Measurement of the relative gamma-ray decay probabilities from the electron capture decay of ^{153}Gd

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Contents of the talk

- Motivation of the study.
- Notes on current evaluations and previous measurements.
- Experimental technique.
- Results of the measurements.
- Comparison of the results.

Gadolinium-153 – Motivation for Study

- Gadolinium-153 is used as a line source for SPECT.[1]
- Historically was used in bone absorptiometry, bone mineral density and marrow content studies.[2]
- Many non-destructive testing (NDT) products use the isotope for profiling purposes. (the LIXI profiler[3])
- Identified as a potential radionuclide use for interstitial rotating shield brachytherapy (I-RBST) to replace ^{192}Ir. [4]

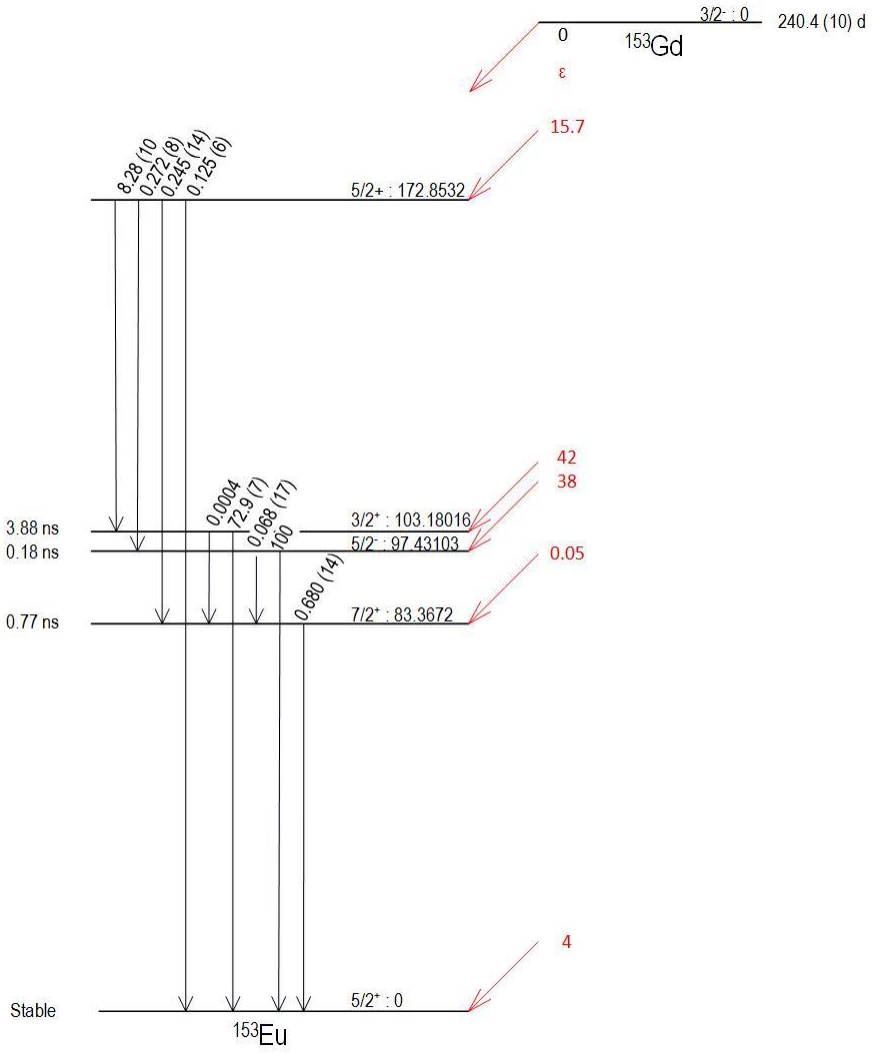
The electron capture of ^{153}Gd

^{153}Gd decays 100% of the time to the A=153 stable isobar ^{153}Eu.

4 excited levels are populated following the decay in the daughter.

Gamma rays seen in previous work can not be placed in accordance to the current evaluated level scheme [5].

This evaluation disagrees with recent evaluation by [6], which includes (7/2+) 269.7 keV state and the 7/2- 151.6 keV state.



Data taken from [5]

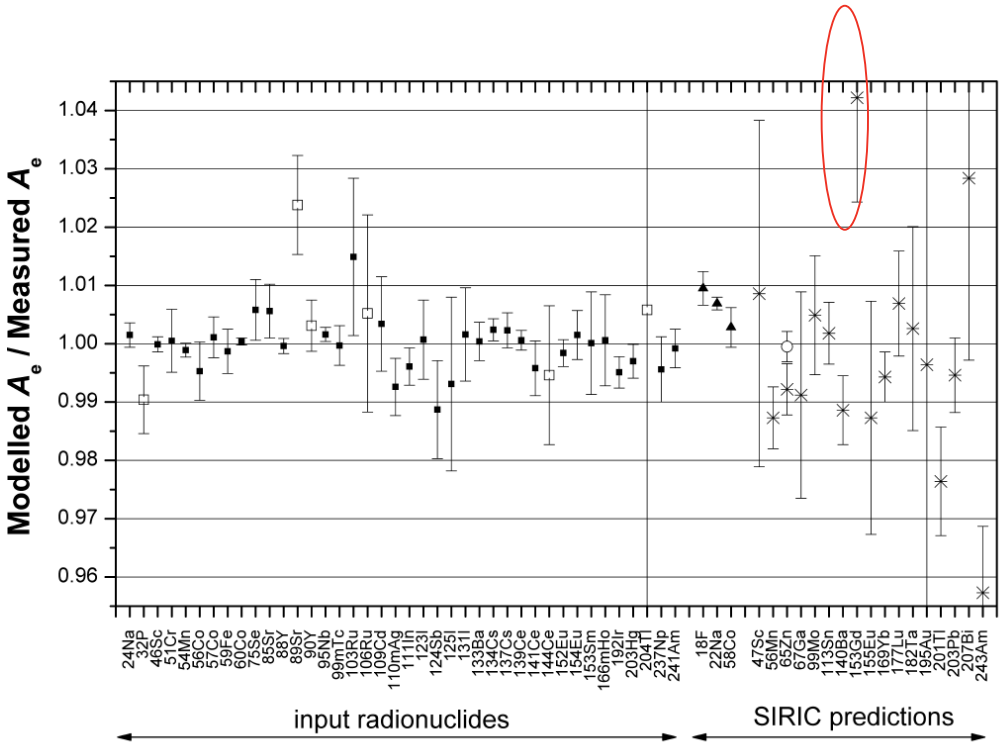
Previous evaluations and comments

- DDEP [5] evaluation has noted several discrepant measurements related to the electron capture decay of ^{153}Gd, "***The values for several γ–rays are discrepant… A good new measurement of the intensities of the weak lines is desirable***".

