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# Determination of fractional electron capture probabilities of Nickel-59 by means of Metallic Magnetic Calorimeters

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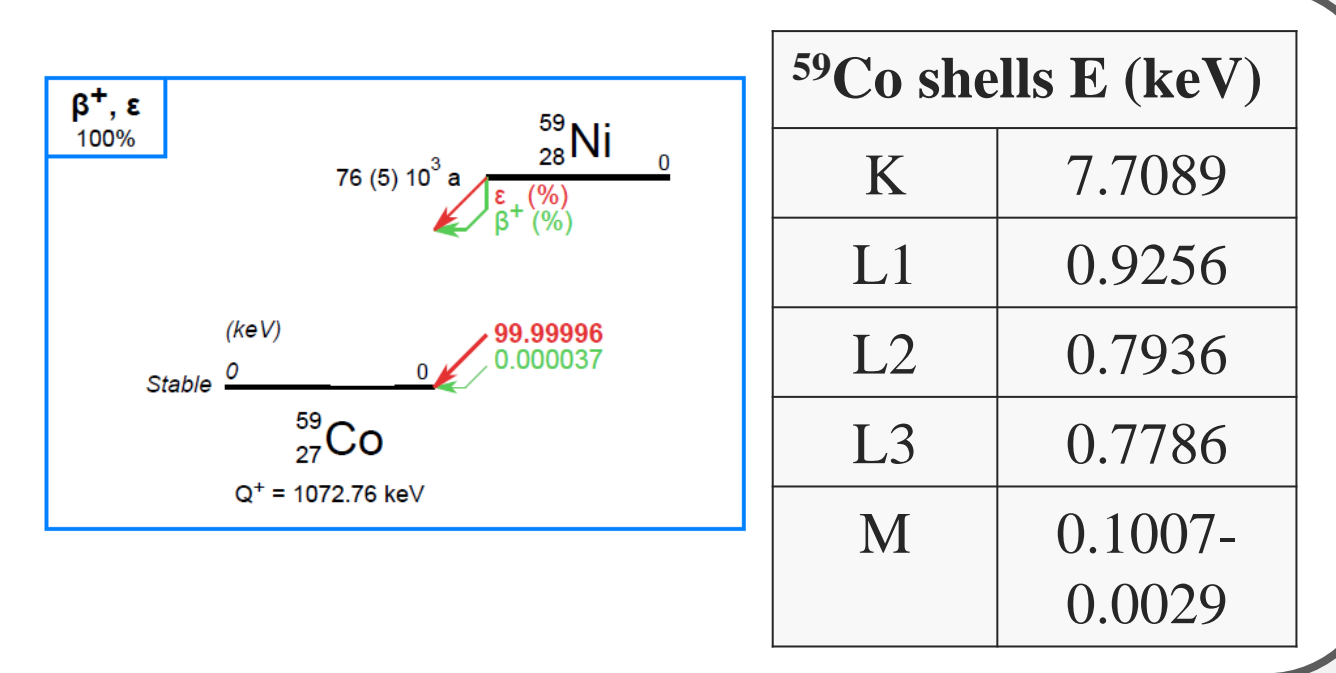
The measurement is part of the European metrology research project MetroMMC (2018-21). In the project, MMCs have been optimized and utilized for decay energy spectrometry of radionuclides decaying via electron capture (EC). Several EC-decaying radionuclides emitting radiation in the energy range between 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 2<sup>nd</sup> forbidden non-unique transition. The main applications where the electron capture probability ratios are required are:

- Isotope geology: <sup>59</sup>Ni is used to date meteorites and determine the abundance of extraterrestrial dust in ice and sediment.
- Nuclear power plant radioactive waste qualification: <sup>59</sup>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  $\gamma$ -ray emission.



## Detection technique: Metallic Magnetic Calorimeters (MMCs)

MMCs with the radionuclide embedded in the absorber  $\rightarrow$  Decay energy spectrometry (DES).

