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Metallic magnetic calorimeters (MMCs) are a type of cryogenic detectors offering the potential to measure beta spectra, in particular of low to medium energy transitions, with unprecedented precision, owing to their very high energy resolution, linearity and detection efficiency [1]. Within this project, MMCs have been optimized for beta spectrometry measurements in five different energy ranges, from few tens of keV up to ~1 MeV. A new detector module housing, inside a magnetic shielding, the MMC, the SQUID used for the readout of the MMC signal as well as a separate collimated radionuclide photon source for energy calibration has been constructed. Several techniques for the fabrication of high-quality radionuclide sources as well as radiation absorbers have been developed. Data analysis with special care not to introduce any spectrum distortion or other artifacts was also addressed. Most of the developed techniques performed as expected or even better, and where this was not the case alternative solutions were found. Combining the outcome of these developments, several beta spectra could be measured with high precision. These decay data can be used by interested third parties and will also serve to validate the theoretical calculation of the shapes of beta spectra developed in another work package of the project.

1 Introduction

Energy-dispersive low-temperature detectors, e.g. metallic magnetic calorimeters (MMCs), enable radionuclide spectrometry with energy resolutions exceeding by far those of conventional detectors such as HPGe or Si(Li) detectors. Also, the energy threshold of radionuclide spectra can be much lower than with conventional techniques. Within the European Metrology Research Project “MetroBeta”, beta spectrometry based on MMCs with radionuclide sources embedded in noble metal absorbers has been developed. The aim of this Good Practice Guide is to provide an overview of all steps which are necessary to bringing into service a high-precision beta spectrometer based on MMCs.

Within this project five MMC detector designs were developed to span a wide range of beta spectrum endpoint energies of radionuclides of interest. In fact, to perform a high-precision calorimetric measurement, certain design parameters of an MMC detector can be optimized for the endpoint energy of the radionuclide. Based on the endpoint energy the required absorber dimensions are determined by Monte Carlo simulations, thus fixing the corresponding heat capacity at the MMC detector operating temperature. Based on the absorber heat capacity the MMC detector is designed. A detailed description of this process is found in the section Metallic magnetic calorimeters for high-precision beta spectrometry.

The operation of MMC detectors requires a very low temperature environment which is provided by a dilution refrigerator. The readout of the paramagnetic temperature sensors of MMC detectors is done by SQUID sensors. In addition to suitable SQUID sensors, specific SQUID readout electronics is necessary, and the refrigerator must be wired for both MMC and SQUID operation. A magnetically shielded detector module is to be devised to house the MMC detector and the SQUID sensor assembly as well as a collimated photon reference source for energy calibration. These requirements are detailed in the third section Requirements for an MMC-based beta spectrometer.

Reaching an operating temperature below 20 mK is time-consuming and has a non-negligible cost. Therefore, the basic functions and some operational parameters of the MMC detectors are better checked in advance in a much simpler setup in liquid helium. The parameters to be determined comprise the required heater current, the critical detector current as well as the detector inductance. These tests are described in detail in the section Functional testing of MMC detectors. If the MMC detector chip passes the functional testing it is mounted, together with a SQUID chip, in the detector module and is electrically contacted.

The radionuclide to be measured must be completely embedded inside a metallic radiation absorber that is then fixed onto the paramagnetic temperature sensor of the MMC detector. The physical and chemical properties of the radioactive source deposit are of crucial importance for the quality of the measured spectra. In particular, source deposits consisting of ~micrometer large salt crystals can result in significant spectrum distortion, whereas a source consisting of nanometer scale particles or a metallic source deposit provide undistorted spectra. A substantial part of this Good Practice Guide deals with source and absorber preparation techniques which are the most critical and the most difficult part of bringing into service high-precision beta spectrometry based on MMCs. Challenges are the small source dimensions of a few hundred micrometers and the reproducibility of the source and absorber assemblies. The source preparation requires a lot of efforts and time and there is always the risk of failures. Therefore, it can be advantageous to produce...
several sources at a time in a collective process on absorber arrays while controlling every fabrication step to maximize the yield of usable sources. Within the framework of this project, the radionuclides were embedded into the absorber material by different methods. On the one hand, the radionuclide was deposited directly onto an array of half-absorbers. This was achieved with the help of a micro-drop dispensing system. Then, a second array of half-absorbers was diffusion-welded to the first array to ensure a $4\pi$ geometry of each individual absorber. On the other hand, the radioactive material was deposited onto a separate thin source carrier foil which was subsequently placed between two half-absorber foils. Deposition on the source carrier foils was performed both by electrodeposition and by simple drop deposition from a radioactive solution. In the latter case, the crystals forming during drying of the solution were cracked into nanometer small particles by mechanical processing of the source carrier foil. The source carrier foils were subsequently placed between two half-absorber foils. The parameters of the diffusion welding process depended strongly on the composition of the embedded source and on the absorber material. Several tests were done to achieve suitable parameters. The quality of the embedded source is controlled after every preparation step by different techniques. In the end, the source is glued onto an MMC detector with the help of a placement system.

Beta spectra with endpoint energies higher than a few 100 keV measured by MMC detectors can be distorted by the escape of bremsstrahlung from the absorber. Therefore, the combination of different absorber materials in bilayers was explored with the aim of reducing the number of generated bremsstrahlung photons while maintaining a high absorption efficiency for the photons and at the same time the small absorber dimensions required for high energy resolution. All source and absorber preparation techniques as well as the quality control for reproducible source and absorber assemblies are described in the fifth section Source and absorber preparation techniques.

The fifth section Data analysis discusses the acquisition of large volumes of data and their offline analysis. Custom-built measurement routines based on LabView control all parameters of the data acquisition. The signals are continuously stored as binary data. The spectrum is obtained offline by digital signal processing comprising software triggering, application of dead-time and digital filtering.

The last section Results presents the beta spectra of $^{14}$C, $^{151}$Sm and $^{99}$Tc which have been measured by means of MMCs within this project. In this section the results of the high-precision beta spectrometers based on MMC detectors are shown and compared between both independent systems as well as with theoretical calculations being the result of WP 1 “Theoretical calculations of beta spectra”.

2 Metallic magnetic calorimeters for high-precision beta spectrometry

This section gives an overview of the individual steps that must be pursued to optimize, design and fabricate metallic magnetic calorimeters (MMCs) to be used for high-precision beta spectrometry.

2.1 Detector optimization

To perform a high-precision calorimetric measurement of the beta spectrum of a radionuclide of interest, a suitable absorber has to be chosen first. In particular, the absorber material and the absorber dimensions must be selected such that the total energy emitted during a beta decay event, except for the energy taken away by the emitted neutrino or antineutrino, is entirely deposited within the absorber. For making a proper choice, Monte Carlo simulations of different combinations of absorber materials and absorber dimensions are performed. The result of these simulations is a set of potential absorbers that allows to perform the measurement of beta spectrum of the radionuclide of interest. Using the actual absorber material and absorber dimensions, the heat capacity of each of these absorbers at the MMC operating temperature can be calculated using the well-known equations for the electronic heat capacity, the phononic heat capacity, etc. as they are derived within solid state physics. The absorber with sufficient stopping power and the lowest heat capacity at the MMC working temperature is then the absorber of choice for the measurement of the beta spectrum of the radionuclide of interest since the achievable energy resolution depends on the intrinsic heat capacity of the absorber.

The heat capacity of the selected absorber is the starting point for the detector optimization process. Since the thermodynamic properties of the paramagnetic temperature sensor of an MMC detector can be modelled very well using equilibrium thermodynamics and all noise contributions can be, at least empirically, described quite well, the signal-to-noise ratio and hence the achievable energy resolution can be calculated for a certain detector configuration. Here, the absorber heat capacity, the sensor area and thickness, the concentration of the magnetic ions within the sensor material, the area and inductance of the meander-shaped pickup coil, the magnetic field configuration created by a persistent current running through the pickup coil, the SQUID used for detector readout, the pickup coil geometry as well as the MMC operating temperature enter. By stepwise changing the detector configuration where the range of possible detector parameters is restricted by experimental boundary conditions, e.g. the available cooling power and base temperature of the refrigerator used to cool down the detector or the availability of suitable SQUIDs, or available fabrication techniques, e.g. realizable linewidths of the meander-shaped pickup coil, a large set of energy resolution values is created. The lowest value for the energy resolution defines then the ‘optimal’ detector geometry for a given radionuclide of interest.

Obviously, the optimal detector configuration is different for each radionuclide of interest since the range of endpoint energies of beta emitting isotopes is rather large and thus, the absorber geometry and material differ significantly. Hence, in principle, a unique detector has to be simulated, designed and fabricated for each radionuclide. Of course, even though this approach yields beta spectra with the highest precision, it is very time- and resource-consuming in practice, particularly when including the resources needed during the detector fabrication within microfabrication facilities. For this reason, a different approach is most often used and so also within this project. This alternative approach is based on the simple fact that the minimum of the energy resolution of an MMC detector is rather broad and, for example, even a 50% mismatch of the sensor and absorber heat capacities, often result only in a minor degradation of the achievable energy resolution, e.g. of the order of 10%. Including this degradation, MMC based beta spectrometers still yield spectra with the highest precision available today as compared to other techniques. Following this approach, a small set of MMCs can be simulated, designed and fabricated where each detector is optimized for a certain absorber heat capacity value. Within this project, five different detectors have been designed that cover an absorber heat capacity range (at an operating temperature of 20 mK) between 8 pJ/K and 1.7 nJ/K. These absorbers are suitable for measuring beta spectra with endpoint energies in the range between a few 10 keV and ~1 MeV.

For the measurement of the beta spectrum of a certain radionuclide, one starts with the choice of a suitable absorber and calculates the absorber heat capacity at the MMC detector operating temperature. Out of the set of available detectors, one then selects the detector that best matches the calculated absorber heat capacity value.

The approach of using a small set of detectors that cover a wide heat capacity range greatly reduces the time- and resource-consumption. However, all detectors within such a set have a distinct configuration that
can be significantly different from one detector to the other. While geometry dependent parameters such as the linewidth or the pitch of the meander-shaped pickup coil can be adjusted quite easily for each detector and in general many different coil geometries can be combined on a single wafer, parameter as the sensor thickness or concentration of magnetic ions within the paramagnetic material are often incompatible with each other and in principle cannot be combined on a single wafer. For this reason, each detector would require an individual fabrication cycle, having a strong influence on the time and resources that need to be spent within a project.

To overcome this issue, one can again use the fact that the minimum of the achievable energy resolution is rather broad. This allows to find a compromise for those detector parameters that are in principle incompatible with each other, i.e. the thickness of the temperature sensor or the concentration of the magnetic ions. After fixing these parameters to a certain value, an additional detector optimization cycle is performed, this time with some parameters fixed and only varying the parameters that can be easily combined or changed on a single substrate, e.g. pickup coil related geometry parameters. The result of this procedure is a set of detector configurations that can be fabricated within a single fabrication cycle and that can be used for a wide range of endpoint energies. The energy resolution is in most cases degraded by less than a factor of 2 and is still sufficient to yield high-precision beta spectra. This procedure has provided the detector parameters that are summarized within table 1 and that have been chosen for designing the detectors within this project.