- Recent evaluation in 2010 [6] rebuilds decay scheme (different to the DDEP evaluation) also calls for the need for new measurements of the decay. ***"…there are large discrepancies among the measured relative intensities of 54.2, 68.2 and 151.6 keV rays, so a new better measurement is desirable."***

Notes on absolute intensities

- BIPM-Monographie 7, "*Measurement modelling of the International Reference System (SIR) for gamma emitting radionuclides*" found the following discrepancy between modelled and experimental data in the case ^{153}Gd for the measured activity and the SIRIC predicted value according to a well understood least squares fitting to the efficiency of the counter. Similar nuclides with similar decay properties were not in disagreement.



- It was stated, "***The discrepancy for the ^{153}Gd is probably due to nuclear data… The efficiency curve therefore provides motivation for to investigate the decay data of that radionuclide."***

Previous measurements of the decay of ^{153}Gadolinium

γ (keV)	I_γ												
	Heath (1974)	Sergienko et al. (1974)	Grigorev et al. (1981)	Singh et al. (1985)	Subrahmanyeswara et al. (1988)	Venkateswara et al. (1988)	Chechev et al. (1992)	Chand et al. (1992)	Egorov et al. (1993)	Kumar et al. (1995)	Laurec et al. (1996)	LRSW	Evaluated values
Kα1							204(4)		208(8)			205(4)	205(4)
Kα2							114(2)		114(4)			114(2)	114(2)
Kα		321(11)		150.2(35)	340.51(380)	313(8)		302.4(78)		323.3(82)		325(15)	325(15)
Kβ1							65.2(14)	60.5(20)	65(3)	69.2(19)		65.1(17)	65.1(17)
Kβ2							17.5(4)	15.9(6)	17.5(7)	16.84(26)		16.95(27)	16.95(27)
Kβ		78(11)		32.9(5)	84.90(84)	78.9(11)		76.4(21)				82.6(23)	82.6(23)
14.06383(20)				0.054(9)	0.146(15)	0.09(1)		0.11(3)	0.10(2)	0.051(5)		0.068(17)	0.068(17)
19.81296(19)				0.089(9)	0.072(11)	0.006(1)		0.06(2)	< 0.03	0.019(3)			0.00040(4)[a]
21.2287(8)					0.07(2)				< 0.03	0.078(16)		0.075(12)	0.075(12)
54.1934(4)		< 0.01		0.091(3)	0.058(8)					0.027(2)			0.021(5)[b]
68.2557(5)		0.04(1)			0.071(7)	0.035(14)		0.064(17)		0.072(11)		0.060(8)	0.017(4)[b]
69.67300(13)	7.8(2)	8.4(3)		8.35(32)	8.60(15)	8.31(13)	8.41(22)	7.97(20)		8.20(26)	8.20(16)	8.27(8)	8.27(8)
75.42213(23)	0.30(3)	0.26(8)		0.26(8)	0.278(31)	0.27(1)		0.28(2)		0.26(2)		0.272(8)	0.272(8)
83.36717(21)	0.80(8)	0.70(7)		0.69(7)	0.673(42)	0.69(3)		0.66(2)		0.71(4)		0.680(14)	0.680(14)
89.48595(22)	0.30(3)	0.23(7)		0.23(6)	0.218(26)	0.22(2)		0.29(2)		0.22(2)		0.245(14)	0.245(14)
97.43100(21)	100(5)	100	100	100	100	100	100(3)	100.0(15)	100	100	100.0(2)	100	100
103.18012(17)	73.5(10)	71.0(15)		71.1(15)	74.80(68)	69.6(10)	73.4(17)	73.7(12)		72.1(14)	73.7(4)	73.3(5)	73.3(5)
151.6245(12)	0.0130(13)	< 0.06	0.011(2)	0.31	0.060(15)	0.02(1)		< 0.01		0.021(1)			0.082(14)b
166.5548(15)			0.0010(10)										0.0010(10)
172.85307(21)	0.130(13)	0.10(10)	0.147(15)	0.28	0.144(26)	0.10(2)		0.13(1)		0.12(1)		0.128(5)	0.128(5)

[a] Calculated from $I_{ce}(LM) = 1.17$ and $\alpha(19,E2) = 3220$.
[b] Calculated from $I_\gamma(152):I_\gamma(68):I_\gamma(54) = 100(8):21(2):26(4)$ (adopted from Helmer, 2006) and $I_\gamma(21\,keV) = 0.075(12)$.

Taken from [6]