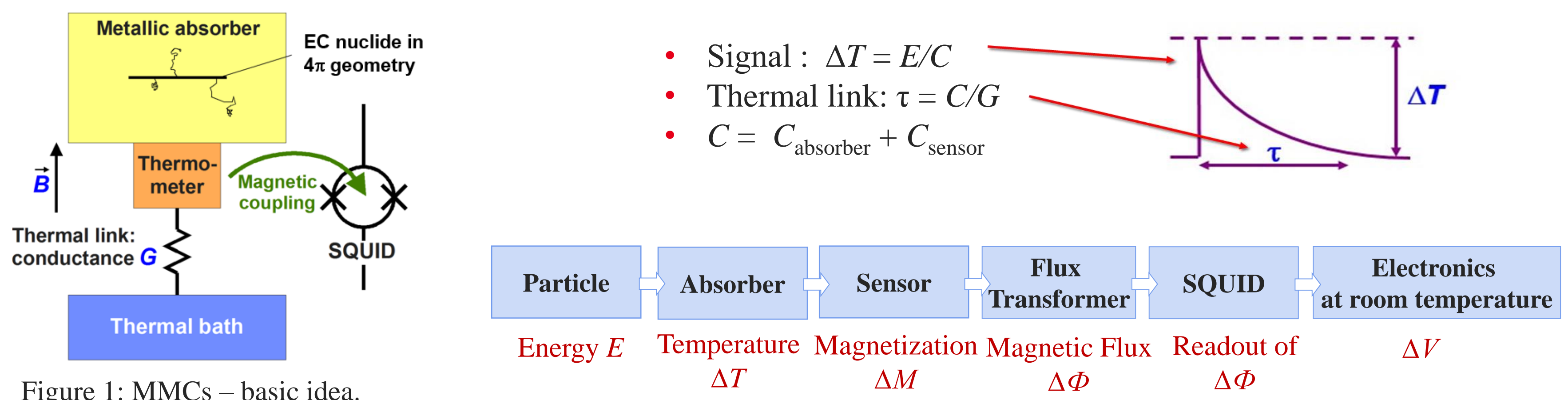


Figure 1: MMCs – basic idea.