### Table 1: Overview of detector parameters based on absorber dimensions

<table>
<thead>
<tr>
<th>absorber type</th>
<th>XS</th>
<th>S</th>
<th>M</th>
<th>L</th>
<th>XL</th>
</tr>
</thead>
<tbody>
<tr>
<td>material</td>
<td>Au</td>
<td>Au</td>
<td>Au</td>
<td>Au</td>
<td>Au - Cu</td>
</tr>
<tr>
<td>shape</td>
<td>circle</td>
<td>circle</td>
<td>octagon</td>
<td>octagon</td>
<td>octagon</td>
</tr>
<tr>
<td>diameter</td>
<td>480 µm</td>
<td>600 µm</td>
<td>725 µm</td>
<td>1655 µm</td>
<td>3065 µm</td>
</tr>
<tr>
<td>thickness</td>
<td>30 µm</td>
<td>70 µm</td>
<td>90 µm</td>
<td>300 µm</td>
<td>350 µm</td>
</tr>
</tbody>
</table>

### MMC detector type

<table>
<thead>
<tr>
<th>MMC detector type</th>
<th>XS</th>
<th>S</th>
<th>M</th>
<th>L</th>
<th>XL</th>
</tr>
</thead>
<tbody>
<tr>
<td>detector area</td>
<td>(249 µm)²</td>
<td>(335 µm)²</td>
<td>(538 µm)²</td>
<td>(1427 µm)²</td>
<td>(2663 µm)²</td>
</tr>
<tr>
<td>single detector inductance</td>
<td>3.4 nH</td>
<td>3.1 nH</td>
<td>8.0 nH</td>
<td>56 nH</td>
<td>196</td>
</tr>
</tbody>
</table>

#### 2.2 Detector design and layout

As described later, special techniques are applied to embed the radionuclide of interest within the absorber. Some of these techniques are incompatible with the actual detector used for performing the measurement. An obvious example is the use of temperatures above 500 K in order to perform diffusion welding of absorbers made of Au which are well-known to significantly degrade the performance of MMCs due to diffusion and subsequent oxidation of Er ions within the sensor material. For this reason, the absorber is often prepared separately and afterwards glued or low-temperature diffusion-welded to the actual MMC chip. In summary, this means that ‘mechanical force’ needs to be applied to the MMC chip that potentially causes damage of critical structures.

In order to avoid such a potential damage, the sensor on which the absorber is to be attached is spatially separated from the bond pads used for contacting the chip as well as the region where many vertical interconnect accesses are placed, see figure 1. The detector configuration itself, i.e. the linewidth and pitch of the meander-shaped pickup coil, the sensor area etc. are adopted from the detector optimization calculations (see section 2.1). Using a proper thickness of the insulation layer (see section below) guarantees that the electrical insulation between sensor and pickup coil can be maintained, e.g. even after applying pressure to the absorber during a gluing or diffusion-welding process.
A simple absorber geometry is a cylinder whose diameter is chosen such that the source embedded in the center of this cylinder is surrounded by sufficient material to reach an absorption efficiency of 100%. Practically speaking, however, a square-shape absorber is mostly simpler to realize. We have chosen an octagonal sensor geometry which is about equally well adapted to cylindrical as to square-shape absorbers. The sensor is connected to a large on-chip heat bath (HB in figure 1) via a sputter-deposited normal metal link made of Au. The ratio between length and cross-section of the link is matched to the target decay time of the detector. This offers the possibility to thermally connect the on-chip heat bath to the sample holder, which is directly attached to the base plate of the refrigerator.

Since the detector module is different for each research group using MMCs, the positioning of the MMC and SQUID chips relative to one another can differ and it is preferable to provide not only one but many interface configurations (bond pads, green in figure 1) that allow to connect the detector. This is the reason why the MMC detector chips fabricated within this project provide three complete sets of bond pads located at the left, lower and right side of the detector chip. Electrically, all associated bonding pads are routed to a common summing point such that the detector performance is not affected by the choice of the actual bond pad set.

2.3 Detector fabrication

The detector fabrication is likely one of the most critical steps during the development of an MMC based beta spectrometer. This section describes some challenges related to the fabrication of MMCs and how these challenges were overcome. In the end, we shortly summarize the different fabrication steps required to fabricate a well-working detector.

The magnetic field within the sensor material which is required for magnetizing the temperature sensor is created by a persistent current running through the pickup coil. The magnitude of this persistent current is rather large, i.e. 10 mA to 100 mA. These values are in the ballpark of the critical current density of the pickup coil that must be close to the critical current density of bulk material. This challenges the fabrication
process of thin superconducting films which are very well-known to often show critical current densities being orders of magnitudes smaller as compared to the bulk value. In addition, any vertical interconnect access (“via”) within the pickup coil must be able to handle these large currents, too, resulting in stringent requirements on the interface transparency between superconducting layers stacked on top of each other. Within this project, the pickup coil was fabricated by using Nb thin films sputter deposited at highest possible rate in a highly pure, low pressure Ar atmosphere. For enhancing the interface transparency between both Nb layers required to produce the pickup coil, hard Ar ion cleaning was used.

Diluted alloys of erbium hosted in a noble metal, as for example Au:Er or Ag:Er, are most often used as sensor material for MMCs. Within the source material or in the final deposited layer, the Er ions are protected against oxidation by the surrounding host material and hence show their very well-known paramagnetic behaviour. However, during deposition it easily happens that residual oxygen present in the deposition chamber oxidizes Er ions resulting in an antiferromagnetic behaviour, i.e. a temperature insensitivity at low temperature. This puts stringent requirements on the background pressure of the deposition chamber as well as on the rate used for the deposition of the film and motivates the choice of sputtering at high rate in a UHV sputtering system as deposition method for the temperature sensors for the MetroBeta detectors.

Table 2: Individual MMC fabrication steps

<table>
<thead>
<tr>
<th>Layer</th>
<th>Material</th>
<th>Thickness</th>
<th>Process</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>Si-Wafer</td>
<td></td>
<td>Sputter deposition followed by UV-photolithography with positive photoresist and reactive ion etching</td>
</tr>
<tr>
<td>1</td>
<td>Nb</td>
<td>250 nm</td>
<td>Anodic oxidation of Nb of layer 1</td>
</tr>
<tr>
<td>2a</td>
<td>Nb₂O₅</td>
<td>~50 nm</td>
<td>UV-photolithography with negative photoresist followed by sputter deposition</td>
</tr>
<tr>
<td>2b</td>
<td>SiO₂</td>
<td>150 nm</td>
<td>UV-photolithography with negative photoresist followed by sputter deposition</td>
</tr>
<tr>
<td>2c</td>
<td>SiO₂</td>
<td>150 nm</td>
<td>UV-photolithography with negative photoresist followed by sputter deposition</td>
</tr>
<tr>
<td>3</td>
<td>AuPd</td>
<td>~150 nm to 200 nm</td>
<td>UV-photolithography with negative photoresist followed by sputter deposition</td>
</tr>
<tr>
<td>4</td>
<td>Nb</td>
<td>600 nm</td>
<td>UV-photolithography with negative photoresist followed by sputter deposition</td>
</tr>
<tr>
<td>5</td>
<td>Au</td>
<td>300 nm</td>
<td>UV-photolithography with negative photoresist followed by sputter deposition</td>
</tr>
<tr>
<td>6</td>
<td>Ag:Er</td>
<td>3 µm</td>
<td>UV-photolithography with negative photoresist followed by sputter deposition</td>
</tr>
<tr>
<td>7</td>
<td>Au</td>
<td>2 µm</td>
<td>UV-photolithography with negative photoresist followed by electroplating</td>
</tr>
<tr>
<td>8</td>
<td>Au</td>
<td>5 µm</td>
<td>UV-photolithography with negative photoresist followed by electroplating</td>
</tr>
</tbody>
</table>

Table 1 clearly shows that the temperature sensor can easily cover an area of several mm², in particular for detectors suitable for measurements of beta spectra with endpoint energies in the range of ~ 1 MeV. The meander-pickup coil is placed below the sensor, it needs to be galvanically isolated from the temperature sensor to avoid short circuits and excess current noise. This demands for the deposition of very well insulating dielectric layers between the pickup coil and the temperature sensor. Most easily, this can be achieved by depositing rather thick insulation layers. However, since the magnetic field created by the persistent current in the pickup coil as well as the magnetic coupling between the magnetic ions and the pickup coil decrease exponentially in vertical direction, the sensor should be placed in close vicinity to the pickup coil, suggesting a rather thin dielectric layer between sensor and pickup coil. Overall, the fabrication process has to be optimized in a way such that thin dielectric layers with a thickness in the range of 100 nm to 300 nm that insulate over rather large areas can be reliably produced. Within this project, a combination of anodic
oxidation of the surface of the pickup coil made of Nb creating a ~ 50 nm thick Nb$\text{O}_5$ layer and a 300 nm thick SiO$_x$ layer sputter deposited in an Ar/O$_2$ plasma was used. This ensures insulation over areas larger than 4mm$^2$ making the fabrication of even the largest MetroBeta detectors possible.

Since the absorber in which the beta emitter is embedded is glued on top of the sensor it is very likely that the sensor-absorber interface is not homogeneous over the entire sensor area. In addition, the sensor might be larger than the absorber area in many cases. Both facts potentially create position dependences which can result in a distortion of the measured beta spectra as well as degradation of the energy resolution. To avoid these effects, the sensor should be heated up homogeneously by a beta event. For this reason, a thermalization layer is placed on top of the sensor having a lateral thermal conductance that is much larger than the lateral thermal conductance of the temperature sensor. In this case, the heat created by a beta event within the absorber first distributes laterally within the thermalization layer (due to its much larger lateral thermal conductance) and then reaches the sensor over the full sensor area. Within this project, the thermalization layer was made of highly pure Au that was electrodeposited on top of the sensor. Since the residual resistivity ratio (RRR) of such a layer can easily be a factor of 20 larger than the RRR of the sensor, the condition of having significantly different lateral thermal conductance is easily met.

After having discussed the most important challenges related to the fabrication of MMCs for high-precision beta spectrometry, the individual fabrication steps are summarized in table 2.
3 Requirements for an MMC-based beta spectrometer

This section gives an overview of the components which are required to set up a high-resolution beta spectrometer based on MMCs. First, a millikelvin refrigerator is needed to achieve the low working temperatures of MMC detectors. It is important that the refrigerator has a good temperature stability and a low vibration level. Also, the wiring of the refrigerator should be suitable to operate MMCs.

The readout of an MMC detector is performed employing SQUID sensors. Working with highly magnetically sensitive SQUIDs requires an excellent shielding of electromagnetic interference (EMI).

A detector module housing the MMC chip and SQUID sensor assembly at the millikelvin temperature platform of the refrigerator is required. Furthermore, analogue electronics to operate the SQUID sensors as well as a digitizer for the data acquisition and an additional hardware bandpass filter are required.