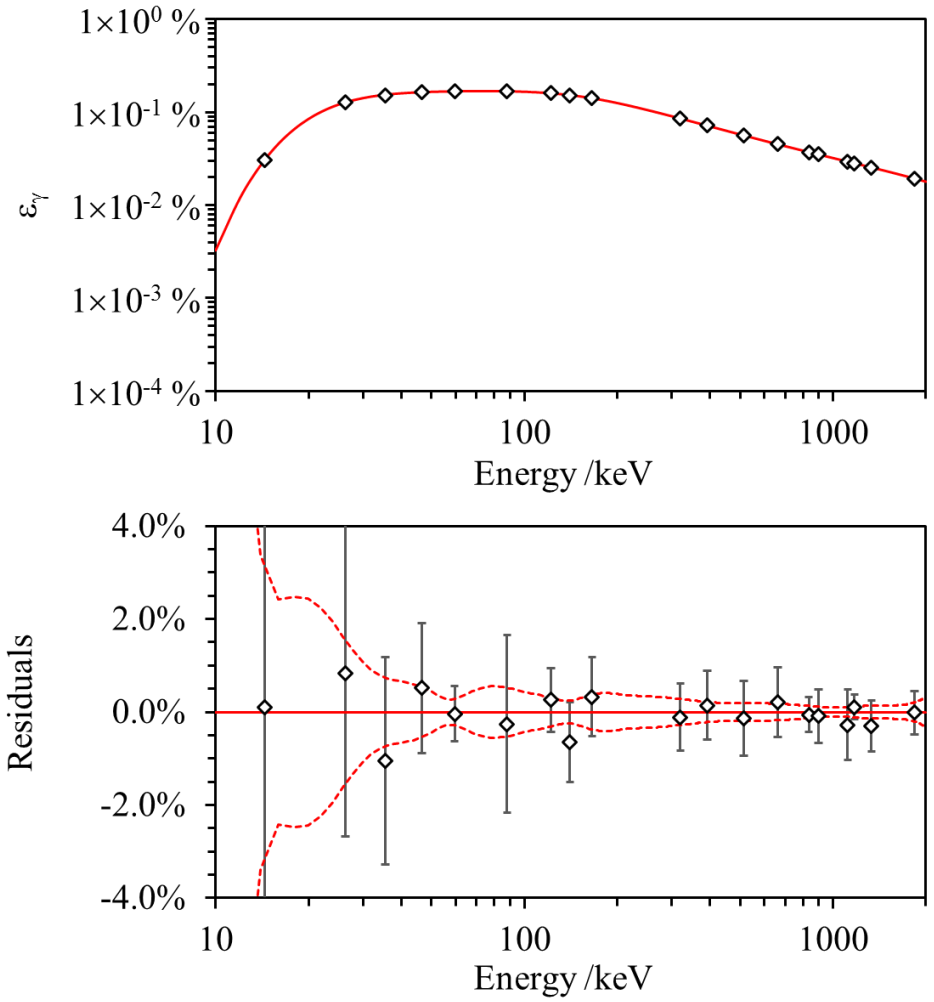
Experimental technique

- Three flame sealed 2ml BS ampoule sources of chemically pure ^{153}Gd were prepared by dispensing ~1g of ^{153}Gd from a 0.5M HCl solution.

- Two HpGe detectors (BART and LOKI) measured the three sources over a period of 5 years, for acquisition times between 86400 s and 250000 s.

- Spectra were fitted in Genie 2000.

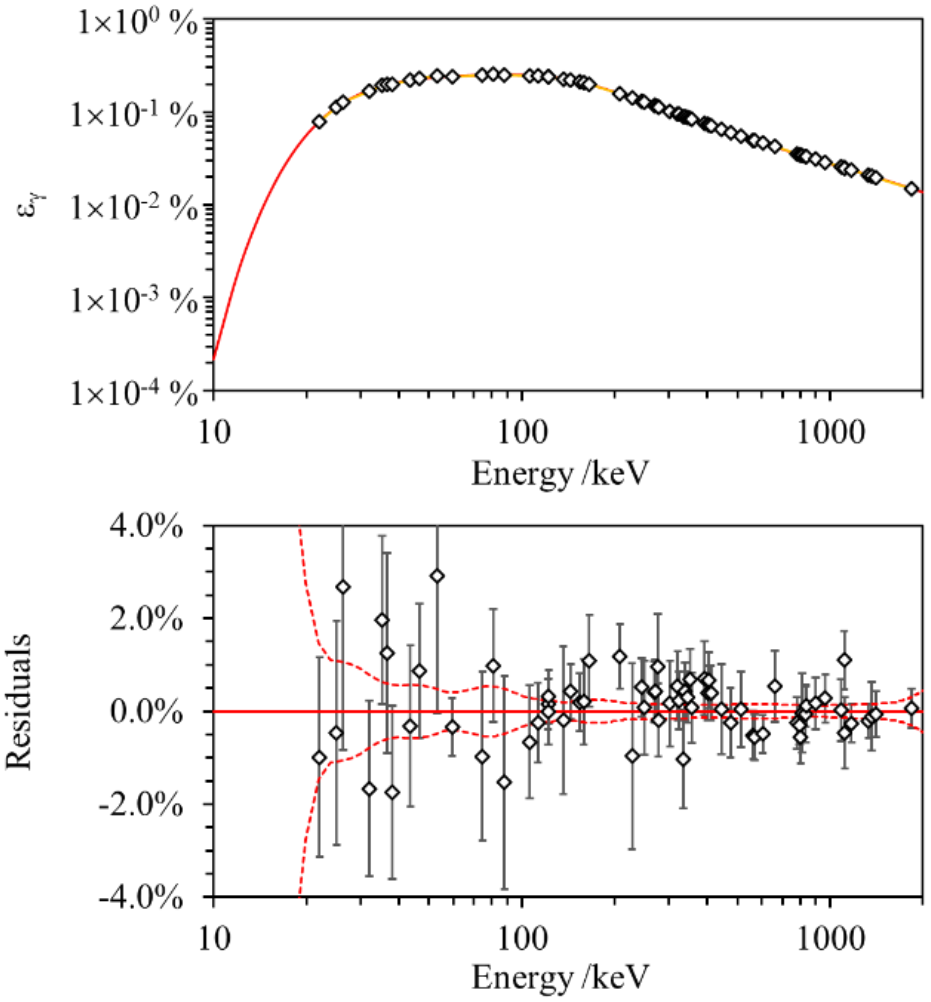
Calibration of the two HpGe detectors – BART



Specifications of BART

- ORTEC GMX detector, 28.9% relative efficiency
- 10 cm thick aged Pb walls covered in 0.5mm Cd and 0.7mm Cu.
- Optical bread boards mounted on the coffin to allow for highly reproducible source mounting.
- FWHM(1.33 MeV) = 1.79 keV
- Source distance to end cap = 250 mm