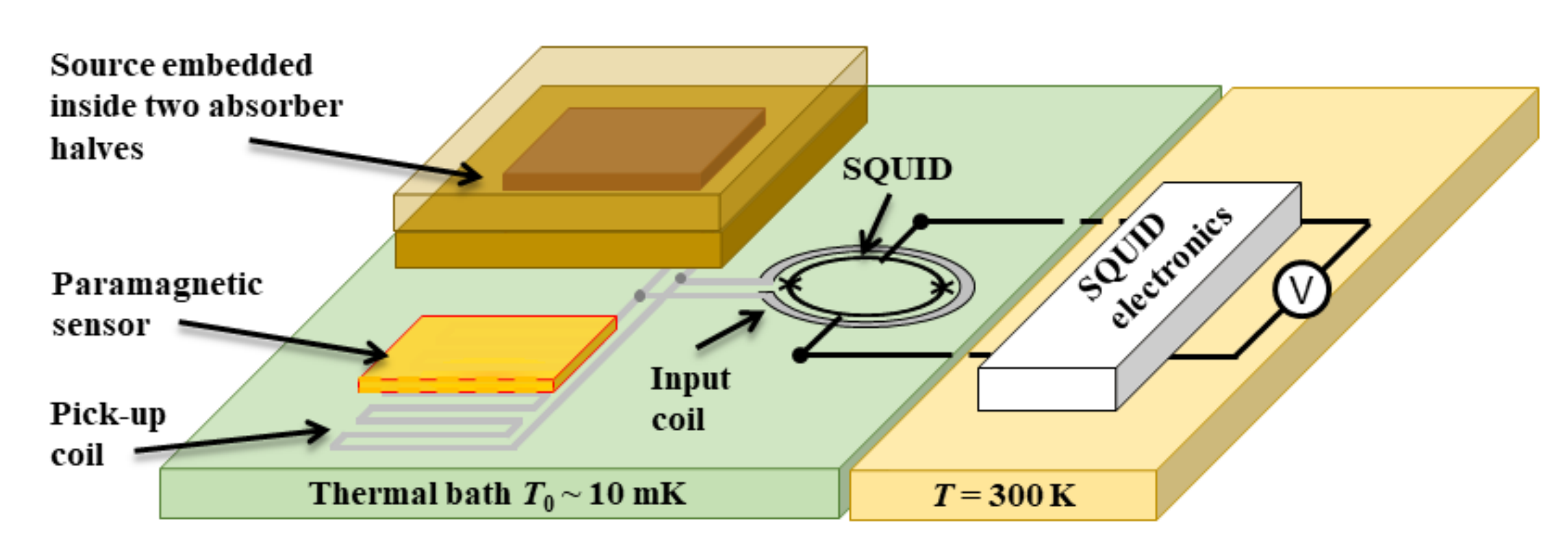


Figure 2: MMC readout by DC SQUID magnetometer via a superconducting flux transformer.

## Absorber and source preparation

### I. Electrodeposition Method

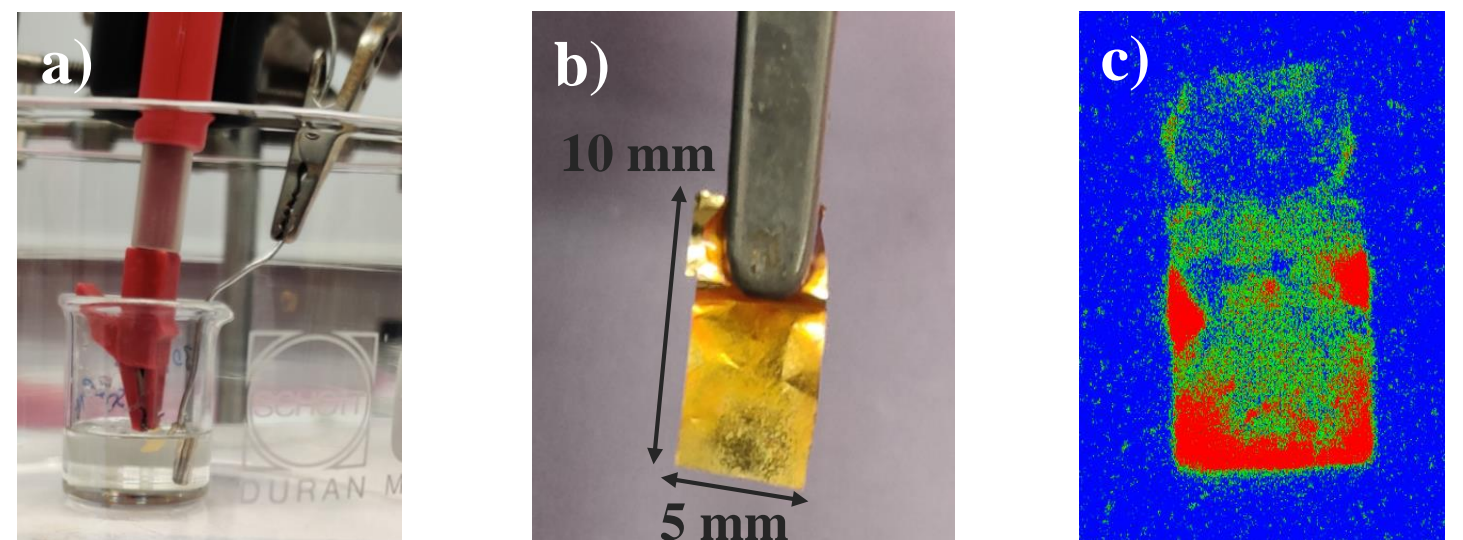


Figure 3: a) Electroplating setup b) Source foil after plating c) Autoradiography of the source foil.