3.1 Requirements for the millikelvin refrigerator

To reach the low working temperature of MMCs a dilution refrigerator is needed with operating temperatures between 1 K and 10 mK. Dilution refrigerators provide continuous cooling power in this temperature regime and operate without any moving part at the lowest temperature stage. The cooling power of the refrigerator should be adequately high with a low parasitic heat flow. Modern dilution refrigerators are customer-friendly systems which work reliably for a long period of time without requiring any service. In figure 2 an easy-to-use cryogen-free pre-cooling dilution refrigerator sold by BlueFors Cryogenics is shown which was installed at PTB for this project. Its cooldown time from room temperature to a base temperature below 10 mK is less than 24 hours. The mixing enthalpy of two isotopes of helium – $^3$He and $^4$He – is used to obtain low temperatures. The pre-cooling is achieved by a pulse tube cooler. The flanges of the 50 K and the quasi-4 K radiation shields are connected to the two stages of the pulse tube. The whole radiation shield assembly is housed in a common vacuum can. The last and coldest stage is the mixing chamber plate.

![Figure 2: Dilation refrigerator at PTB](image)
MMC detectors are very sensitive thermometers requiring a high base-temperature stability. A standard dilution refrigerator has a temperature stability of a few 10 μK. To reach a higher accuracy a better temperature stabilization is needed. Different approaches are possible. In this project a superconducting reference device based on the transition temperature of different superconducting materials was tested. Preliminary tests indicate a temperature stability of a few 100 nK stabilizing the temperature at 30 mK.

Due to the requirements of a high-resolution beta spectrometer based on MMC detectors read out by SQUID current sensors the wiring of the refrigerator has to be custom-made. Four readout channels for MMCs and SQUIDs were installed comprising 30 leads of CuNi alloy ("alloy 30") with a diameter of 0.2 mm. This wiring was continuous from the cryostat top flange at room temperature to the mixing chamber plate. The CuNi leads were routed in a stainless-steel braided hose acting as an EMI shield. Heat sinks along the MMC/SQUID wiring are required and installed at every temperature plate.

When working with highly magnetically sensitive SQUIDs it is advisable to assess the lab environment with regards to EMI and, if necessary, suppress external magnetic signals by an additional magnetic shielding, e.g. mumetal shields placed inside the refrigerator vacuum shields.

### 3.2 Requirements on the SQUIDs

The maximum energy of a radionuclide of interest not only defines the MMC detector design and dimensions, but also a suitable a SQUID sensor for its readout. As mentioned before, the radionuclide has to be embedded into an absorber material to achieve a high-resolution beta spectrum. Therefore, the dimension of the absorber has to be large enough to stop every beta particle within the material. In turn, the heat capacity of the absorber material influences the design of the MMC detector. The heat capacity of the MMC sensor should be equal to that of the absorber, which affects the inductance of the meander shaped pickup coil. The MMC detectors developed within this project are of double-meander type. For this detector type optimal magnetic flux signal coupling is achieved when the inductance of the MMC detector is twice as large as the inductance of the designated SQUID input coil (i.e. $L_M=2\cdot L_N$). An overview of the used absorber types and their resulting heat capacity are shown in table 3. The MMC detector types, which were designed for the considered absorber types, are listed in the same column. Based on the inductance of each MMC detector type a suitable SQUID sensor is selected. The SQUID sensors used in this project have been developed at PTB [2] for different applications with sensor operation temperatures below 1K and had been used before to read out other MMC detectors.

<table>
<thead>
<tr>
<th>absorber type</th>
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<th>M</th>
<th>L</th>
<th>XL</th>
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<td>480 μm</td>
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<td>725 μm</td>
<td>1655 μm</td>
<td>3065 μm</td>
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<tr>
<td>thickness</td>
<td>30 μm</td>
<td>70 μm</td>
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<td>350 μm</td>
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<table>
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<th>M</th>
<th>L</th>
<th>XL</th>
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<td>detector area</td>
<td>(249 μm)$^2$</td>
<td>(335 μm)$^2$</td>
<td>(538 μm)$^2$</td>
<td>(1427 μm)$^2$</td>
<td>(2663 μm)$^2$</td>
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<tr>
<td>single detector inductance</td>
<td>3.4 nH</td>
<td>3.1 nH</td>
<td>8.0 nH</td>
<td>56 nH</td>
<td>196 nH</td>
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<table>
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<th>S</th>
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<td>2 nH</td>
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<td>65 nH</td>
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</tbody>
</table>
As mentioned above, the Metrobeta-MMC detectors are of double-meander type. Hence, both meanders are read out by a single SQUID sensor. This configuration degrades the MMC-to-SQUID flux coupling compared to a single-meander MMC read out by a matched SQUID. However, it has several advantages. Firstly, it allows to simultaneously detect the polarization-decoded signals of two detectors. Secondly, it avoids having to run the rather high persistent field current through the SQUID input coil, which can degrade the SQUID performance. And thirdly, since a double-meander setup can be made in a gradiometric configuration, the net magnetization change for a change in base temperature is strongly suppressed. This design approach renders the detectors significantly less sensitive to base temperature variations during long-term measurements. If the double-meander is slightly off-balanced – as is the case for the detectors developed in this project – this common-mode suppression is only slightly degraded, but the resulting small net magnetization sensitivity to base temperature variations can be used in a twofold way: to check if the magnetization as a function of temperature corresponds to the design behaviour, and as a means to correct the measured pulse heights for base temperature variations.

3.3 Detector module for a dilution refrigerator

For the operation of MMC detectors read out by SQUID sensors a detector module suitable for measurements in a dilution refrigerator was designed. This module holds not only the MMC detector and SQUID sensor assembly but also a reference source for energy calibration. In figure 3 the detector module produced for this project is shown. The body of the detector module consists of a gold-coated copper support structure. The MMC detector and the SQUID sensor are glued to the detector module with insulating varnish and separated by a slit in the copper body to reduce parasitic thermal coupling between both devices. This is necessary due to the higher working temperature of SQUID sensors of nearly 200 mK.

![Detector module of gold-plated copper with a holder for a reference source](image)

Superconducting NbTi wires and copper wires connect the SQUID electronics via printed circuit (PC) boards to the MMC detector and the SQUID sensor. Further electrical connections between the PC boards and the microfabricated devices are realized with superconducting Al bonds. A proper thermalization of both devices is achieved with additional Au bonds between their heat baths and the gold-plated copper support structure. The distance of the reference source to the absorber can be adjusted. There is also an additional
superconducting magnetic shielding made of aluminium to reduce electromagnetic interference. The detector module is mounted in the centre of the mixing chamber plate and connected via Lemo 2.B connectors to the SQUID electronics operating at room temperature.

### 3.4 Readout electronics and data acquisition

To operate the MMC detector and to read out the SQUID sensor analogue electronics are required. The SQUID electronics that have been used within the project, are state-of-the-art commercial high-performance SQUID readout electronics from Magnicon. It enables flux locked loop operation of the SQUID sensors and provides additional current sources. The later are employed to apply persistent MMC meander currents of up to 150 mA and to actuate the persistent current switch with currents of up to 20 mA. All signals from and to the MMC and SQUID sensor are controlled via the SQUID electronics. It is connected to the cryostat wiring directly on the top flange of the dilution refrigerator via LEMO connectors. Other electronics hardware components are a connector box and an output signal cable. The software-based “SQUIDViewerTM” electronics control is based on LabVIEW. The digitization of the SQUID sensor output signal is done in the PTB system using a commercial 2-channel digitizer from AlazarTech (Model ATS 9462) with a maximum sampling rate of 180 MS/s per channel and a resolution of 16 bits. The data acquisition is controlled by self-built LabVIEW programs.
4 Functional testing of MMC detectors

The classification of MMC detectors is separated into a basic functional test and an advanced characterization. The former includes measurements of the values of all relevant MMC connections at room temperature, the determination of the required heater current and a validation of the critical detector current. The advanced characterization covers the determination of the detector inductance and a check of the detector magnetization.

For both characterizations the same high-performance DC SQUID electronics including the SQUIDViewer™ software is used. An equal system is also utilized for the high-resolution beta spectrometer based on MMCs.

MMC detectors contain electrostatically sensitive parts. Therefore, the build-up of electrostatic charges must be avoided when handling or measuring MMC detectors, thus a full ESD protection is mandatory!

4.1 Basic functional test

The basic functional test is less complex and leads to a useful evaluation of the MMC detector quality. One part of the measurements is performed at room temperature and the other part takes place at 4 K.

Resistance measurements at room temperature

A first quality check of MMC detectors is done via resistance measurements at room temperature. For this purpose, a needle prober or a multimeter is used. Figure 4 shows the layout of an MMC detector (type XL) and simplified circuit scheme of a double-meander MMC detector with persistent current switch (PCS). The readout of the MMC with a suitable standard SQUID is done via the terminals +/− SQ. The terminals +/− H connect the PCS to the SQUID electronics and a meander current is injected via the terminals +/− I.

![Figure 4: Left: Layout of an MMC detector chip (type XL; HB indicate on-chip heat bath pads). Right: simplified circuit scheme of double-meander MMC detector with a persistent current by-pass between +/− I terminals, terminals to connect to SQUID input coil +/− SQ, and terminals to connect to PCS heater resistor +/− H](image)

The resistance values are measured for all possible combinations of contacts including the on-chip heat bath (HB). This covers the combinations +/− SQ, +/− I and +/− H. Additionally, the combination SQ/H and SQ/HB must be checked to detect any parasitic connections.

In case that all resistance values are reasonable, the MMC detector is glued onto a test sample holder providing bond pads as well as areas for soldering small Cu wires. The following measurements require low temperatures, e.g., a liquid helium bath, and a low-temperature sample holder. The wiring of the sample holder should be compatible to the standard pin assignment of the SQUID electronics, see figure 5.
Figure 5: Standard pin assignment of the SQUID electronic

Determination of the heater current of the persistent current switch

Terminal +SQ is soldered to the channel 1 voltage connection of the SQUID electronics +V1. -SQ is linked to the ground connection GND1. The heater contact terminal +H is connected to I1, which is an additional current source of the CSE-1 extension board. Correspondingly, terminal -H is connected to the ground I1234. Before the MMC detector is cooled down to 4.2 K, a 4-wire resistance measurement is performed to check the room temperature resistance values at the terminals +/- SQ and +/- H.

As mentioned above, the SQUID electronics is used for the determination of the heater current of the persistent current switch. With the help of the output signal cable, the voltage drops of channel 1 is measured between the terminals +/- SQ using, e.g., an oscilloscope.

The Bias palette of the control software is opened and the value of the bias current I_b is set to 180 µA. Next, the Extension2 palette is selected. Here, slide I1 of channel 3 is chosen and the Connect switch is activated. The measurement is started with no current flowing through the heater terminals +/- H. While increasing the current in steps of 100 µA, the voltage drops at the terminals +/- SQ is monitored with the oscilloscope. Since one of the +/- SQ ports is connected to the part of the PCS underneath the heater, a finite resistance is expected which appears as soon as the PCS is actuated. On the oscilloscope, a sudden change is observed in the voltage drop.

Determination of the detector critical current

The detector critical current, i.e. the maximum supercurrent that a given detector can carry, is obtained by means of a 4-wire measurement at the terminals +/- I and by a 2-wire measurement at the terminals +/- H. Therefore, the terminal +/- I is linked to +V1 and to the additional current source I0. Terminal -I is connected to the ground of both channels, GND1 and I0GND. Again, the heater terminals +/- H are connected to the additional current source I1 and its corresponding ground I1234. The room temperature values of all terminals must be checked before cooling down the MMC chip.

