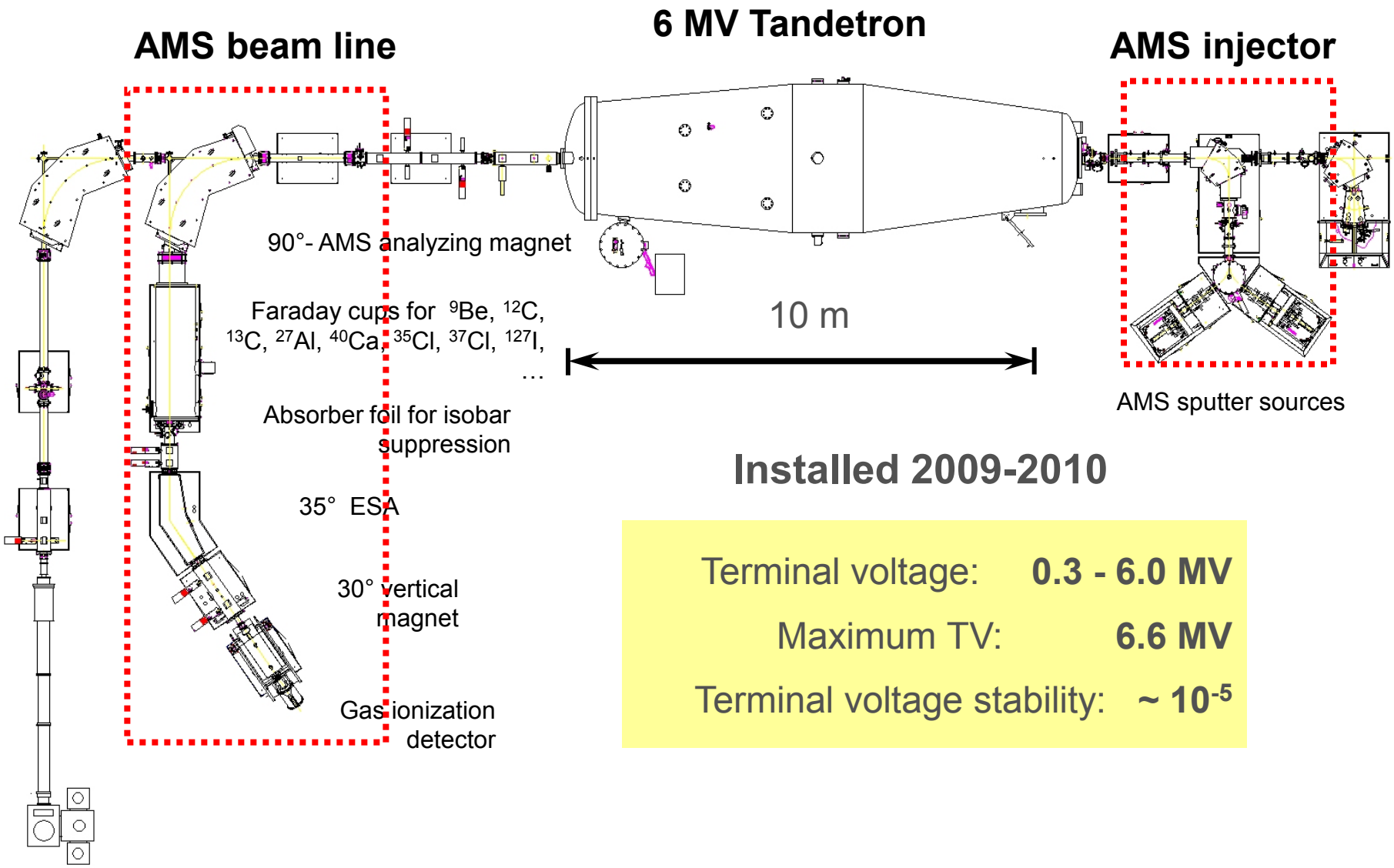
DREsden AMS setup with a 6 MV Tandetron



Installed 2009-2010

Terminal voltage:	**0.3 - 6.0 MV**
Maximum TV:	**6.6 MV**
Terminal voltage stability:	**~ 10^{-5}**

Thank you for your attention!

I have DREAMS

HZDR
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HELMHOLTZ GEMEINSCHAFT