The PrimA-LTD Project

Towards New Primary Activity Standardisation Methods Based on Low-temperature Detectors

Frederic Juget

Vaud University Hospital Center (CHUV) Institut de Radiophysique, Lausanne, Switzerland

E-mail: Frederic.Juget@chuv.ch

Metallic microcalorimeters (MMCs) are detectors of radiation or particles that are operated at very low (< 0.1 K) temperatures. Over the last two decades, MMC detectors have been developed for energy dispersive spectrometry in the X-ray and gamma ray energy range with exceptionally high resolution. They are an enabling technology for precision experiments in particle physics [1,2], and their potential for applications ranging from nuclear forensics to X-ray astronomy is being explored actively.

MMCs measure the temperature rise of a metallic absorber, usually gold, that occurs when energy is deposited in the absorber. The device comprises the absorber element and paramagnetic metallic sensor that are weakly thermally coupled to a heat bath. The paramagnetic sensor is exposed to a weak magnetic field to create a temperature dependent sensor magnetization. Following the calorimetric detection principle, a temperature rise causes a change in the magnetic susceptibility of the paramagnetic material, e.g. Ag:Er, which leads to a change in the sensor magnetization. This change in magnetization can be precisely measured using a superconducting quantum interference device (SQUID) sensor.

MMCs have been employed for radioactive decay energy spectrometry (DES). Here, the radioactive source is typically enclosed by the absorber which is designed, in terms of its dimensions, to realise a 4p detection geometry, i.e. to achieve a full absorption of the energy of the emitted particles. The EMPIR project PrimA-LTD, “Towards new primary activity standardisation methods based on low-temperature detectors,” aimed to develop a new primary decay scheme-independent activity standardization method using MMCs and to improve on fundamental decay data. The project involves 5 European countries (France, Germany, Portugal, Spain, and Switzerland) and has been active for 3 years until mid-2024. Two MMC spectrometers are operated by the project partners, one at the Laboratoire National Henri Becquerel (LNHB) of the Commissariat à l’Energie Atomique (France) and the second one at the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig (Germany). Given achievable detection efficiencies close to 100% in ideal conditions, high energy resolution and linearity as well as low background, MMCs are close to ideal detectors for standardization in radionuclide metrology. Their use is mainly limited by their comparatively slow recovery time limiting the acceptable activity per detector element to a few Becquerel.

Within the PrimA-LTD project, new MMC detector types have been developed enhancing the flexibility of MMC measurements. Typically, MMCs have been used for single measurements only by gluing a composite source-absorber element to the detector pixel [3,4]. The new and innovative detector design developed in the project now allows for thermal coupling of external source-absorber elements to the detector via gold wirebonds (Fig. 1). Thus, the absorber can be easily replaced and the detector be reused. Two reusable detector variants have been designed [5]. One, named RoS-L, is optimized to measure the energy spectrum of the ^{241}Am alpha decay and ^{129}I beta decay. The second variant, named RoS-M, is optimized for measurements of electron capture nuclides such as ^{55}Fe. The RoS detectors are designed to match external absorbers having a heat capacity of 400 pJ/K (RoS-L) and 110 pJ/K (RoS-M) at a temperature of T = 20mK.

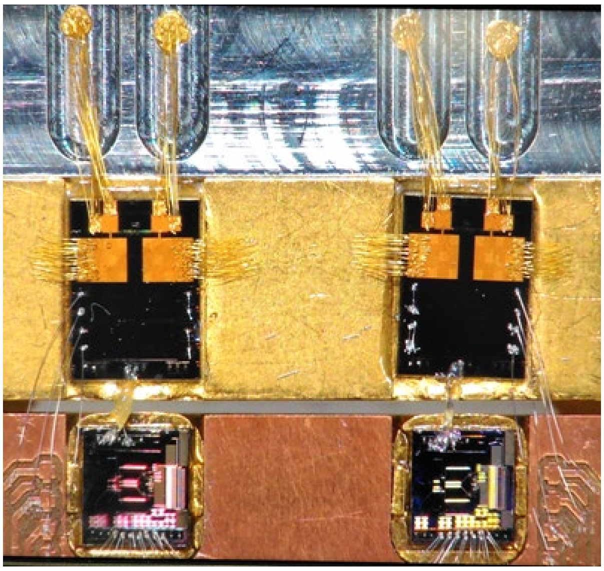

Fig. 1: Measurement configuration with reusable MMC detectors. External source-absorber elements (top) are thermally connected to the RoS-type MMC detectors (middle) via gold bond wires. The MMCs are read out by SQUID current sensors (bottom).

Both variants have been configured to be fabricated on the same Si wafer. Each detector chip has a size of (4 × 5) mm^2 and contains one two-pixel detector of one of the variants, which can be read out by a single SQUID. The pixel size is (1120 × 1070) µm^2 with an area of the Ag:Er$_{475\text{ppm}}$ sensor of about 1 mm^2. The RoS-M variant has a smaller pixel size of (490 × 500) µm^2 with no bond pad in its center. However, the pixels are equipped with wire bond pads at their edges. From numerical simulations, the theoretical energy resolution at 20 mK is ΔE_{FWHM} = 38.5 eV and ΔE_{FWHM} = 19.5 eV for the RoS-L and RoS-M variants, respectively.