The current flowing through terminals +/- I is increased until the superconducting meander “quenches” to determine the detector critical current. This value should be larger than the necessary persistent current for later measurements. For this purpose, channel 3 of the Extension2 palette is selected and the current source I0 is connected. The current I0 is increased in 1000 µA steps until a sudden change in the voltage drop is observed on the oscilloscope. The corresponding current value is the detector critical current for an inactive PCS.

Further measurements are needed to check whether the rest of the detector, i.e. the pickup coils as well as the other circuitry, remains superconducting when the PCS is actuated. First, a current larger than the
required heater current is applied by the additional current source $I_1$ at terminals $+/\cdot$-$H$. Now, the current source $I_0$ is activated and its current is increased in 1000 µA steps while monitoring the voltage drop with the oscilloscope. The jump-value corresponds to the detector critical current for an actuated persistent current switch.

Another check of the detector is done as follows: a current significantly smaller than the detector critical current is applied at terminal $+/\cdot$-$I$ with the additional current source $I_0$. Now the current value of $I_1$ is increased in 100 µA steps and monitored. The current value $I_0$ at which a sudden change is observed corresponds to the heater current for a given detector current.

In case that all measurements are reasonable the MMC detector passed the first test and is suitable for an advanced characterization at temperatures below 1 K in a dilution refrigerator.

### 4.2 Advanced detector characterization

Advanced characterization concerns the determination of the MMC detector inductances and its magnetization within a temperature range from 20 mK to 200 mK. The measurements are done in a dilution refrigerator using a SQUID sensor with a well-known input coil inductance and the high-performance SQUID electronics from Magnicon. The MMC detector and the SQUID sensor are glued onto the detector module and connected to the SQUID electronics via aluminium bond wires.

The biasing of the SQUID sensor into its working point is done via SQUIDViewer™, and the electronics is subsequently operated in FLL mode. This mode will lock to the adjusted working point and the flux locked loop will generate a dc offset flux, resulting in a dc offset feedback voltage at the output of the SQUID readout electronics. The signals of interest in the advanced detector characterization are referenced to this dc offset signal.

### Wire bonding instructions

The MMC detector chip and the SQUID sensor chip are glued onto the detector module. Afterwards, the electric connections to the SQUID sensor are done by means of Al bond wires. The SQUID sensor chip layout is shown left in figure 6. The wire bond pads $+/\cdot$-$F$, $+/\cdot$I, $+/\cdot$FX, -$+$IFX, and -$+/\cdot$+V are bonded to the corresponding pads on the PC board.

The MMC layout (type XL) is shown in the center of figure 6. One set of the MMC terminals $+/\cdot$-$H$ and $+/\cdot$-$I$ are connected to the corresponding pads on the PC board.

Heat-sinking of the MMC and SQUID chips is realized by means of Au bond wires. Typically, about ten Au wire bonds are used to connect the heat bath pads of the MMC detector and of the SQUID sensor with the detector module or the PC board.

Figure 6: Left: Layout of a SQUID sensor (type S). Center: Layout of an MMC detector (type XL). Right: An image of an MMC detector connected to a SQUID sensor via bond wires; around the MMC and SQUID chips PC boards are glued onto the detector module to provide electrical contacts.
Finally, the MMC detector is connected to the SQUID sensor input coil. These connections are done with single Al wire bonds from the MMC detector bond pads +/-SQ to the SQUID sensor bond pad +/-IN. The wire connection between MMC detector and SQUID sensor should be as short as possible, without touching each other, so as to reduce the stray inductance associated with the chip-to-chip connection.

Inductance of the detector

The inductance of MMC detectors is experimentally determined by means of a Johnson-Nyquist current noise measurement. This measurement is performed using the MMC-SQUID configuration subsequently employed in the beta spectrometer, as shown in figure 6. For this purpose, no persistent current is applied to the MMC detector, and the measurement temperature \( T_{\text{meas}} \) should be above the critical temperature of the Al bond wires, nearly 1.2 K, but below the critical temperature of Nb (material of MMC detector double-meander and SQUID sensor input coil), nearly 9 K.

Normal-conducting Al bond wires represent an Ohmic resistor in series to both the MMC detector double-meander and the SQUID sensor input coil. Therefore, the configuration is modelled as a LR-circuit with \( L \), the total circuit inductance consisting of double-meander inductance and SQUID sensor input coil inductance, and \( R \), the resistance of Al bond wires. The frequency response of the Johnson-Nyquist current noise \( \sqrt{S_I} \) is of 1st order low-pass type and can be expressed as

\[
\sqrt{S_I(f)} = \sqrt{\frac{4 k_b T_{\text{meas}}}{R}} \frac{1}{\sqrt{1 + f^2/f_c^2}}
\]

with the characteristic frequency \( f_c = R/(2\pi L) \) and \( k_b = 1.38 \cdot 10^{-23} \) J/K.

![Figure 7: Measured Johnson-Nyquist current noise spectrum for the estimation of the MMC detector inductance. An MMC double-meander detector type L was read out using a SQUID current sensor of type XS at \( T_{\text{meas}} \) = 4.3 K.](image)
The measured Johnson-Nyquist current noise spectrum is approximated using the above expression with $L$ and $R$ as fit parameters. An example of this fit is shown in figure 7. The SQUID sensor input coil inductance as well as its coupling are known from separate SQUID sensor characterization measurements, see table 3. Furthermore, typical resistance values of Al bond wires are of the order of 1 mΩ. Fits that would drastically differ from those bond wire resistance values indicate inconsistency with the circuit model. The MMC detector inductance is deduced from the fit parameter value of $L$ for the total circuit inductance.

**Magnetization of the detector**

Several temperature-dependent measurements are needed to determine the magnetization of an MMC detector. For this characterization, the magnetic flux change in the SQUID sensor is recorded as a function of the varied bath temperature.

Using one of the additional current sources integrated in the SQUID electronics a short pulse is applied to the heater resistor of the PCS via $I_I$, while at the same time a dc current is applied to the persistent current terminals using the current source $I_0$. This procedure results in a well-defined persistent current in the double-meander circuit. Actuating the PCS causes an increase of the temperature of the cryostat. During the subsequent cool-down of the bath temperature the temperature dependent measurements of the MMC detector magnetization can be performed. Alternatively, the bath temperature can be varied by applying a current to the mixing chamber (MX) plate heater resistor. During the measurements the MX plate heater current is decreased by means of a software-based PID controller and the bath temperature $T$ together with the magnetic flux change are recorded in sync. Typically, the magnetization measurements are done for five different persistent current values (10 mA, 20 mA, 40 mA, 60 mA and 80 mA). The total magnetic flux change observed over the temperature range of the magnetization measurement should linearly increase with the applied persistent current, see figure 8.

![Figure 8: Example of a magnetization measurement: the magnetic flux change in the SQUID in units of a flux quantum $\phi_0$ is measured as a function of the bath temperature for different persistent currents injected into the MMC detector.](image)
5 Absorber design and fabrication

The absorber material should have a high stopping power for beta particles, must allow for a fast thermalization, should be available in a highly pure form while being chemically stable and easy to machine. For these reasons, noble metals such as gold or silver are suitable absorber materials. Based on these considerations highly pure (5N) Au foils were selected for most of the source/absorbers prepared during this project.

Reliable and reproducible source preparation for MMC detectors is desirable, and the fabrication of multiple sources at once is advisable.

Au foils with a size of 21 mm × 21 mm are used as a basis to obtain a good mechanical stability of the structured foil. On this area different absorber sizes can be produced. In figure 9 an example of a structured Au foil with the two largest absorber types is shown. The absorbers have an octagonal shape resembling the shape of the MMC detector.

Depending on the thickness $d$ of the Au foil, laser cutting or milling techniques can be employed to produce arrays of individual absorber elements.

![Figure 9: Example of an absorber array for the two largest absorber types](image)

5.1 Laser cutting

Structuring the absorber arrays by laser cutting is suitable for thin foils ($d < 90 \mu m$). The Au foil is fixed on a glass slide with photoresist for this technique. This is necessary for stability reasons during the process. Glass slides are suitable sample holders because they do not chemically react with the Au foils. The absorber design is cut by a laser beam. After this step, the structured Au foil is released from the glass slide with the help of acetone and an ultrasonic bath. The thin Au foils must be handled carefully and should never be dried with compressed air.
A disadvantage of structuring a Au foil by laser cutting was the formation of residues of evaporated gold leading to rough edges and gold bulges on the surface, see figure 10 (left). For thick foils the gold bulges are a serious problem.

5.2 Milling

Milling of Au foils has been found to be suitable for Au absorber foil thicknesses \( d \geq 90 \) μm. The foils are glued onto a disposable substrate using double-sided adhesive tape. After milling thorough solvent-based cleaning of the foils is advisable. Milling provides well-defined edges, see figure 10 (right). However, milling of Au foils (with thicknesses \( d < 90 \) μm) was found not to be reliable to format absorber elements due to mechanical damage.

5.3 Absorber design for high energy beta spectra

Beta spectra with endpoint energies higher than a few 100 keV can be distorted by energy escape processes, for instance via bremsstrahlung losses. Although every beta electron is stopped within a given absorber, some bremsstrahlung photons may escape. This effect causes some of the electrons with high initial energies to be detected with reduced energies. The probability of bremsstrahlung production increases with the electron energy, therefore only rather high energy beta spectra are concerned. The cross section of bremsstrahlung production also strongly increases with the atomic mass of the material in which the electron decelerates. Hence, a low atomic mass absorber material would have the advantage of reducing the number of generated bremsstrahlung photons. However, this would also have two disadvantages. Firstly, a larger absorber is then required to stop the electrons, thus degrading the energy resolution and enhancing the energy threshold of the detector. Secondly, the absorption probability for the photons of a low atomic mass material is lower, thus partially counteracting the advantage of reduced bremsstrahlung production.

A better choice may be a bilayer absorber. An inner low atomic mass layer would reduce bremsstrahlung production where the electrons have still high energy, and a high atomic mass outer layer would reduce the total absorber thickness required for stopping the electrons and at the same time enhance the absorption probability for the photons.

The case of \(^{36}\)Cl, the highest energy beta spectrum measured within this project \( (Q_{\beta} = 709 \text{ keV}) \), has been studied via a series of Monte Carlo simulations. As a first step, the minimum absorber dimensions required to fully stop all electrons up to 709 keV were determined for five absorber materials or material combinations: gold (Z=79), silver (Z=47), copper (Z=29), and silver-gold and copper-gold bilayer absorbers. In a second step, full Monte Carlo simulations were performed for the five previously dimensioned absorbers in order to evaluate the spectrum distortion by bremsstrahlung escape. A realistic theoretical spectrum considering beta spectra measurements based on MMC of \(^{36}\)Cl was used as an input for the simulations and the output is the distribution of the energy that would be deposited in each of the absorbers, i.e. the expected shapes of the measured spectra. Figure 11 shows a comparison of the results of these Monte Carlo simulations. For better visibility, two energy ranges are shown in enhanced viewgraphs in the bottom part of figure 11. It can be
observed that indeed the spectrum distortion decreases with the atomic number for the monolithic gold, silver and copper absorbers. The spectrum distortion of a silver-gold bilayer absorber is weaker than for monolithic gold or silver absorbers and comparable with a monolithic copper absorber, and for a copper-gold bilayer absorber it is the smallest of all studied absorber configurations.

