Determination of Fractional Electron Capture Probabilities of ^{59}Ni by means of Metallic Magnetic Calorimeters

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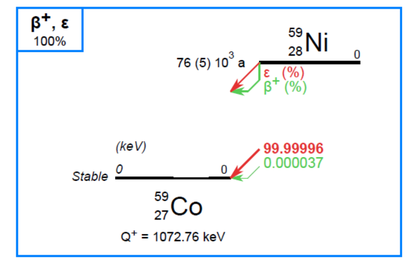
The measurement is part of the European metrology research project MetroMMC (2018-21). In the project, MMCs have been optimized and utilized for decay energy spectrometry of radionuclides decaying via electron capture (EC). Several EC-decaying radionuclides emitting radiation in the energy range between 20 eV and 100 keV were selected.

The project's main objectives are to measure fractional electron capture probabilities and absolute X-ray emission intensities. To improve theoretical models and calculations of electron capture and subsequent atomic relaxation and to validate the theoretical data with the high-precision experimental data from this project.

Motivation

Nickel-59, one of the radionuclides chosen for the MetroMMC project, decays mainly by EC to the ground state of Co-59 and has a long half-life of 76 000 years. The decay is a 2nd forbidden non-unique transition. The main applications where the electron capture probability ratios are required are:

- Isotope geology: ^{59}Ni is used to date meteorites and determine the abundance of extraterrestrial dust in ice and sediment.
- Nuclear power plant radioactive waste qualification: ^{59}Ni is produced in nuclear reactors as an activation product of Ni-cladded control rods and other steel structural components. It is present in nuclear waste, and its detection is very difficult because of its long half-life and no γ-ray emission.



^{59}Co shells E (keV)	
K	7.7089
L1	0.9256
L2	0.7936
L3	0.7786
M	0.1007-0.0029

Detection technique: Metallic Magnetic Calorimeters (MMCs)

- MMCs with the radionuclide embedded in the absorber → **Decay energy spectrometry (DES).**

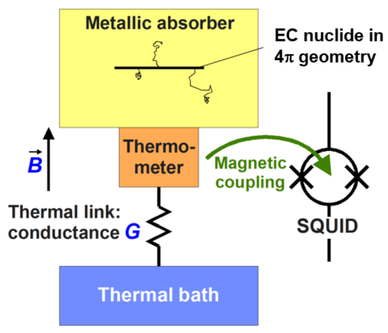
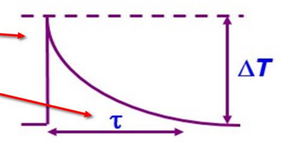


Figure 1: MMCs – basic idea.

- Signal : $\Delta T = E/C$
- Thermal link: $\tau = C/G$
- $C = C_{absorber} + C_{sensor}$



Particle	Absorber	Sensor	Flux Transformer	SQUID	Electronics at room temperature
Energy E	Temperature ΔT	Magnetization ΔM	Magnetic Flux $\Delta \Phi$	Readout of $\Delta \Phi$	ΔV

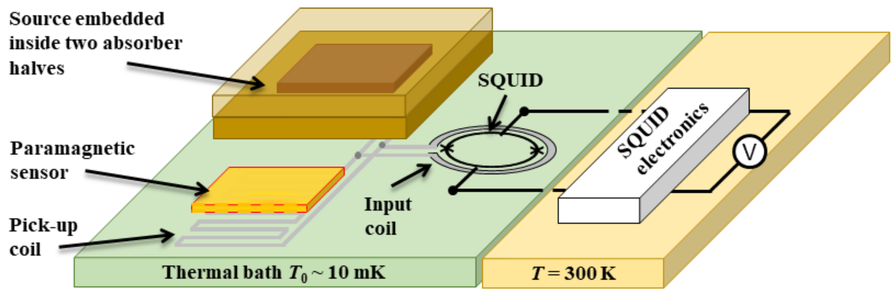


Figure 2: MMC readout by DC SQUID magnetometer via a superconducting flux transformer.

Absorber and source preparation

I. Electrodeposition method

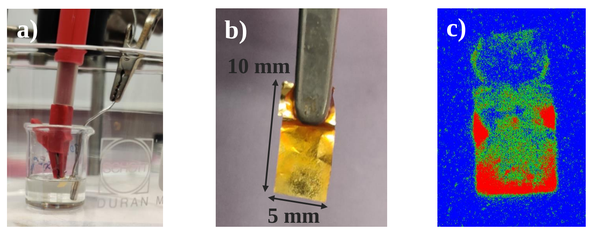


Figure 3: a) Electroplating setup b) Source foil after plating c) Autoradiography of the source foil.