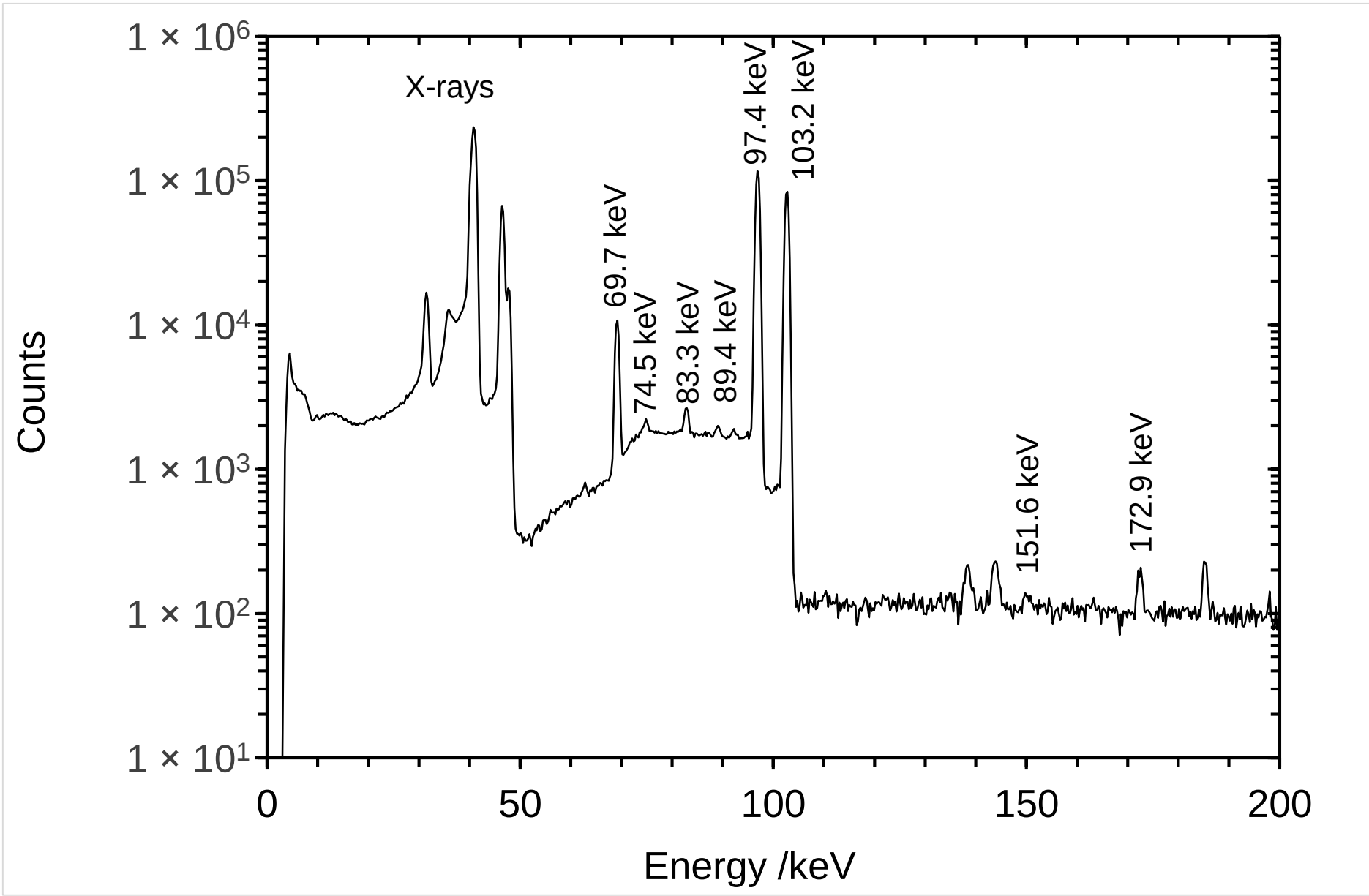
Calibration of the two HpGe detectors - LOKI



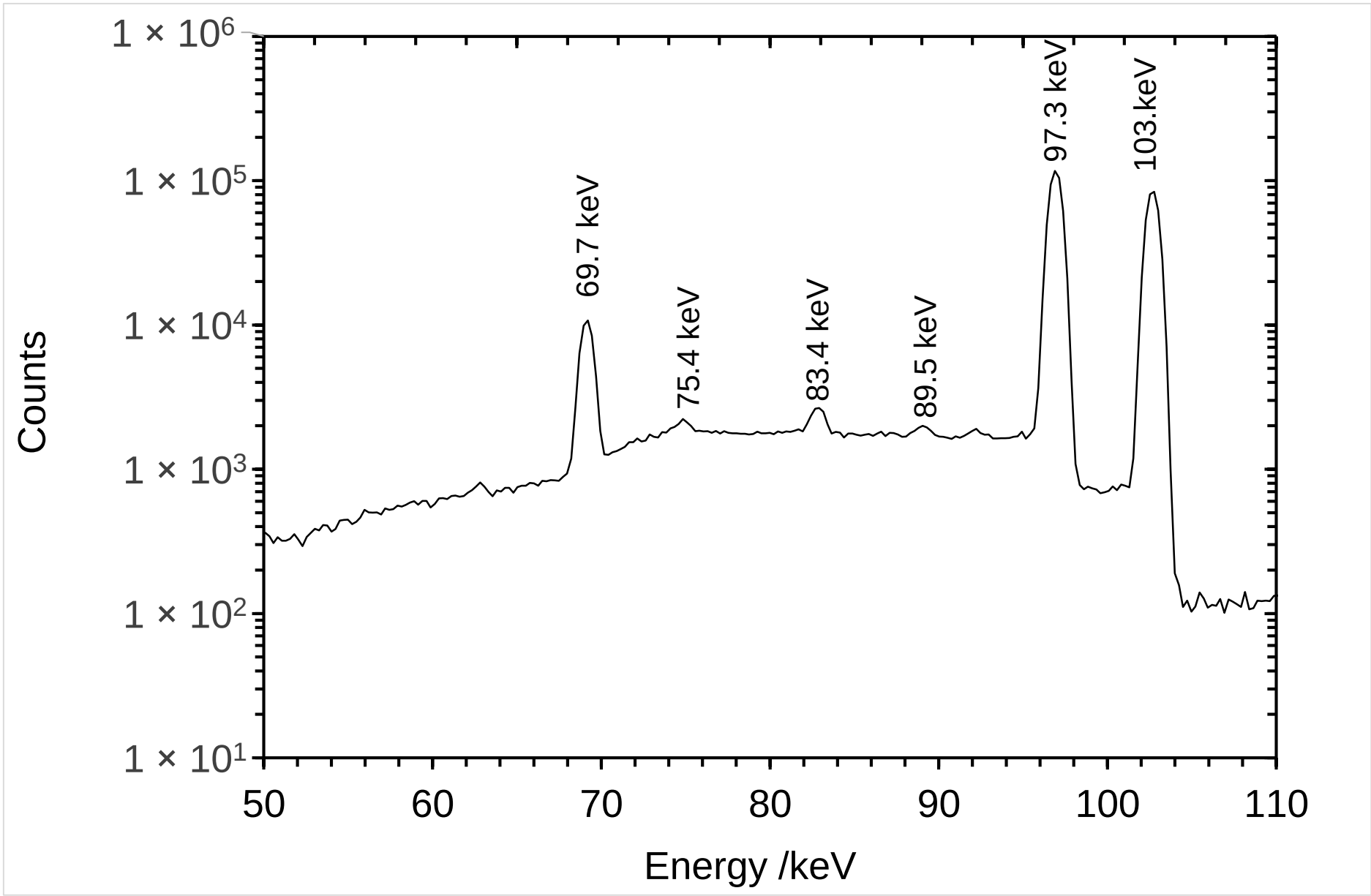
Specifications of LOKI

- GEM series ORTEC detector
- 10 cm thick aged Pb walls covered in 0.5mm Cd and 0.7mm Cu.
- Optical bread boards mounted on the coffin to allow for highly reproducible source mounting.
- FWHM(1.33 MeV) = 1.68 keV.
- Source distance to endcap = 290 mm.