For the measurement of the beta spectrum of $^{36}$Cl, a Au-Cu bilayer absorber is considered consisting of a gold foil and a copper foil. Oxidation of the copper surface when exposed to air may cause difficulties in a subsequent diffusion welding process. An additional thin noble metal layer of <100 nm deposited onto the copper surface can prevent this oxidization.

Monte-Carlo simulations of Au-Cu bilayer absorbers

Using the Monte Carlo software EGSnrc [3], several simulations were carried out to compare the energy escape reduction capacity of three absorber configurations:

1) Au-XL-absorber, $d = d_{Au} = 300 \ \mu m$
2) Au-Cu-XL-absorber, $d_{Au} = 200 \ \mu m$ and $d_{Cu} = 150 \ \mu m$.
3) Au-Cu-XL-Au-absorber, $d_{Au} = 200 \ \mu m$, $d_{Cu} = 150 \ \mu m$ and $d_{Au\_layer} = 50 \ \text{nm}$
Figure 12: Plots of 1000 simulated particle tracks for electrons having an initial energy 800 keV, showing the cross-sections of Au-XL-absorber (left), Au-Cu-XL-absorber (center) and Au-Cu-Au-XL-absorber (right).

The third configuration was simulated to obtain an upper bound of the effect of adding the thin layer of Au/Pd. In figure 12, the simulated tracks of 1000 electrons with initial energy 800 keV starting from an isotropic point-like source at the center of the absorber are plotted. The electron and photon tracks are depicted in red and yellow, respectively.

Qualitatively, the Au-XL-absorber has better stopping power for the electrons but more photons escape. The opposite is the case for the Au-Cu- and Au-Cu-Au-XL-absorber. These features become more obvious when performing a quantitative simulation of $2 \times 10^6$ electrons with initial energy 800 keV, to record the photons that escape the absorbers, as shown in table 4:

Table 4: Comparison of energy escape simulations for the three absorber types; using a centered point-like isotropic source, $2 \times 10^6$ electrons with initial energy 800 keV were simulated.

<table>
<thead>
<tr>
<th>Absorber type</th>
<th>Au-XL</th>
<th>Au-Cu-XL</th>
<th>Au-Cu-Au-XL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of primary electrons</td>
<td>2,00E+06</td>
<td>2,00E+06</td>
<td>2,00E+06</td>
</tr>
<tr>
<td>Total energy initialized in MeV</td>
<td>1,60E+06</td>
<td>1,60E+06</td>
<td>1,60E+06</td>
</tr>
<tr>
<td>Total energy escape in MeV</td>
<td>30766</td>
<td>13232</td>
<td>13290</td>
</tr>
<tr>
<td>Proportion of energy escape</td>
<td>1,92%</td>
<td>0,827%</td>
<td>0,831%</td>
</tr>
<tr>
<td>Number of escaped photons</td>
<td>118220</td>
<td>64511</td>
<td>64611</td>
</tr>
</tbody>
</table>

Surface count spectra, i.e. the energy distribution of the photons that escape the respective absorber configuration, are depicted in figure 13.

The simulations show that using a bilayer absorber reduces the energy escape by approximately 57% for electrons having an initial energy of 800 keV while the effect of having an additional thin layer of Au/Pd should be negligible.

Figure 13: Surface count spectra plots of the three absorber configurations.
6 Source Preparation

One aim of this project was to improve existing source preparation techniques. The source preparation is essential for high-precision beta spectra based on MMC detectors. The radioactive material must be completely embedded into the absorber material to ensure that every beta decay is detected and – in the ideal case – the decay energy is completely deposited into the absorber and entirely thermalized, i.e. transformed to heat. It has been known, e.g., that the presence of large (of order micrometers) salt crystals, that can form upon radionuclide deposition from a charged salt solution, can result in spectrum distortions. The reason is that a fraction of the decay energy deposited in the salt crystals is expended in the formation of crystal defects or long-lived electron-hole pairs and therefore not thermalized.

For the absorber/source preparation within this project, the radioactive material was embedded into the absorber by two different methods. Using the first method, the radionuclide is firstly directly deposited onto one half of the absorber. Subsequently, the second half of the absorber is welded onto the first part to form a $4\pi$ geometry. For the second method, the radioactive material was deposited onto a separate thin noble metal foil that acts as a source carrier foil; this foil was subsequently placed between two half-absorber foils, and this foil stack was diffusion-welded together.

The parameters of the diffusion welding processes depended strongly on the embedded source material and the absorber material. Several tests have been carried out to achieve suitable parameters. It is advisable to perform quality control checks of the embedded sources.

There are several methods to deposit a radionuclide onto metal foils. Ion implantation [4] has the potential to provide ideal sources with the radionuclide implanted directly into the absorber material, but it is a complex and costly technique which requires dedicated facilities. Here, electrodeposition [1] as well as drop deposition were investigated.

6.1 Electrodeposition

Sources from a number of radionuclides can be, depending on their electrochemical potential, prepared by electrodeposition onto electrically conducting substrates, either metal plates of foils, or metallized polymer membranes. The substrate acts as the cathode at the bottom of a deposition cell made e.g. from PTFE, and an anode is immersed in the electrolyte containing the radionuclide. Within the project, $^{151}$Sm and $^{99}$Tc were electroplated on silver respectively gold foils on diameters of 8 mm to 12 mm. $^{99}$Tc can be deposited in metallic form, whereas in the case of $^{151}$Sm an oxide/hydroxide layer form. Since these source carrier foils are much larger in area than the MMC detector compatible absorbers, a small piece of each foil with the desired activity had to be cut and sandwiched between two half-absorber foils.

![Figure 14: The setup used for electrodeposition. Also shown is the shadow mask.](image_url)
It has been tried to electrodeposit radionuclides directly onto the individual absorber elements of an array. A well-established setup for electrodeposition was adapted for this approach, shown in figure 14. Here, the absorber material formed the cathode, a rotating cylindrical grid of platinum was the anode and the radioactive material was dissolved in the electrolyte. A shadow mask was designed with openings in the middle of each absorber within the array. Unfortunately, this approach failed, presumably because of the reduced surface area of the anode due to the mask. The activities deposited were negligible and no parameter set could be found to change this. Thus, this technique was not followed any further.

6.2 Drop deposition and dispersion of the activity in the absorber material

The simplest way to produce radionuclide sources is drop deposition by manual pipetting using pycnometers with a fine capillary. Since the formation of large crystallizations should be avoided, carrier-free solutions are preferable, if available. However, most commercially available radionuclide solutions are charged with relatively large amounts (compared with the radionuclide concentration) of inactive carrier salt in order to enhance the thermodynamic stability of the solutions. Simple drying in air will then often result in rather large crystallizations, especially because the solutions tend to concentrate on very small areas while drying. There are various ways to remedy this problem by either distribute the activity on a larger area or by fine dispersion of the radioactive material in the absorber metal.

One way to avoid the shrinking of the drop of solution while drying is freeze-drying. The drop must be frozen immediately upon deposition and then be freeze-dried under coarse vacuum. The ice of the frozen droplet sublimes away and the radioactive material deposits on the entire area covered by the initial droplet. This technique requires a dedicated apparatus comprising a very efficient cooler and a vacuum pump.

Another way to avoid the shrinking of the drop of solution is to cover the substrate surface by a thin layer of latex microspheres by electro-spraying prior to the deposit of the radionuclide solution. The solution will spread on the entire surface covered by the microspheres due to capillarity and therefore also dry on the same surface area. However, it has not been fully clarified if the presence of the latex microspheres in the absorber alters the thermodynamic properties of the sample or if they tend to distort the spectrum in a similar way as salt crystals.

If neither of the two methods can be applied or if they result in still too thick deposits – especially in the case of long-lived nuclides requiring rather large quantities of salt – there are different ways to disperse the radioactive material in the absorber material. One of them consists in absorbing the radioactive solution into a sample of gold nano-foam whose pore size, of the order of tens of nanometers, constrains the size of individual salt crystals. The nano-foam can be prepared by de-alloying gold-silver alloy by chemical etching under controlled conditions. This approach has been tested with alpha-decaying nuclides, but not yet with beta emitters.

Another method consists in repeated folding and laminating of the metal foil with the radioactive material. This mechanical processing breaks the salt crystals to increasingly small particles embedded in the metal. After a number of cycles, particle sizes of only few nanometers have been observed. However, if the individual layers in the metal foil stack become too thin, they tend to fall apart. This does not only destroy the sample under preparation but is also problematic in terms of radioprotection. To avoid this problem, it is advisable to proceed to intermediate diffusion welding cycles once the individual layer thickness decreases to below a few micrometers.

**Micro dispensing system**

Drop deposition by an automated micro dispensing system was the best approach realized in this project to obtain sources reproducible in terms of activity. Several tests were conducted using a commercial micro dispensing system (microdrop MD-P-826). This system is based on a piezo-driven inkjet printing technology. The core of this system is a pipette surrounded by a piezo actuator. This unit has a storage volume of 25 µl for liquids. The drop volume depends on the used liquid, the nozzle diameter of the pipette and the pulse parameters applied by the piezo actuator. The liquids are filled from a glass vial into the pipette. The drop size and the dispense position are controlled by two cameras. The overall travel range of the pipette is 200 mm × 200 mm × 100 mm. For each radionuclide a new pipette is used to avoid the risk of cross-contaminations.
Figure 14: Left: The installed micro dispensing system with the support structure for the absorber array (red). Center: Calibration of the drop size with the help of a camera perpendicular to the flight direction of the drop. Right: Dispensing a liquid onto an absorber array. To the upper right the vertical camera used for positioning is shown.

Typical radionuclide carrier solutions contain certain salt loads (e.g. $^{36}$Cl in 0.05 mol/l NaOH, 33 mg/l NaCl). They are deposited with a placement accuracy of 20 μm and drop volumes of less than 50 picolitres. This demonstrates high reproducibility for the drop dimensions, placement accuracy and source activity. However, drop sizes of 200 pl are optimal for this project. They are easier to achieve and have a linear trajectory. In contrast, manual drop deposition allows a minimum volume of 0.1 µl and a placement accuracy of around 500 μm with a low reproducibility.

For the micro dispensing system, a support structure was designed with an intake for an absorber array holder which was also used as the lower part for the diffusion welding press, see figure 16 and 18. The support structure has openings for vials filled with the radioactive solution or cleaning liquids. These positions are saved in the software-based positioning system.

To avoid cross-contaminations a dispensing cycle must be accurately planned and controlled. Firstly, the typical drop size of a pipette is determined and adjusted with distilled water or with carrier solution, seen in figure 16. Then, the dispense positions, relative to the middle of each absorber, are verified by the horizontal camera (figure 16) and listed in the dispensing program. The pipette is cleaned a few times with distilled water before it is filled with the radioactive solution. A filling time of 3 sec corresponds to a volume of around 25 µl. This is controlled by weighing the glass vial before and after the dispensing cycle. Before the dispensing program is started the drop size and the positioning accuracy are controlled again.