### II. Micro-drop deposition on gold nanofoam

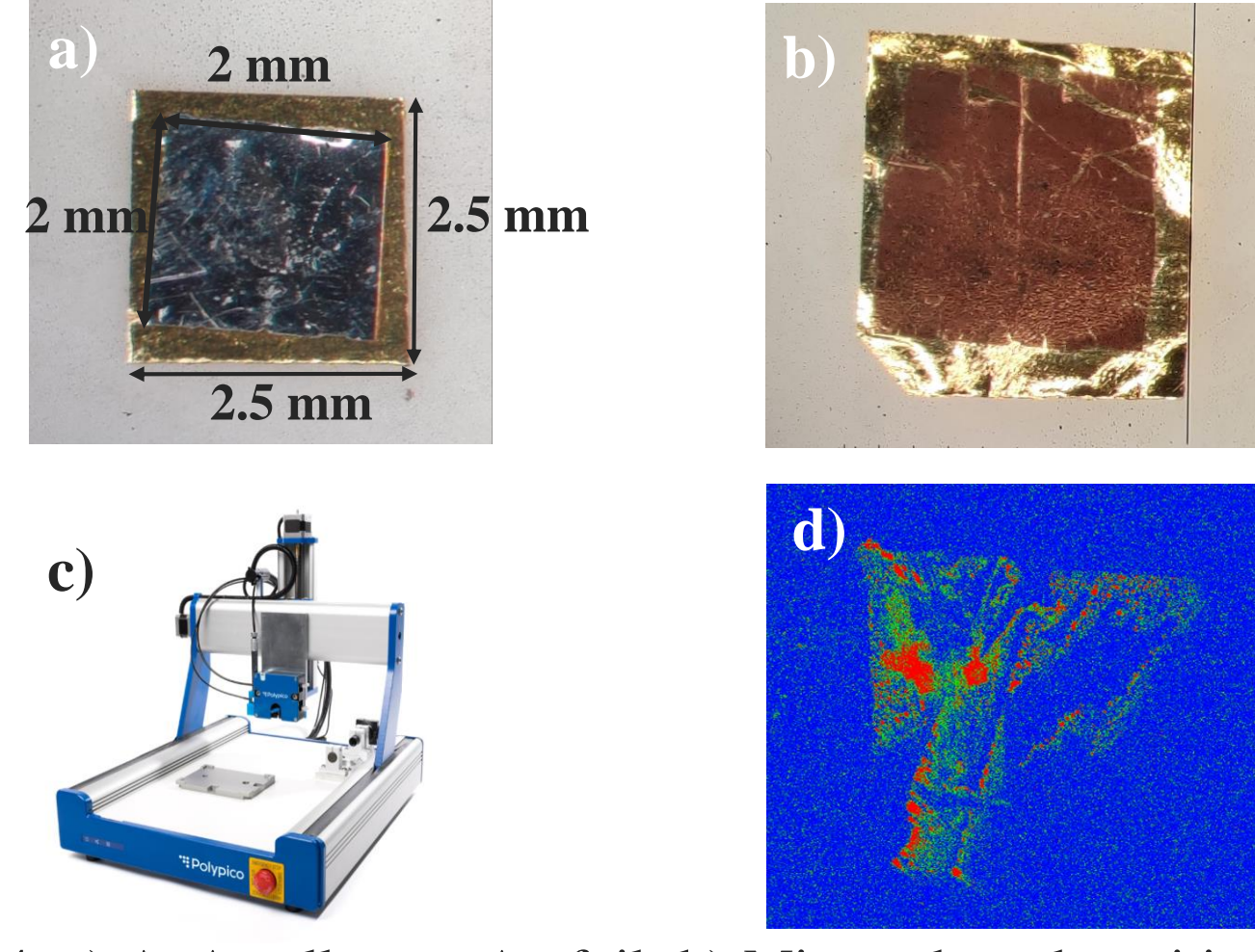


Figure 4: a) AuAg alloy on Au foil b) Micro-drop deposition on gold nanofoam c) Micro-drop dispenser d) Autoradiography of the source foil.

- The electrodeposition using an in-house adapted two-electrode cell setup, as shown in Figure 3a.
- Anode: platinum wire and cathode: gold foil (10x5x0.005 mm<sup>3</sup>).
- Electrolyte: 5 mL of DMSO & 100  $\mu$ L of 1 N HCl + 0.1 N HNO<sub>3</sub> + NiCl<sub>2</sub>.
- 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.