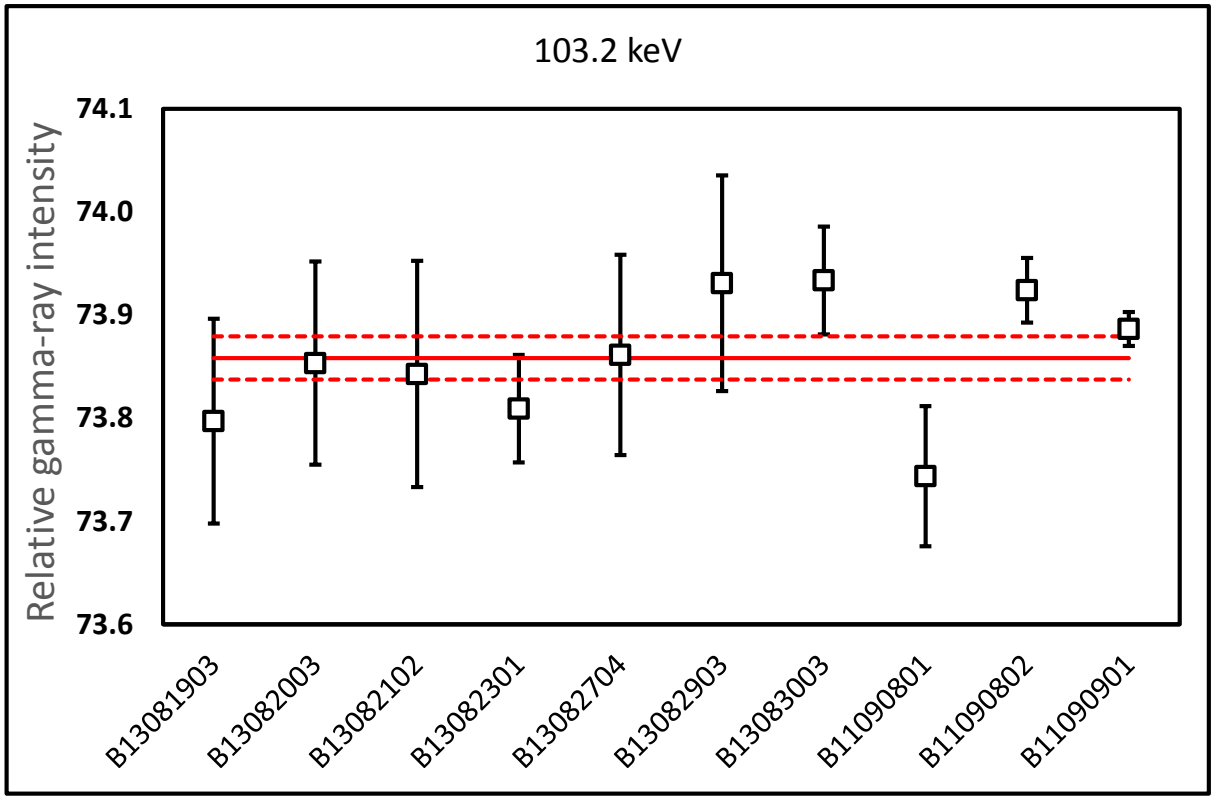
Experimental results – LK16071901



Experimental results – LK16071901



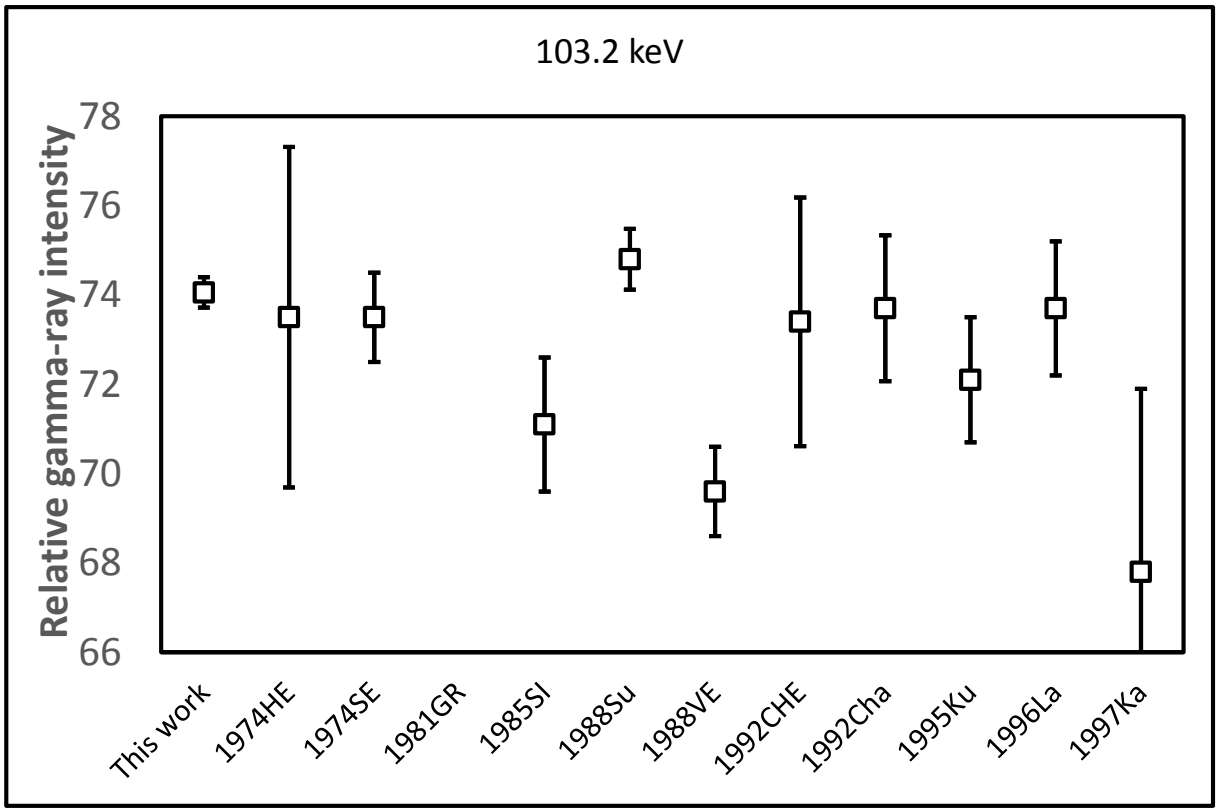
Experimental results



Energy /keV	Mean	Chi²
69.6	7.8455	0.09
89.46	0.3249	0.61
97.43	100.0	
103.18	73.98	0.11
151.62	0.0157	0.27
172.85	0.1312	0.12

Measurements for BART can be seen with good fidelity, corroborated by very acceptable reduced χ^2 for reported intensities.