Normally, a drop pattern, which is shown right in figure 16, is dispensed to limit the crystal growth caused by the salt load of the carrier solution. Therefore, the distance between each drop should be large enough that the drops could not converge. Also, the dropped solution must be dried before a new drop could be deposited at the same position. Only low activities are loaded to the micro dispensing system which limits the specific

Figure 15: Comparison of drop disposition done manually (left) and by a micro dispensing system (center). In both cases an overall volume of 0.5 µl was dispensed. In case of the micro dispensing system a drop pattern was often used (right).
activity of the radioactive solution in the glass vial. Considering the processing of normally 36 absorbers with up to 0.5 µl, the automated drop depositing exceeds 12 hours. After dispensing the pipette has to be cleaned immediately. Otherwise, the pipette could be blocked due to residual salt crystals forming in the glass capillary from the remaining solution.

After a dispensing cycle, the quality of the sources and the reproducibility of the micro dispensing system are checked.

6.3 Check of unsealed sources

Depending on the salt load of the radioactive solution the position of the dried radioactive material is controlled optically with the horizontal camera of the micro dispensing system. Pictures of every absorber are taken and evaluated to ensure that the radioactive material is deposited in the middle of each absorber.

Another part of the quality check of the absorber array is done by autoradiography. Based on this technique the deposited activities could be estimated, an example of this method is shown in figure 17 (right). Here, two different activities, 5 Bq left and 2.5 Bq right, are deposited. Optically, a check of the activity was not possible but based on the results of the autoradiography the position and the different activities of the deposited material could be confirmed. An exposure time of 24 hours for an unsealed source is sufficient to observe also the shadow of the Au foil which is useful for mapping. Possible effects of outgassing are seen with the help of this method.

![Figure 16: After dispensing the open source is controlled optically (left) and by autoradiography (right). The two different activities, 5 Bq left and 2.5 Bq right, are nicely seen in the picture of the autoradiography.](image_url)

6.4 Diffusion welding

Diffusion welding makes use of the principle of solid-state diffusion. Atoms of two solid, metallic surfaces intersperse themselves over time. In addition, the welding process depends on the applied pressure and the process temperature. In figure 18 the design of a press for diffusion welding is shown which is used due to the expected high stress for the material. The press consists of two halves of aluminium, four screws and four guidance pins for the absorber foils. The lower part of the press fits to the intake of the holder in the micro dispensing system. Glass slides, borosilicate glass with dimensions of 0.2 mm × 18 mm × 18 mm, are used as separating layers during the diffusion process. Otherwise, the gold would also be welded to the press itself. For the actual diffusion welding process, the loaded press is placed in an oven, and exposed to a well-defined temperature for a fixed time.
Figure 17: Left: Design of the press used for diffusion welding consisting of two halves of aluminum, four screws and four guidance pins. Right: The lower part of the press with a glass-absorber-source-absorber-glass sandwich and the four guidance pins.

The absorber array with the radioactive material is mounted on a glass aluminium assembly and a second absorber array of the same shape and thickness but without radioactive material is placed on top of it. With the help of the guidance pins the absorber foils cannot shift against each other. The other glass aluminium assembly is put on-top of the absorber-source-absorber sandwich, and the four screws are fastened to produce a pressure. Then, the guidance pins are removed otherwise the gold could also be welded to the pins.

Depending on the deposited radioactive material and the used carrier solution, suitable parameters of diffusion welding can differ significantly. To avoid melting or outgassing of the radioactive material, the parameters were adjusted. Several tests were done to identify suitable diffusion welding process parameters.

For materials with high melting points of all chemical compounds, e. g. \(^{36}\)Cl, the following parameters were determined:

- Torque: 2 Nm per screw
- Oven temperature: 400°C
- Process time: 45 min to reach 400°C and 1 hour at 400°C
- Cooldown: 12 hours

For materials with low melting points of one chemical compound, e. g. \(^{99}\)Tc and \(^{14}\)C, the following parameters were determined:

- Torque: 10 Nm per screw
- Oven temperature: 140°C
- Process time: 45 min to reach 140°C and 40 h at 140°C
- Cooldown: 12 hours
After diffusion welding, the press is carefully opened because there might be broken glass slides, seen in figure 19. The broken glass is removed from the welded gold foils and collected for further tests.

In case of a successful diffusion welding, the radioactive material is enclosed completely by gold without any contaminations on the gold surface. To check this, different control procedures have been developed within this project. Also, the press surface is tested for contaminations by wipe testing. If no contamination is detected, the press is resurfaced for a new diffusion welding process.

**Diffusion welding of Au-Cu bilayers**

Preparation of Au-Cu bilayers is possible by means of diffusion welding. Due to the increased tendency of Cu to oxidize at elevated temperatures, it is, however, advisable to carry out this process in a nearly Oxygen-free atmosphere. Therefore, the welding oven needs to be evacuated and/or purged with inert gas before and during the bilayer preparation.

There exists extensive literature on the Au-Cu solid system as well as interdiffusion at Au-Cu interfaces. The gold-copper phase diagram [5] shows that continuous solid solutions form over a large temperature range. The minimum melting temperature of these solutions is 910°C. For practicality, the preparation of the Cu-Au-bilayer foils should occur before absorber formatting and source deposition. Then, above mentioned limitations with regards to process temperature and pressure that are associated with melting/outgassing of radioactive material are not relevant.

Native surface oxide layers from the Cu foil should be removed directly prior to the diffusion welding process. To this end, a combination of mechanical polishing and wet etch in low-concentration acids can be applied. A glass aluminum assembly using four clamping screws very similar to the press fixture described later in 5.4 can be employed for the diffusion welding process. Suitable process parameters to prepare a 24 mm × 24 mm Cu-Au-bilayer foil are summarized below.

<table>
<thead>
<tr>
<th>Process atmosphere</th>
<th>Argon</th>
</tr>
</thead>
<tbody>
<tr>
<td>Torque</td>
<td>6.2 Nm per screw</td>
</tr>
<tr>
<td>Oven temperature</td>
<td>500°C</td>
</tr>
<tr>
<td>Process time</td>
<td>60 min to reach 500°C and 1 hour at 500°C</td>
</tr>
<tr>
<td>Cooldown</td>
<td>3 hours</td>
</tr>
</tbody>
</table>

Cu surface oxide removal: mechanical polishing and 15 min. wet etch in mixture of acetic acid (conc. 1%) and nitric acid (conc. 1%)
After the Cu-Au-bilayer preparation and their formatting it is typically unavoidable to expose the foils to ambient atmosphere, so that native Cu surface oxide again forms. Consequently, Cu surface oxide removal would be needed to be repeated directly prior to the deposition of the source material and the subsequent final diffusion welding. To simplify this oxide removal processing, it is advisable to deposit a comparably thin, e.g. 100 nm, non-oxidizing capping layer onto the Cu surface of the formatted Au-Cu-bilayer. Gold or gold alloys, e.g. AuPd, are suitable capping layer materials. It can be expected that process parameters that result in reliable diffusion welding of Au absorber foils are useable to perform the diffusion welding of AuPd-capped Au-Cu-bilayers.

6.5 Contamination control and quality check

For a first check the broken glass slides from the diffusion welding process are checked for potential contamination with the help of liquid scintillation (LS) counting [6]. If there were any problems during the diffusion welding process, e.g. too high process temperature causing a melting of the radioactive material, traces could be deposited onto the glass slides. The potentially contaminated glass is mixed with a liquid scintillator and the scintillation photons are counted. Due to the low activities deposited on the absorber array contaminations – if any – are expected to be small. Therefore, LS sample preparation and the counter should be adapted to low-level measurements.

Two LS samples are prepared with 15 ml of a commercial liquid scintillation cocktail offering low background, e.g. Ultima Gold LLT. Standard 20 ml polyethylene vials are used which provide lower background and higher light yield than glass vials. The possibly contaminated glass is filled into one LS sample while the other LS sample is filled with the same amount of contamination-free glass. This has to be done because of the intrinsic activity of borosilicate glass causing background counts. In this project the measurements have been carried out in a Quantulus™ 1220 low-level counter. The Quantulus counter comprises a massive passive shielding (about 630 kg of lead) as well as a guard detector to suppress the background due to (external) gamma-rays. Since no gamma-ray emission from the potential contamination is expected, the guard detector is used to reduce the background significantly.

The counting rate is determined for both LS samples. A typical counting rate for a LS sample with about 240 mg of contamination-free glass is about 24 CPM (counts per minute). If the counting rates of both LS samples are comparable there is no contamination. Otherwise, a contamination is detected, the spectra of the contamination is determined, and the diffusion welding process is revised. Examples of the detection of no contamination as well as of contamination are shown in figure 21.

Another means of quality check of the welded absorber array is autoradiography. This time the exposure time is clearly larger than for an unsealed source. Typically, it took 6 days to observe effects due to not-perfectly closed absorbers. In figure 22 two autoradiographic images taken after diffusion welding of $^{36}$Cl (left) and $^{99m}$Tc (right) are shown. In the case of the embedded $^{36}$Cl sources the emission of bremsstrahlung is mainly observed because of the higher electron energies. However, the right edge of the third absorber in the first row is brighter. This indicates that the absorber is not completely welded and a complete $4\pi$ absorber geometry is not reached. In case of the $^{99m}$Tc absorber array, figure 22 right, a contamination of the gold surface was detected before with LS counting. Here, the radioactive material melted during the diffusion welding due to the high process temperature of 400°C resulting in a contamination of the absorber edges.
Considering all control mechanisms, a suitable absorber source assembly was selected and was separated carefully with a scalpel.

*Figure 21:* Spectra corrected by the background spectrum obtained from LS samples with glass pieces measured in a low-level LS counter. Left: Measurement of glass pieces from the preparation of $^{36}$Cl sources. No contamination with $^{36}$Cl is detected. Right: A contamination was detected in the case of $^{99}$Tc sample preparation.

*Figure 22:* Autoradiographic images after diffusion welding of two different absorber arrays. Left: $^{36}$Cl absorber array. The emission of bremsstrahlung is seen but it can also be concluded that the third absorber in the first row is not perfectly sealed and thus, cannot be used. Right: $^{99}$Tc absorber array. The radioactive material melted during the diffusion welding and contaminated the absorber surface.

The last source preparation step is to couple the absorber to a successfully tested MMC detector. Here, it is important to keep in mind that MMC detectors and SQUID sensors are prone to electrostatic discharge (ESD) damage. ESD safety measures should, therefore, also be applied when absorber elements are mounted to the MMC detectors.

The MMC chip and the SQUID are glued with GE Varnish onto the detector module, and all wiring is finished. The positioning of the source-absorber assembly onto the MMC detector is done with the help of a placement system, commonly called pick-and-place machine. This kind of machines is typically used to place surface-mount devices onto a PC board with high precision.

Shortly before the absorber is glued onto the MMC detector, a two-component epoxy resin adhesive, Stycast 1266, is mixed. Then, small glue drops are placed on-top of the gold stems of the MMC detector with the help of a thin wire. The absorber is accurately positioned so that it does not touch any wiring on the MMC.
detector chip. During the curing of the two-component adhesive at room temperature, a weight, ranging between 500 g and 1000 g depending on the absorber size, is loaded onto the absorber, see figure 23.

All resistance values of the MMC detector and the SQUID sensor are re-checked after the glue hardened which typically is 24 hours after preparation. If all resistance values are reasonable, the detector assembly is ready for the measurement of beta spectra.