The source preparation (absorber and radioactive material) is fundamental for precise measurements with MMCs. Special care must be taken to ensure that the entire energy of each decay is deposited and thermalized in the absorber. A commonly used technique of source preparation for MMC-based decay energy spectrometry is drop-deposition. Manual drop deposition often leads to the formation of large (on the order of a few micrometres) salt crystals which can cause considerable spectrum distortions due to incomplete thermalization.

This issue can be minimized by automatic microdrop deposition in a 2D array of very small individual droplets. It is achieved using commercial micro-dispensing systems depositing single droplet volumes of less than 50 pL in combination with a placement accuracy of better than 20 µm. Another source preparation technique employed in the project involved nano-structured absorber surfaces to reduce spectrum artefacts. An absorber of silver nanofoam was developed to be used for autodeposition of ^{129}I [6].

Employing the RoS detectors, measurements concerning electron capture probability ratios, X-ray, gamma, and electron spectrometry have been performed within the PrimA-LTD project. The L X-ray spectra from ^{241}Am and ^{210}Pb decays show a full-width half-maximum (FWHM) energy resolution of 26 eV and a constant detection efficiency between 5 and 26 keV, allowing for the determination of about 30 relative L X-ray intensities for ^{241}Am and ^{210}Pb.

A further objective of the PrimA-LTD project was to combine source preparation by radioactive ion implantation and optimized MMC detectors to obtain spectra of highest energy resolution. A novel multi-pixel MMC detector [5] to detect the low energy photons ($E < 6.5$ keV) emitted by the electron capture decay of ^{55}Fe ion-implanted into the absorber has been developed. A gold absorber thickness of 12 µm was estimated by Monte Carlo simulations to achieve a detection efficiency of larger than 99.99% in the energy range of interest. To fully integrate a gold absorber of 2× 12 µm thickness into the MMC detector, an optimized electro-deposition process has been developed. An area of 136 × 136 µm^2 is designated to be ion implanted in the integrated absorber elements with lateral size of 170 × 170 µm^2. The total heat capacity of the absorber amounts to 1 pJ/K at $T = 20$ mK. Laser ion resonance excitation and a mass separation to select only the wanted isotope to have a pure source was adopted to implant ^{55}Fe [7].

Currently, the PrimA-LTD project is in its last phase. Ion-implanted ^{55}Fe detectors are being prepared for measurements at PTB and LNHB. ^{129}I electrodeposited sources have been prepared and measured at LNHB and sent to PTB for additional measurements for comparison. Concerning ^{241}Am, the activity of electrodeposited sources was measured using primary measurement techniques in different laboratories of the project partners, to precisely determine their activities. They were measured at LNHB to validate the results from MMC measurements. for activity determination. Both MMC spectrometers of the project partners have been upgraded to now allow simultaneous operation of >10 MMC/SQUID readout channels. This way measurements with very high statistics of isotopes like ^{129}I and ^{55}Fe will be possible. Measurements of ^{55}Fe are of particular interest because they will enable the investigation of very low probability events such as shake-off and shake-up effects.

[1] H. Bhang et al, AMoRE experiment: a search for neutrinoless double beta decay of ^{100}Mo isotope with ^{40}Ca^{100}MoO$_4$ cryogenic scintillation detector. J. Phys.: Conf. Ser. 375 042023 (2012)
http://doi.org/10.1088/1742-6596/375/1/042023

[2] L. Gastaldo et al., The electron capture in ^{163}Ho experiment – ECHo. Eur. Phys. J. Special Topics 226, 1623–1694 (2017)
http://dx.doi.org/10.1140/epjst/e2017-70071-y

[3] M. Paulsen, J. Beyer, L. Bockhorn, C. Enss, S. Kempf, K. Kossert, M. Loidl, R. Mariam, O. Nähle, P. Ranitzsch, M. Rodrigues, Development of a beta spectrometry setup using metallic magnetic calorimeters. J. Instrum. 14(08), 08012 (2019). https://doi.org/10.1088/1748-0221/14/08/P08012

[4] M. Loidl, J. Beyer, L. Bockhorn, J.J. Bonaparte, C. Enss, S. Kempf, K. Kossert, R. Mariam, O. Nähle, M. Paulsen, P. Ranitzsch, M. Rodrigues, M. Wegner, Precision measurements of beta spectra using metallic magnetic calorimeters within the european metrology research project Metrobeta. J. Low Temp. Phys. 199(1), 451–460 (2020).
https://doi.org/10.1007/s10909-020-02398-2

[5] M. Müller, M. Rodrigues, J. Beyer, M. Loidl, S. Kempf, Magnetic microcalorimeters for primary activity standardization within the EMPIR project PrimA-LTD, accepted for publication in J. Low Temp. Phys.

[6] J. Hadid, M. Rodrigues, A. Harouri, Ch. Dupuis, D. Bouville, A. Martin, M. Loidl, L. Ferlazzo, Detachable three-layer Au absorber microfabrication for low-temperature detectors, Micro and Nano Engineering, 20, 100220, (2023).
https://doi.org/10.1016/j.mne.2023.100220.

[7] T. Niemeyer, D. Mowitz, S. Berndt, J. Beyer, H. Dorrer, C.E. Düllmann, A. Göggelmann, R. Hasse, S. Kempf, T. Kieck, N. Kneip, K. Kossert, C. Mokry, M. Müller, O.J. Nähle, D. Renisch, J. Runke, D. Studer , M. P. Takács and K. Wendt, Ion implantation of the electron-capture nuclide ^{55}Fe for measurements by means of metallic microcalorimeters, submitted to Applied Radiation and Isotopes.