Experimental results – Comparison



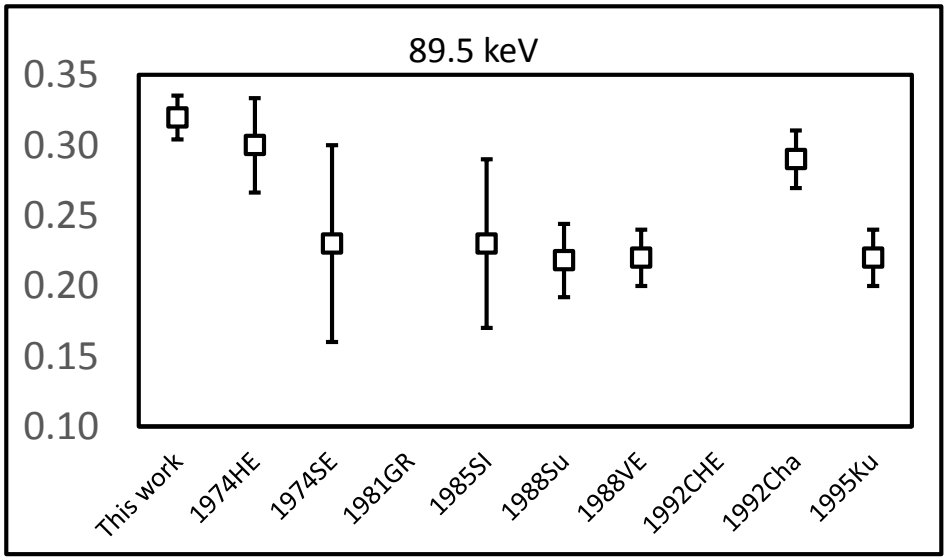
Results from the weighted mean of the two detectors intensity measurements show that there are large ($\sigma > 2$) from some of the previous work, however we consistently agree with work done by Chand et al.

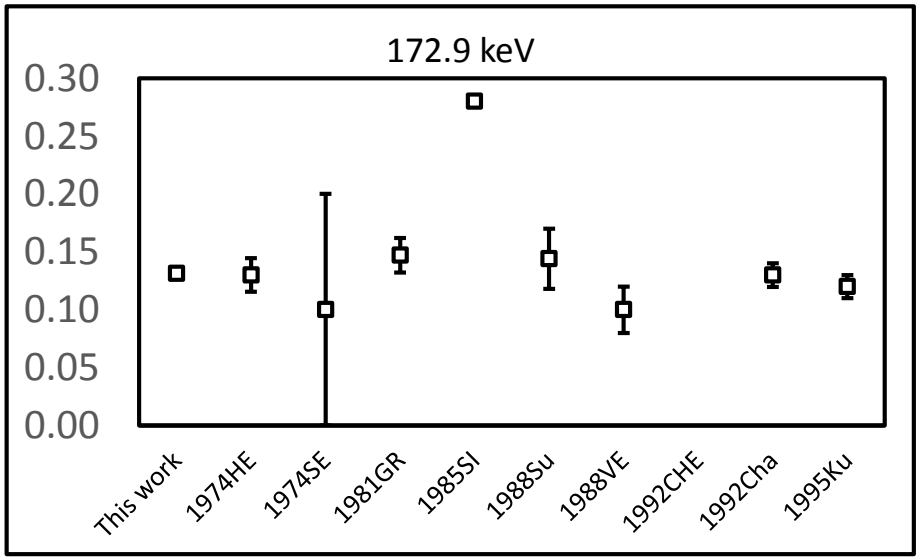
Many measurements have much smaller uncertainty than in previous work.

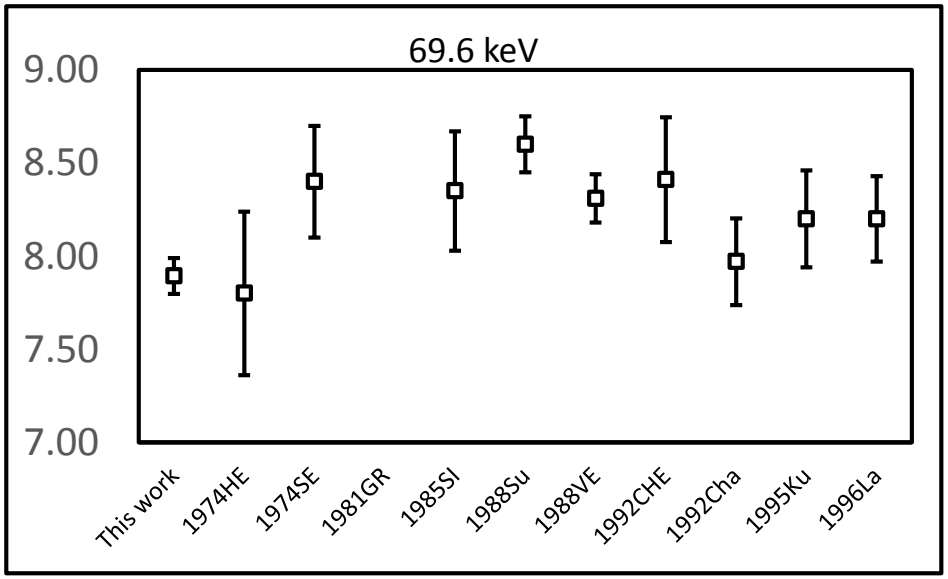
For 103.2 keV:

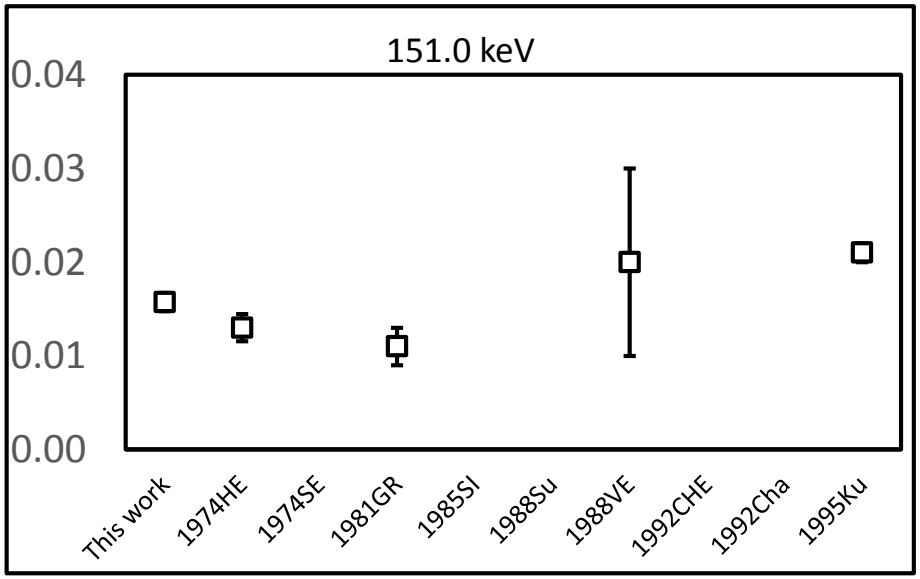
$u(I_{88SU})\% = 0.9\ \%$
$u(I_{NPl})\ \% = 0.5\ \%$