Figure 193: Left: Positioning of the source-absorber assembly onto the MMC detector. Center: The hardware which is used to apply a constant pressure onto the glued source-absorber assembly. Right: Closer look on the mechanism
7 Data analysis

An advantage of this project is the use of two independent MMC-based beta spectrometers: the established spectrometer at CEA and the new system at PTB. Hence, the reliability of a measured beta spectrum shape could be confirmed within this project. To ensure that there are no systematic software problems, the continuous data recording and the offline data analysis were also developed independently as described in this section.

7.1 Data analysis at CEA

The analogue output voltage of the Magnicon SQUID electronics is amplified and filtered with an SR560 amplifier from Stanford Research Systems. The output is connected to a PCI acquisition card P25M from Innovative Integration. The data are recorded continuously with a sampling frequency of $f_s = 100$ kHz and stored on a solid-state drive. The antialiasing low-pass filter frequency on the SR560 amplifier is 30 kHz. The data are recorded during about two weeks. The spectrum is obtained offline by digital signal processing with Matlab with these following steps:

- recording of the pulse timestamp triggering on two thresholds
- application of an extended dead-time windowing
- construction of the average pulse and the average noise spectrum
- determination of the pulse energies and several other pulse parameters
- correction of the temperature drift
- rejection of the pile-up on the pulse decays
- energy spectrum construction

Triggering and pulse timestamps

The time positions of the pulses are found using two trigger thresholds; positive and negative thresholds are required to trigger the pulses of opposite sign from the two sensors of each gradiometric double-meander circuit. The thresholds are applied on streams filtered by a high-order Butterworth band-pass filter with cut-off frequencies of typically 400 Hz and 700 Hz. Positive and negative thresholds are set with fixed values during the entire recording, the timestamps at which each signal passes above and under the thresholds are recorded and named respectively $t_{\text{start}}$ and $t_{\text{stop}}$ (figure 24). A third parameter is stored in the timestamps binary file: the polarity of the signal (+1 or -1) to indicate which threshold is triggered, and 0 is stored if the signal is saturated.
Application of an extended dead-time windowing

Once all pulse timestamps are recorded, an extended dead-time function is applied on the timestamps. The goal of this function is to suppress the timestamps corresponding to pile-up on the pulse baseline or the timestamps from artefact signals (i.e. noise triggering, undershoot). For all timestamps $t_{\text{start}}$, a dead-time window begins with a duration $T_{\text{dead}}$ given by $T_{\text{dead}} = F_{\text{dead}} \times (t_{\text{stop}} - t_{\text{start}})$. $F_{\text{dead}}$ must be larger than the ratio between the duration of raw pulse and a band-pass filtered pulse (see section before and figure 24). In conventional extendable dead-time the duration of the dead-time window is fixed; here, using a window length proportional to the pulse duration allows minimizing the percentage of dead-time. All the signals having a $t_{\text{start}}$ falling in a dead-time window are suppressed and the initial dead-time window is extended by a new dead-time proportional to the duration of the suppressed pulse (figure 25).

Construction of the average pulse and the average noise spectrum

Among the methods used to determine the pulse energies, two of them are used, respectively in time domain (TD) and frequency domain (FD):

- A template pulse $M(t-t_{\text{start}})$ is linearly fitted to the detected pulse $m(t-t_{\text{start}})$ using two parameters
  \[ m(t-t_{\text{start}}) = A_{\text{TD}} \cdot M(t) + B_{\text{TD}} \quad \text{eq. 1} \]
  where $A_{\text{TD}}$ correspond to the pulse energy and $B_{\text{TD}}$ is the baseline level

- The other method is known as the optimal filtering. The pulse amplitude is estimated in the frequency domain using:
  \[ A_{\text{FD}} = \frac{\sum_{n=-N}^{N-1} s_n^* v_n}{\sum_{n=-N}^{N-1} |s_n|^2 J(f_n)} \]
  where $s_n$ is the fast Fourier transform (FFT) of the template pulse normalized to 1 in the time domain, $J$ is noise power spectral density and $v_n$ is the FFT of the detected pulse.

Figure 24: The raw signal is filtered by a Butterworth band-pass filter. Positive and negative thresholds are applied on the filtered signal to find the start and stop timestamps.

Figure 25: Example of the application of extended dead-time on a stream containing pulses and pile-up.
A template pulse is required for both frequency and time domains, and the mean noise power spectral density is required in the frequency domain. The template pulse is obtained by averaging about 100 pulses. The first 5 pulses are chosen “by eye” and the next are automatically selected using upper limits on the standard deviation of the pulse baseline, and on the chi-square ($\chi^2$) between the fit of the template and the detected pulse. The number of samples of the pulse template is a power of two, which is more convenient for the FFT into the frequency domain. The first half of the template pulse is the baseline, and the second half contains the pulse rise and the main part of the pulse decay. The mean noise power spectral density is a mean of 8192 FFTs of selected clean baselines with low standard deviations and with a mean value close to one.

**Determination of the pulse energies and several other pulse parameters**

During this step, each pulse having its $t_{start}$ falling in the live time is analyzed. The pulse is pre-filtered with a low-pass filter with a cut-off frequency of $f_e/10$. This guarantees to have enough samples in the leading edge; the energy resolution with optimal filtering is sensitive to a lack of sampled data points. The pulse baseline level is measured and subtracted from the pulse. Then, the amplitude is determined with the two methods described before. In the time domain, the detected pulses are fitted with template pulses of two different lengths: one template has the full length of the template pulse; the other one has a reduced length, typically about 1 ms to 2 ms. The use of a reduced length gives a better estimation of the parameter $A_{FD}$ (equation 1), because it reduces the contribution of the low frequency noise.

In addition, several other parameters are measured, for example:

- The maximum of the pulse,
- the pulse area,
- the timestamps of the pulse maximum $t_{max}$,
- the 10% to 90% rise time constant,
- the parameter $\chi_{comp}$, i.e. the $\chi^2$ of the fit between the pulse and the full pulse template,
- the parameter $\chi_{red}$, i.e. the $\chi^2$ of the fit between the pulse and the reduced pulse template,
- the standard deviation of the pulse baseline,
- the level of the pulse baseline.

These parameters are used to check the pulse analysis and to reject the remaining pile-up or artifacts.

**Correction of the temperature drift**

The temperature of the detector stage is not regulated, and it is given by the base temperature of the cryostat. Therefore, the pulse amplitudes are subject to a temperature drift which can be corrected using two ways:

1- The first method consists in using the fluctuations of the amplitudes at a given energy as a reference. For example, these amplitudes can be given by a gamma-ray line from the calibration source. It is recommended to use a line with sufficient statistics, in order to achieve a good sampling of the temperature drift, and a line with a relatively high energy compared to the rest of the spectrum, in order to have a good sensitivity to the temperature drift. Therefore, a spline function $f_{amp}(t)$ is determined from these amplitudes as a function of time and all the amplitudes $A$ are corrected by $A_{corr} = A / f_{amp}(t_{start})$ (figure 26).

2- The second method consists in using the baseline level of each raw pulse (pulse without high-pass or band-pass filtering). The baseline level gives the relative temperature of the detector. The plot of the pulse amplitudes for a given energy as a function of the pulse baseline level gives a linear relation that is used to correct the correlation amplitude vs baseline level, and the temperature drift.

Some periods of the recording have strong temperature instabilities, mainly due to the liquid Helium transfer. The pulses detected during these periods are removed as shown in figure 26.
Rejection of the pile-up on the pulse decays

The extendable dead time rejects the pile-up on the pulse baseline; however, it does not reject the pile-up on the pulse decay. To reject these pile-up events, the $\chi^2$ values $\chi_{\text{comp}}$ are plotted as a function of amplitudes (figure 27). Large $\chi_{\text{comp}}$ values correspond to pulses having a shape deviating from the template shape, generally because of a pile-up on the pulse decay, so by rejecting pulses with large $\chi_{\text{comp}}$, these pile-up events are removed. However, the $\chi^2$ depends on the pulse energy, so the dependence must be taken into account. The curve $\chi_{\text{comp}}$ versus amplitudes $A$ is fitted with a quadratic function, $\chi_{\text{fit}} = a_\chi \cdot A^2 + b_\chi$; $\chi_{\text{fit}}$ corresponds to a mean value of the $\chi_{\text{comp}}$ at a given amplitude. Hence the $\chi^2$ limit to reject the pile-up is $\chi_{\text{limit}} = \chi_{\text{fit}} \cdot X \cdot \chi_{\text{fit}}$ (figure 27). Other parameters than listed before can be used to reject pile-up. For example, for some pile-up, the timestamps $t_{\text{max}}$ are shifted to higher values compared to $t_{\text{max}}$ of a single pulse. Or, the 10% to 90% rise time constant would be increased by some close pile-up. Nevertheless, these parameters are less efficient than the parameter $\chi_{\text{comp}}$ to remove pile-up.

Figure 216: Amplitudes of the 59.54 keV pulses as a function of time. Left, in red the spline function $f_{\text{amp}}(t)$ determined using the 59.54 keV amplitudes. Right amplitudes as a function of time after the drift correction and rejection of the periods of He transfer.

Figure 227: The $\chi$ values $\chi_{\text{comp}}$ as a function of the pulse amplitudes. The blue curve is $\chi_{\text{fit}}$, the fit with a quadratic function. The green curve is the $\chi$ value $\chi_{\text{limit}}$ above which the pulses are rejected.
Validation of the data analysis for the spectrum construction

In order to validate the data analysis, a recording was simulated with a noise spectrum and a pulse shape similar to experimental data. Four different pulse amplitudes were simulated with known probabilities and with a total count rate of 5 s⁻¹, which is relatively high in order to introduce a non-negligible probability of pile-up. The energy spectrum was obtained after the data analysis described in this section. The temperature drift was not implemented in the simulated data.

The table 5 summarizes the results between the two methods of pulse amplitude determination in frequency (FD) and time domain (TD). Concerning the pulse energy, after calibration using only one line, the measured energies are in good agreement with the simulated input energies meaning that both analyses do not distort the energy scale by introducing non-linearities. For the energy resolution, as expected, better resolution is obtained in the frequency domain; however, the analysis in TD degrades the resolution by only 15%. In the case of large pile-up probability, TD analysis would be a good compromise because the duration of the reduced template pulse is short, leading to a smaller dead time percentage and a smaller pile-up probability compared with FD analysis.

The probabilities measured at the different energies with the two methods are in very good agreement with the probabilities used to simulate the data. So, the digital signal processing of the data is validated.

Table 5: Results obtained from the spectrum of simulated data with optimal filtering in the frequency domain (FD) or with a reduced template pulse in time domain (TD). The results are compared with the input parameters introduced in the simulated data (Sim).