- Figure 4a: Au<sub>0.3</sub>Ag<sub>0.7</sub> (wt%) alloy (2x2x0.006 mm<sup>3</sup>) diffusion welded on gold foil (2.5x2.5x0.01 mm<sup>3</sup>).
- Figure 4b: Gold nanofoam by dealloying through wet etching with 20% HNO<sub>3</sub> 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 4 $\pi$  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.

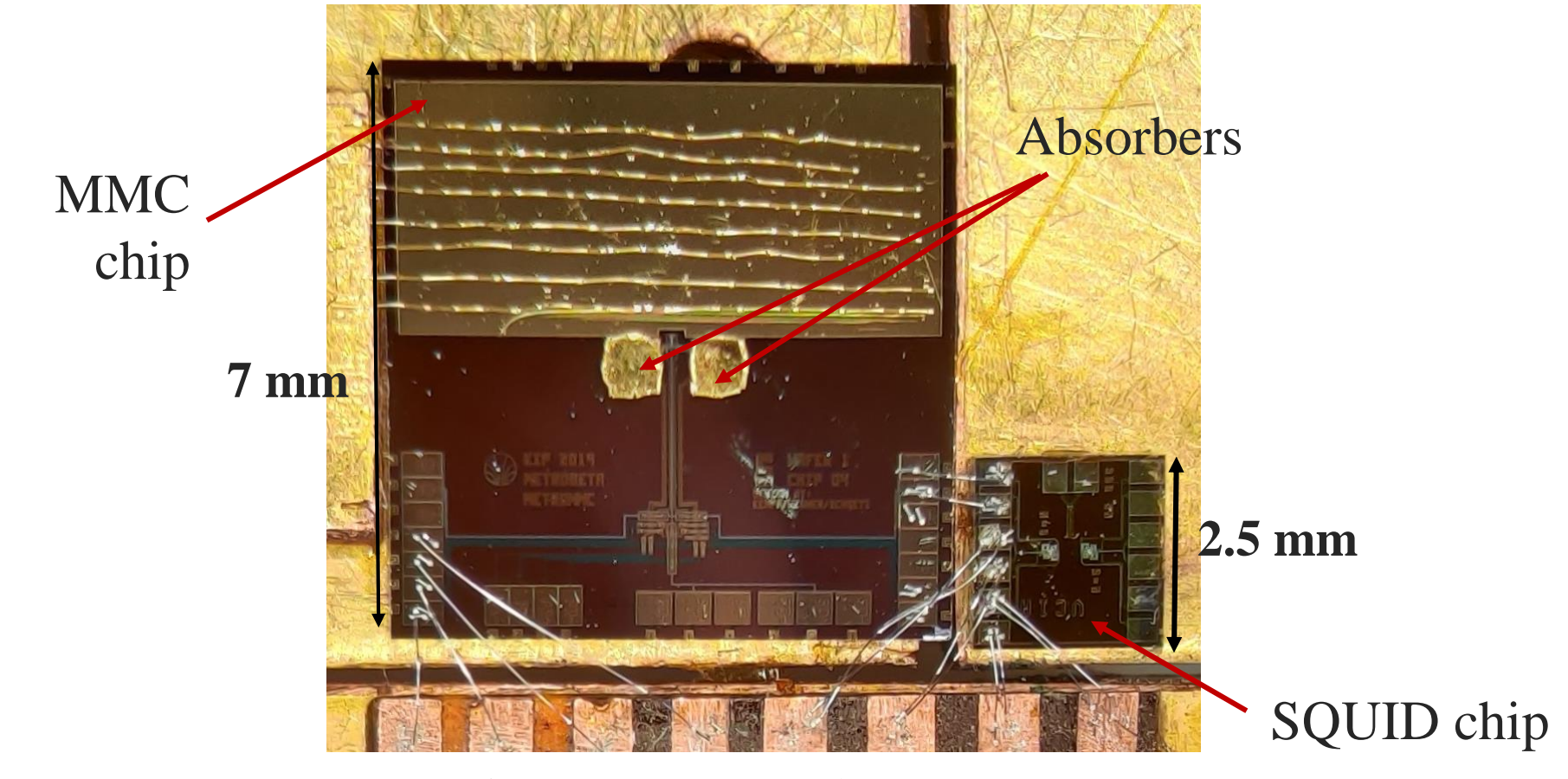


Figure 6: MMC detector setup.