Experimental results – Comparison









Comparison with evaluated data

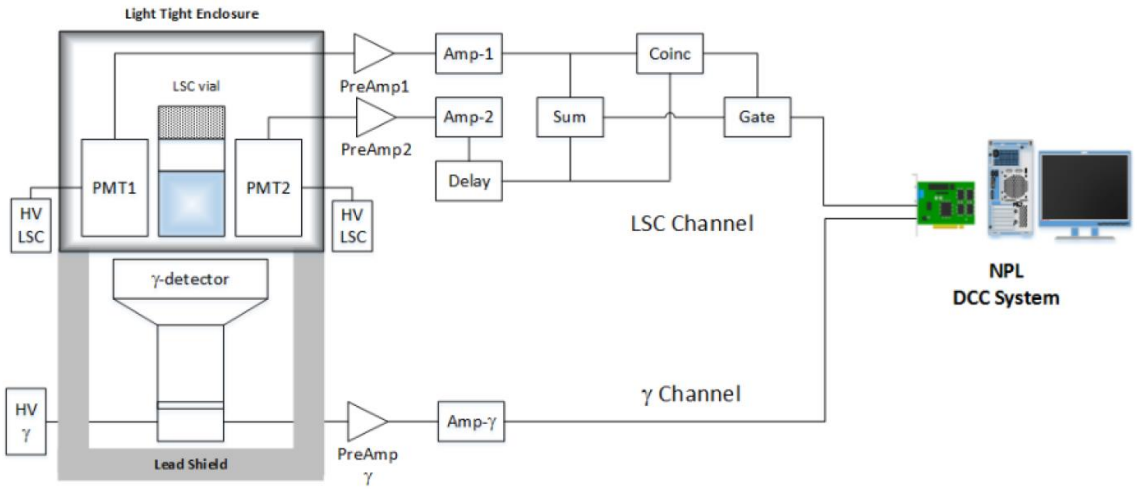
	H. Xiaolong (2010)	Helmer et al (2004)	NPL (this work)
69.67	**8.27 (8)**	**8.28 (10)**	**7.89 (9)**
89.46	**0.245 (14)**	**0.245 (14)**	**0.320 (16)**
97.43	**100**	**100**	**100**
103.18	**73.3 (5)**	**72.9 (7)**	**74.1 (3)**
151.62	**0.082 (14)**	**0.017 (4)**	**0.01573 (10)**
172.85	**0.128 (5)**	**0.125 (6)**	**0.1313 (18)**

Absolute intensity

Very preliminary DCC calculation of the source using $4\pi\beta$ liquid scintillation counter reported specific activity of **512.5 (25)** kBq g^{-1}@ 19/7/2011 12:00 UTC

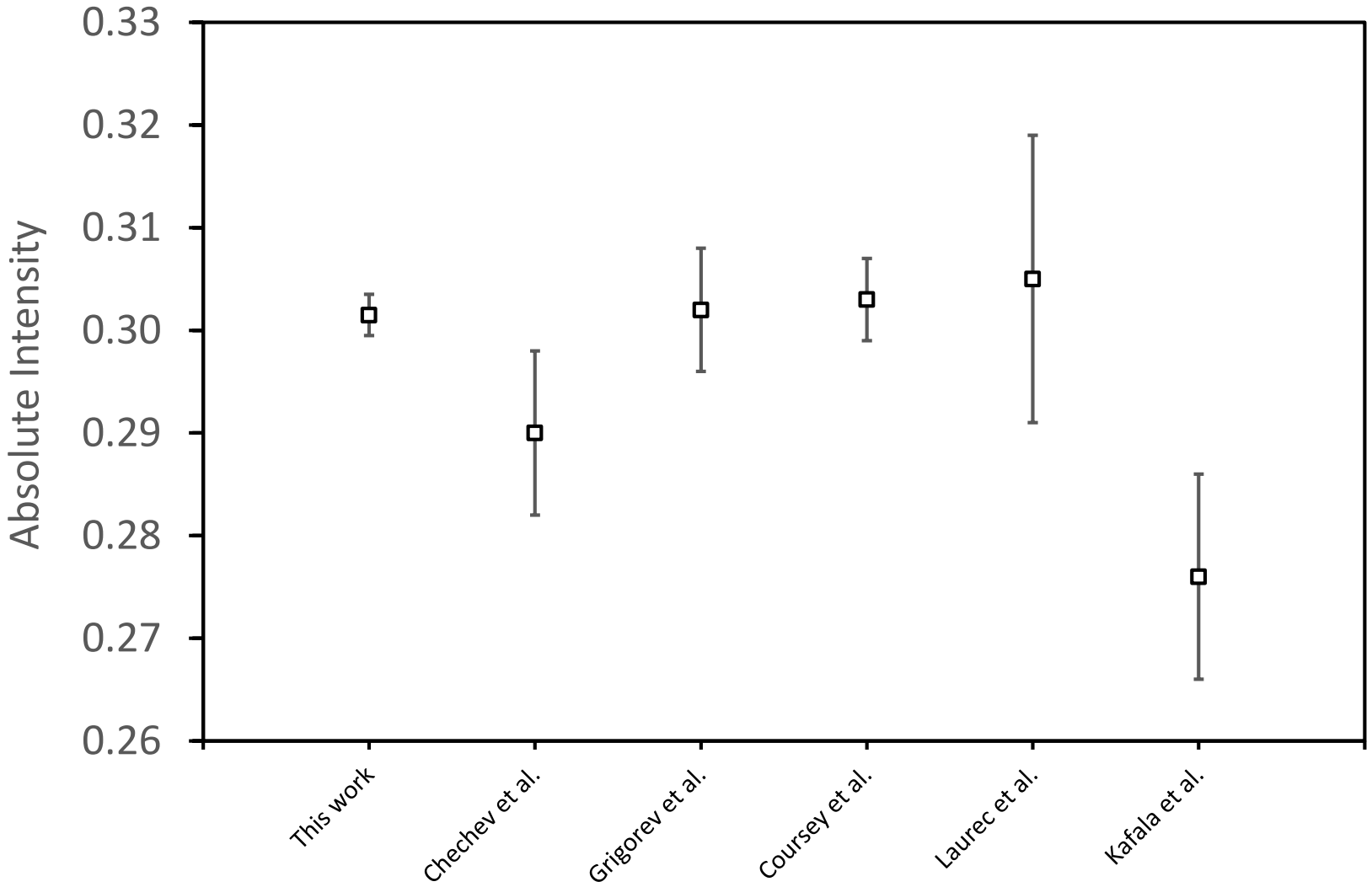
Using this value with it is possible to calculate the absolute intensity value for the 97.3 keV emission, according to the adopted value of the half life of 240.4(10) days.