- The electrodeposition using an in-house adapted two-electrode cell setup, as shown in *Figure 3a*.
- Anode: platinum wire and cathode: gold foil ($10 \times 5 \times 0.005$ mm^3).
- Electrolyte: 5 mL of DMSO & 100 µL of 1 N HCl + 0.1 N HNO_3 + $NiCl_2$.
- A constant current source at I = 10 mA and V = 150 V was used for 20 min.
- The initial mass activity of the radioactive solution is 81 kBq/g.
- Activity obtained on the source foil (*Figure 3b*): 2.77 kBq.
- *Figure 3c* shows the autoradiography of the activity distribution of the source.

II. Micro-drop deposition on gold nanofoam

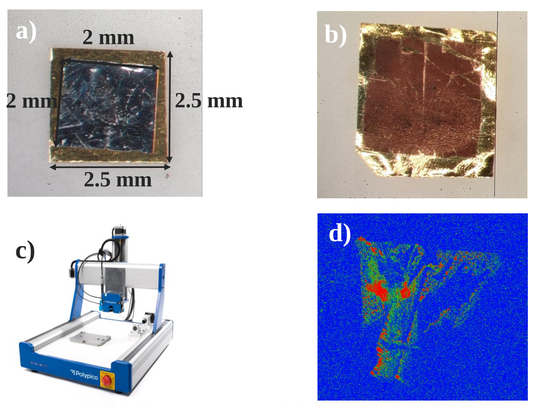


Figure 4: a) AuAg alloy on Au foil b) Micro-drop deposition on gold nanofoam c) Micro-drop dispenser d) Autoradiography of the source foil.

- *Figure 4a*: $Au_{0.3}Ag_{0.7}$ (wt%) alloy ($2 \times 2 \times 0.006$ mm^3) diffusion welded on gold foil ($2.5 \times 2.5 \times 0.01$ mm^3).
- *Figure 4b*: Gold nanofoam by dealloying through wet etching with 20% HNO_3 for 2 hours at 50 °C, dissolving silver, and forming nanoporous gold.
- *Figure 4c*: Micro-drop dispenser setup. For a 3 Bq activity, the drop volume was 41 nL (1216 drops); for 4 Bq, it was 55 nL (1671 drops).
- *Figure 4d*: Autoradiography of one of the sources.

Experiment setup:

Decay energy spectrometry (DES)

- In DES measurement mode, the source is fully enclosed between the two absorber halves (*Figure 5a*) to have a 4π detection geometry (*Figure 5b*).
- The decay energy from the source, except the neutrino, is thermalized inside the absorber. The obtained energy spectrum shows a single peak for each capture electron shell.

Figure 5: a) Source foil on one half of Au absorber b) Complete absorber-source ensemble.

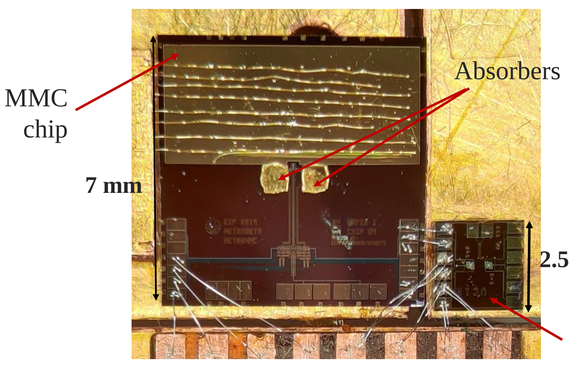


Figure 6: MMC detector setup.

Spectrum analysis and results

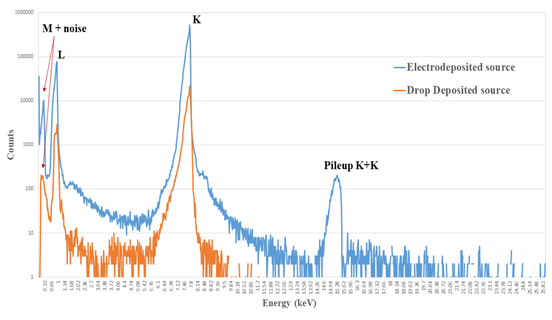


Figure 7: Comparison of both source's total energy spectra of ^{59}Ni.