## Spectrum analysis and results

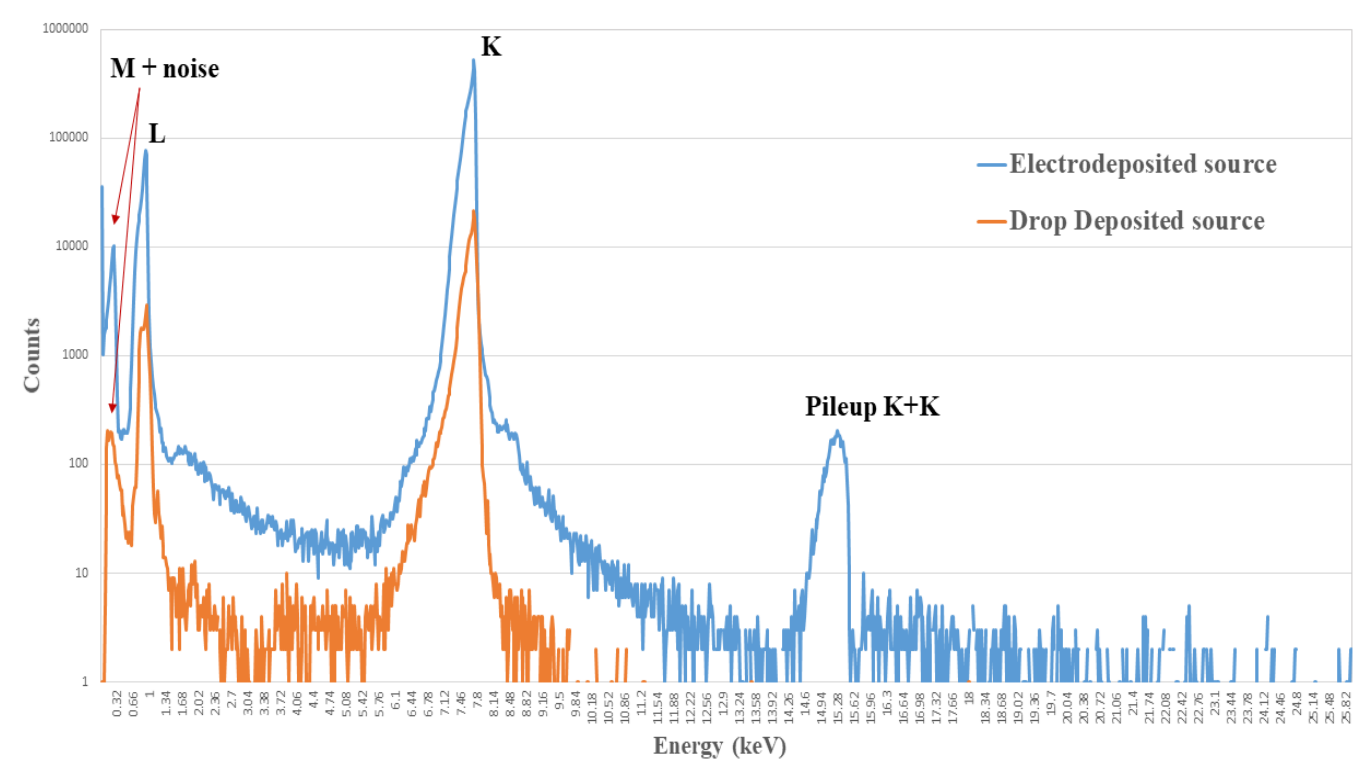


Figure 7: Comparison of both source's total energy spectra of <sup>59</sup>Ni.