By taking into account the well known efficiency and the density corrections required for the liquid source and the uncertainty in the mass dispensed, the rate across the early (first year) measurements were calculated and the (decay corrected) DCC value for specific activity used to find the absolute intensity.



The absolute intensity has been calculated to be **0.3015 (20)**

Absolute intensity comparison



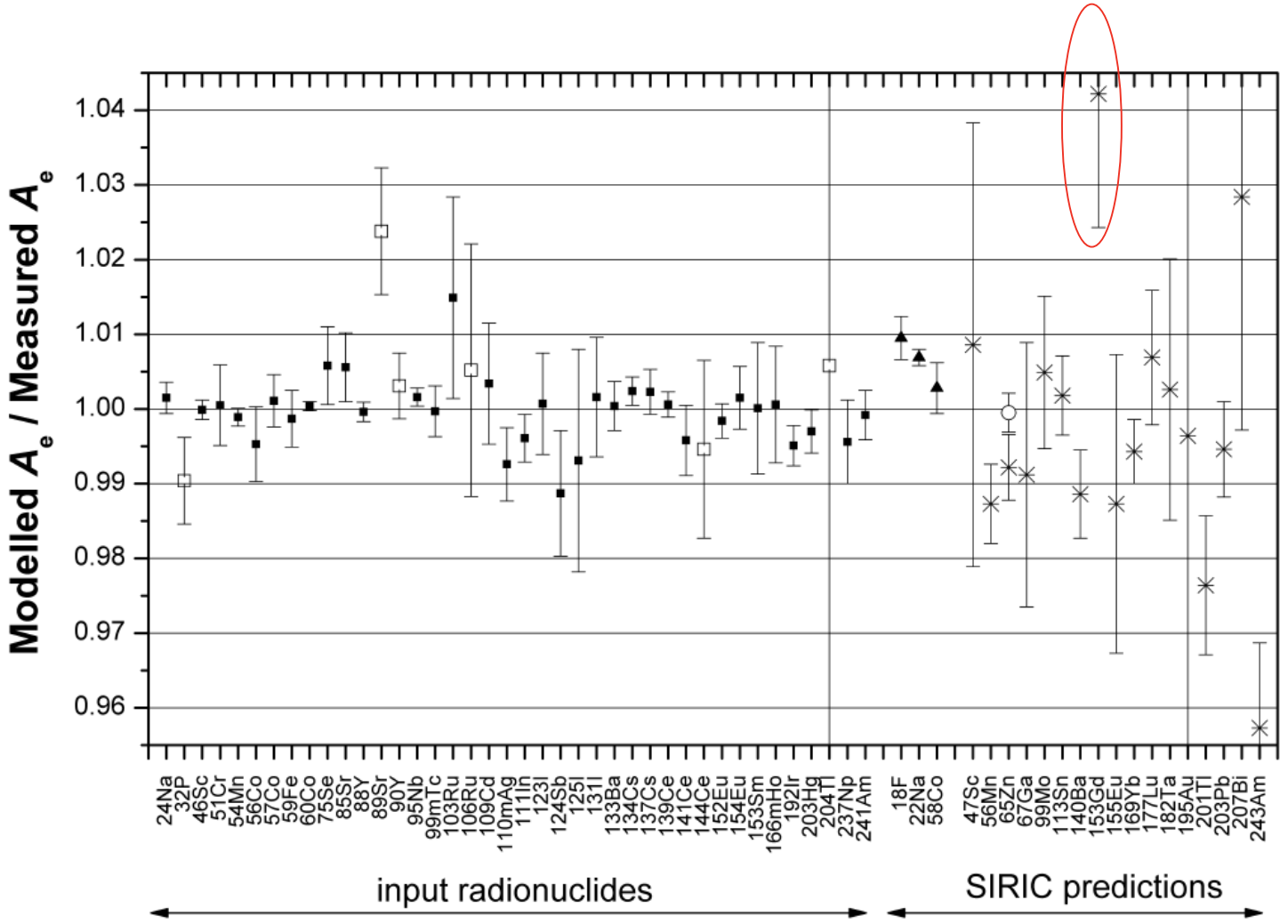
Absolute instensity Comparison remarks

- The adopted evaluated value for the absolute intensity of the 97.3 keV emission is 0.290 (8)

- The central value of the ***preliminary*** value from this work (0.3015 (20)) is 4% higher than the accepted value.

- This could be the reason for the discrepant value from the measured and the modelled value in SIRIC predictions.

Absolute Intensity Comparison remarks



Conclusion

- Present measurements disagree with the DDEP evaluated data.
- Measurements of the seen emissions have been observed with more experimental precision than previous studies.
- No evidence that the 269 keV level in ^{153}Eu is populated during this decay (no 96.8, 118.1 or 166.5 keV gamma ray identified)
- Previously reported gamma emissions in the purported level schemes could not be reliably identified in acquisitions (68.9 or 54.7 keV emission). 151 keV seen suggests 7/5- state is populated.
- Current liabilities within the detector set-up have caused the 75.42 keV and the 83.36 keV emissions to be unreliable and discarded.
- Absolute activity of the source measurement disagrees with current accepted absolute emission probability of the 97.43 keV γ ray.

Future work

- Acquire new material and perform new measurements.
- Perform complimentary half life study.
- Calculate the absolute activity of the new ^{153}Gd material.
- New evaluation of this decay.

Thank you

Department for Business, Energy & Industrial Strategy

FUNDED BY BEIS

References:

[1] R.C Hendel *et al*, J. Nucl. Med, **43(2)**, (2002)

[2] H. W. Wahner *et al*, Radiology,**156(1)**, (1986)

[3] J.E Pascente, (2000), http://www.ndt.net/article/wcndt00/papers/idn713/idn713.htm

[4] W. A. Engel *et al*, Phys Med Biol., **58(4)**, (2013)

[5] M. Be *et al* , Monographie BIPM-5 -Table of radionuclides, **2**, (2004)

[6] H. Xiaolong, Appl. Rad. Iso., **68,** (2010)

[7] M.G Cox et al, Monographie BIPM-7 (2007)

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Uncertainty budget (103.2 keV)

Component	Contribution
Statistical	0.37%
Fit	0.15%
Continuum	0.20%
Source attenuation	0.13%
True coincident summing	0.10%
Mass uncertainty	0.02%

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