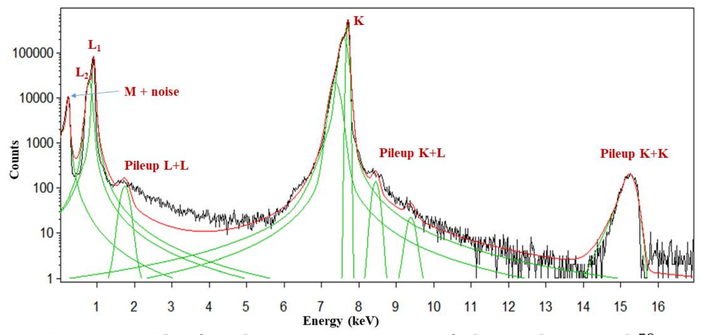


Figure 8: The fitted energy spectrum of electrodeposited ^{59}Ni.

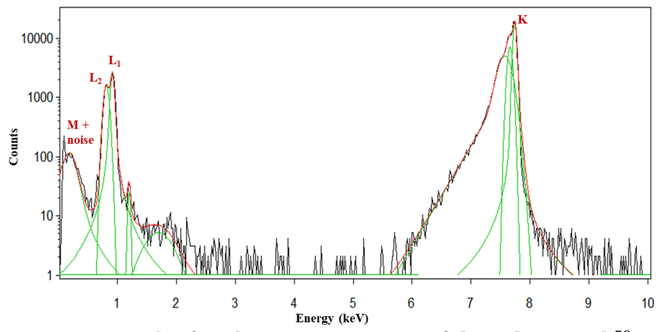


Figure 9: The fitted energy spectrum of drop deposited ^{59}Ni.

- Fractional electron capture probability ratios

Measurement	P_L/P_K
Electrodeposition	0.1154 (11)
Drop deposition	0.1122 (22)
BetaShape *[3]	0.1159 (26)
Literature	0.1210 (20) [4]

* Theoretical calculation code developed at LNHB.

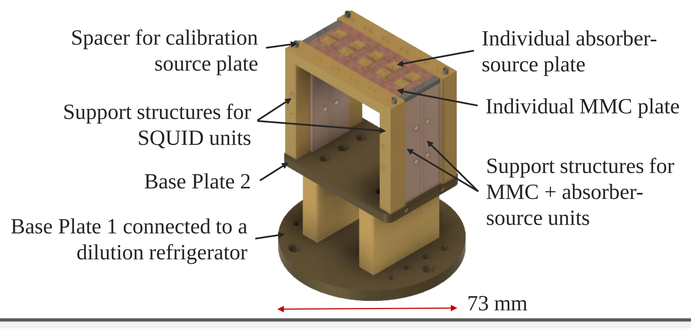
- The ratio of peak areas corresponds to the fractional capture probabilities. The quoted uncertainties comprise the statistical uncertainties and an estimation of the uncertainty due to the data analysis.
- The pulses had a 10-90% rise time of 170 µs in both sources, with exponential decay resulting in a 1/e decay time of 400 µs in the electroplated source and 440 µs in the drop-deposited source.
- In drop-deposited sources, due to the loss of a portion of the energy of Auger electrons in the source material causes a low energy tail (0.95% for K peak), less pronounced in electroplated sources (0.38% for K peak).

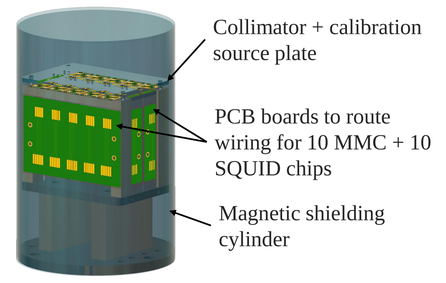
The fractional electron capture probability ratios from both sources are compatible with each other and the recent BetaShape calculation value.

The follow-up EMPIR project: PrimA-LTD (2021-2024)

- To develop a new primary method for decay scheme independent activity determination, high energy resolution, and statistics that exceed 10^8 events/spectrum.
- High statistics measurements of ^{129}I (beta spectrum shape; branching ratio) and ^{55}Fe (shake-up and shake-off effects) will be performed by the use of multiple detectors.

➢ Upgraded MMC spectrometer setup to acquire measurement statistics of >10^8 events per spectrum.





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