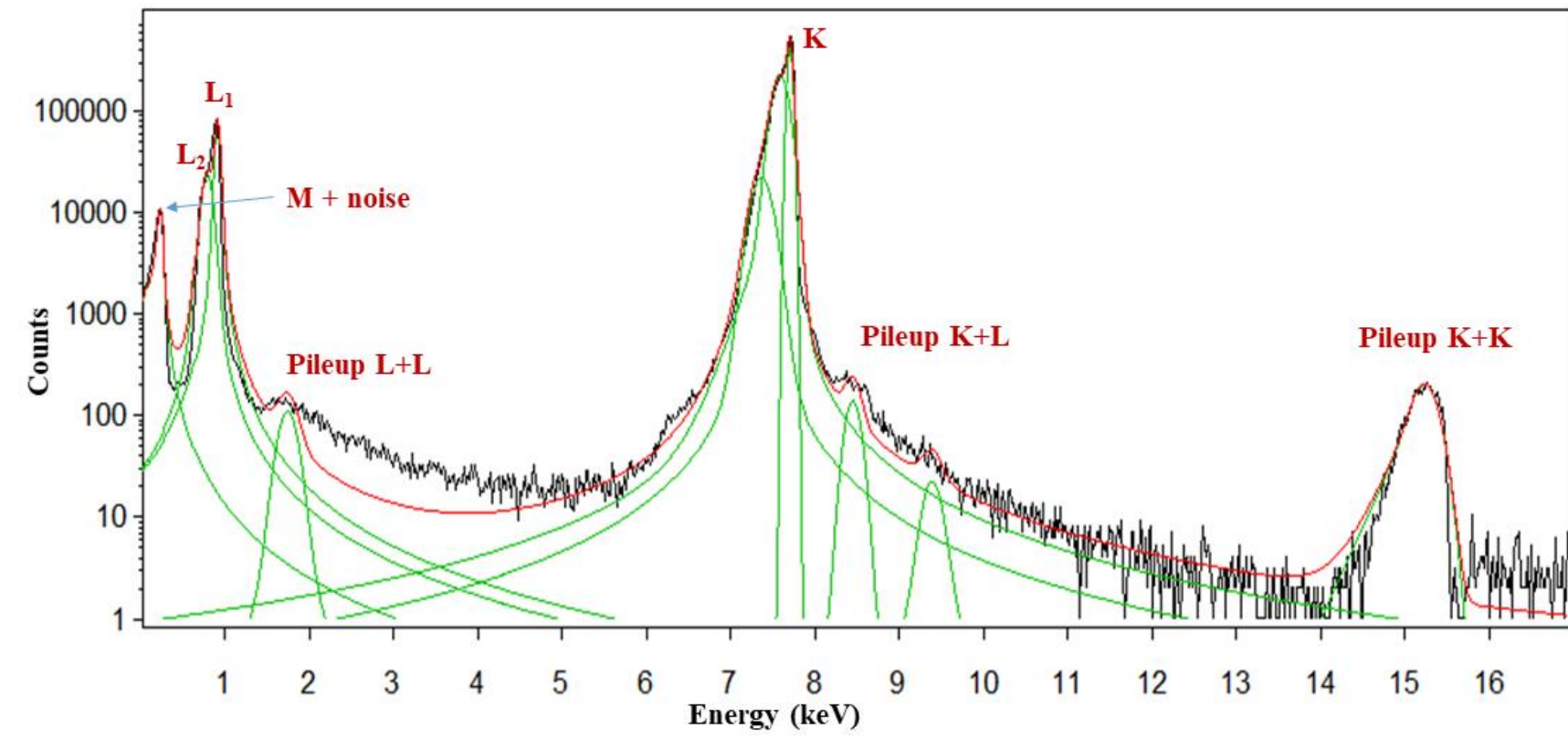


Figure 8: The fitted energy spectrum of electrodeposited <sup>59</sup>Ni.

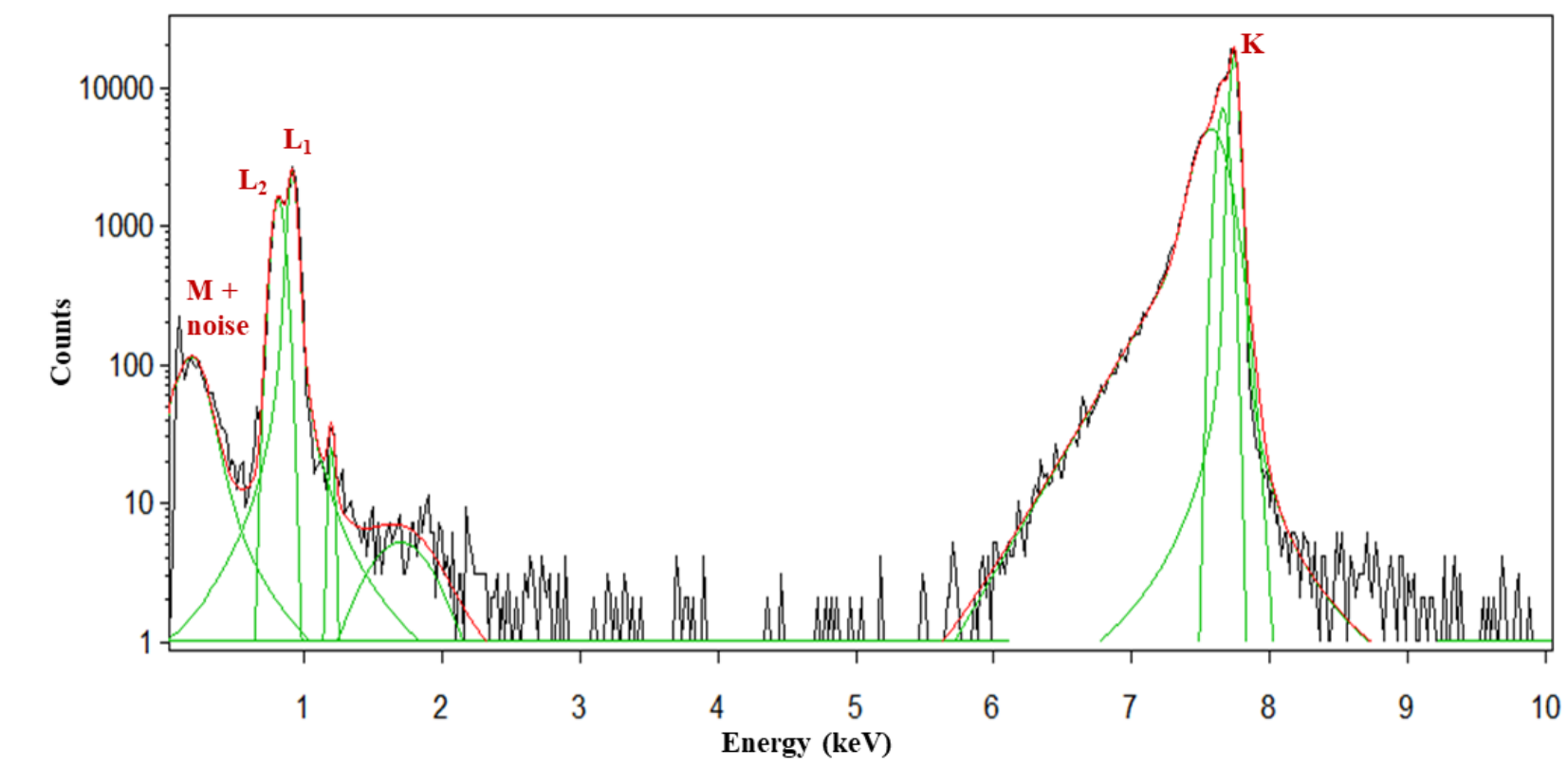


Figure 9: The fitted energy spectrum of drop deposited <sup>59</sup>Ni.

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  $\mu$ s in both sources, with exponential decay resulting in a 1/e decay time of 400  $\mu$ s in the electroplated source and 440  $\mu$ s 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<sup>8</sup> events/spectrum.
- High statistics measurements of <sup>129</sup>I (beta spectrum shape; branching ratio) and <sup>55</sup>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<sup>8</sup> events per spectrum.