<table>
<thead>
<tr>
<th>Energy (eV)</th>
<th>FWHM resolution (eV)</th>
<th>Probability</th>
<th>Deviation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Simulated  TD  FD</td>
<td>Simulated  TD  FD</td>
<td>Simulated  TD  FD</td>
</tr>
<tr>
<td>4000</td>
<td>3999.7  3999.8</td>
<td>0.2500 (8)  0.2491 (8)  0.2488 (9)</td>
<td>-0.36  -0.48</td>
</tr>
<tr>
<td>15000</td>
<td>14999.9  14999.9</td>
<td>0.2500 (8)  0.2492 (8)  0.2492 (9)</td>
<td>-0.32  -0.32</td>
</tr>
<tr>
<td>50000</td>
<td>49999.9  50000</td>
<td>0.2500 (8)  0.2493 (8)  0.2499 (9)</td>
<td>-0.28  -0.08</td>
</tr>
<tr>
<td>100000</td>
<td>100000  100000</td>
<td>0.2500 (8)  0.2498 (8)  0.2506 (9)</td>
<td>-0.08  0.24</td>
</tr>
</tbody>
</table>

Data analysis at PTB

The output voltage of the Magnicon SQUID electronics is also amplified with an SR560 amplifier from Stanford Research Systems which is connected to an AlazarTech digitizer (ATS9462) with a 4-lane PCI express interface. It comprises two channels which are sampled at 16-bit resolution, each having a maximum sampling rate of 180 MS/s per channel with an input voltage ranging between ±200 mV and ±16 V. Additionally, an on-board acquisition memory with 720 MB/s data throughput to PC memory is used for continuous streaming mode.

The custom-built measurement routine developed using LabView controls the parameters of the digitizer. The signals are continuously saved as binary data by using the on-board memory. To achieve manageable data size a new data file is generated after a suitable period. The base temperature and a header including all relevant parameters are saved in separate files. A measurement typically takes one week. The spectrum is obtained offline by digital signal processing by python™ [7] using the file format hdf5 (hierarchical data format 5 [8]).

While the used DAQ hardware is different from the system used at CEA and the software uses its own implementation in a different programming language, the principle procedure is done in analogous way to the CEA approach and will not be described in detail.

The analysis algorithms have not been validated as has been done with the CEA implementation, but tests on short measurement segments have shown no noticeable problems in the resulting spectra. A histogram of the pulse energies of such a measurement of the 99Tc beta spectrum is depicted in figure 28, the calibration peak is clearly visible along with typical characteristics of the spectrum. A Full Width at Half Maximum (FWHM) Gaussian fit of the main peak of 241Am at 59.54 keV indicates that the local energy resolution is approximately ΔE = 590 eV.
Figure 28: Pulse histogram for a $^{99m}$Tc ($E_{\text{max}} = 293.8$ keV) source with an external calibration source (13 h test measurement, sample activity ~ 4 Bq). The calibration peak of $^{241}$Am at 59.54 keV is fitted with a Gaussian curve (inset).
8 Results

Within this project the beta spectra of $^{14}$C, $^{151}$Sm and $^{99}$Tc have been measured. Also, a first attempt of the more challenging measurement of the spectrum of $^{36}$Cl has been made at PTB with the fabrication of bilayer absorber arrays. In this section the results of the high-precision beta spectrometers based on MMC detectors are shown and compared between both independent systems.

8.1 $^{14}$C

The spectrum of $^{14}$C has been measured using a source prepared by conventional drop deposition, but from a carrier-free, high specific activity solution. After drying, the deposit was barely visible, even under an optical microscope, so the source can be considered to be of high quality. It was deposited on a 25 µm thick gold foil, thick enough to stop all beta particles up to the end point of the spectrum (156.5 keV). Since the $^{14}$C atoms are bound in a volatile organic compound (1,3-thiazole), diffusion welding was not a viable option for source enclosure. The foil with the source was just folded over and slightly pressed. Gold is highly ductile and keeps its shape once folded; no beta electrons can escape from being absorbed in the gold absorber. Gold is a good thermal conductor at very low temperature, so the thermal contact between the two halves of the absorber through the bending is sufficient. The absorber (Au, 1 mm$^2 \times (2 \times 25$ µm)) was placed on an MMC chip whose size best matches its heat capacity ($C_{abs} = 67$ pJ/K at 20 mK). It was fixed to the sensor by a small quantity of a low temperature compatible epoxy glue (Stycast 1266). To ensure a good thermal contact between absorber and sensor, very small glue drops are placed between the gold stems, taking care that the height of the drops exceeds the stem height (5 µm). Due to the thermal shrinkage of the glue the absorber is, at low temperature, pressed onto the gold stems and the metal-metal contact provides a high thermal conductivity between absorber and sensor.

The experimental conditions during the spectrum measurement were far from optimal. During the cooling phase, the glue layer fixing the MMC chip to its holder broke, the chip was then only suspended by the gold and aluminum bonding wires used for electric and thermal contacts. This had two consequences degrading the detector performance. One of them is that the thermal time constant of the detector was longer than expected, leading to a large fraction – more than 50% – of piled-up pulses that had to be removed from the data set. After 10 days of data acquisition at a rate of ~ 7 counts per second, the final spectrum, shown in figure 29, contains 2.7 million events.

![Figure 29: Beta spectrum of $^{14}$C measured with an MMC (blue). The lines at 22 keV, 25 keV and 88 keV are K X-ray and gamma ray lines from an external $^{109}$Cd source and the broad line at around 6 keV, a clipped double line, is the K $\alpha + \beta$ line of an external $^{55}$Fe source, these lines are used for energy calibration. The weak lines at around 10 keV are escape lines. The red line is a theoretical spectrum calculated with the code BetaShape [9,10]. The increase of the experimental spectrum at low energies may be due to the degraded detector performance; a new measurement under improved conditions should clarify this question.](image-url)
The other consequence is that the MMC was subject to vibrations, resulting in an energy resolution near 200 eV (FWHM), about a factor 5 worse than expected from the absorber heat capacity. Nevertheless, the detector performance was much better than in the measurement published in [1]: The energy resolution was improved by a factor five, while the energy threshold was reduced from ~ 5 keV to ~ 700 eV. The measurement will be repeated after regluing the MMC chip to fully exploit the performance that can be expected from the MMC.

8.2 $^{151}$Sm

A $^{151}$Sm source was electrodeposited on a 10 µm thick silver foil. The resulting samarium oxide/hydroxide layer was blackish and was considered not to be ideally thin. Therefore, the mechanical processing consisting in repeated folding and laminating of the metal foil with the radioactive material, as described in section 5.6, was applied to the silver foil with the $^{151}$Sm source. A piece with an expected activity of ~ 7 Bq (0.8 mm × 0.8 mm × 7 µm) was cut from this source foil and was sandwiched between two silver foils (0.9 mm × 0.9 mm × 15 µm each) and the three foils were diffusion-welded to form the absorber (heat capacity: 29 pJ/K at 20 mK). Figure 30 shows the silver absorber containing the $^{151}$Sm source on the MMC chip together with the SQUID chip. The performance of the MMC during this measurement was as expected. An energy resolution ranging from about 45 eV (FWHM) at 6 keV to 70 eV at 25 keV and an energy threshold of 250 eV were observed. It should be mentioned here that under optimal conditions the energy resolution of an MMC is nearly energy-independent since it is only limited by noise. Various effects like temperature fluctuation of the thermal bath can, however, add some energy-dependent terms. The thermal time constant (1/e) of the detector was 460 µs; at a count rate of 8.7 s$^{-1}$, the fraction of piled-up pulses was very low. After 14 days of data acquisition, the final spectrum contains 10.2 million events. $^{151}$Sm is the only non-pure beta emitter measured within the MetroBeta project: It has a main $\beta^-$ decay branch ($Q_\beta = 76.3$ keV) to the ground state and a second $\beta^-$ decay branch to the 21.54 keV excited level of $^{151}$Eu. Both transitions are first forbidden non-unique. The recommended values for the respective probabilities of the two decay paths are 99.07 (4) % and 0.93 (4) % [11]. The de-excitation of the 21.54 keV excited state is highly converted; only 3.4% of the gamma transition lead to the emission of gamma-rays (total probability: 3.24 (13) × 10$^{-4}$), the rest leads to the emission of conversion electrons and subsequently X-rays and/or Auger electrons. The detector absorber with its given dimensions, fixed to stop all beta electrons up to the beta $Q$ value, does also stop all conversion electrons, more than 99% of all X-rays and more than 95% of the 21.54 keV gamma photons. The result is that for practically all beta decays to the excited level the sum of the beta energy and the gamma energy is absorbed. So, the measured spectrum for the decays to the excited level is shifted by the energy of the gamma transition and starts at 21.54 keV, leading to a step in the recorded overall spectrum.

![Figure 30: MMC chip with the silver absorber enclosing an $^{151}$Sm source. The SQUID chip and electrical contacts are also shown.](image-url)
Since the maximum energy for this beta branch equals the $Q$ value minus the gamma transition energy, the end point of both measured spectra is the same, 76.3 keV. As it is not possible to distinguish events from the two decay branches, both spectra are superimposed in one experimental spectrum.

The measured spectrum is shown in figure 31 together with theoretical spectra calculated with the code BetaShape for both decay paths. The spectrum corresponding to the decay to the ground state was fitted to the experimental spectrum in the energy range from 10 keV to 20 keV. It can clearly be seen that above 21.5 keV the experimental spectrum lies higher than the fitted spectrum of the main decay branch. The spectrum corresponding to the decay to the excited level was shifted by 21.54 keV and fitted to the experimental spectrum in the energy range 26 keV – 40 keV. The area lying between the two theoretical spectra, corresponding to the probability of the decay to the excited state of $^{151}$Eu, amounts to 2.6% of the total. We do not state any uncertainty on the measured probability, firstly because the theoretical spectra are preliminary, secondly because the theoretical spectrum does not fit the experimental spectrum below 6 keV, and thirdly because the fitting procedure was rather coarse. Nevertheless, this value is in clear contradiction with the recommended value for the probability of the decay to the $^{151}$Eu excited level. Concerning the beta spectrum shape, the discrepancy between experiment and theory at low energies is most likely due to an incomplete control of the atomic effects in the theoretical calculation of this first forbidden, non-unique transition.

![Figure 31: Beta spectrum of $^{151}$Sm measured with an MMC (blue) together with theoretical spectra calculated for the two beta decay branches, to the ground state (red) and to the 21.54 keV excited level of $^{151}$Eu (green). The spectrum from the transition to the excited state is shifted towards higher energies by 21.54 keV, the energy of the gamma transition that is detected in sum with the beta particle energy. Therefore, both measured spectra end at the same energy, 76.3 keV. The energy calibration was performed with an external X-ray source composed of $^{55}$Fe and $^{109}$Cd.](image)

8.3 $^{99}$Tc

The beta spectrum of $^{99}$Tc was measured both at PTB and at CEA, with sources prepared by different techniques, in different cryogenic setups and different detector modules, with different data acquisition systems and using different data analysis routines.

At CEA, a $^{99}$Tc source was electrodeposited on a 10 µm thick gold foil. The deposit is extremely thin and should be metallic technetium. However, the deposition yield and the resulting activity per surface area were lower than expected. Therefore, a sufficiently large piece of this foil had to be folded three times to reduce its area to a size (~0.5 mm × 0.7 mm) small enough to enclose it in an MMC absorber. This source foil was then sandwiched between two gold foils (0.9 mm × 0.9 mm × 74 µm each) and this stack was diffusion welded.
The final absorber has a heat capacity of 350 pJ/K at 20 mK, much larger than the previous detectors. The pulses had a rise time (10% - 90%) of 14 µs and a decay time (1/e) of 2.15 ms. Data was acquired during 13.7 days and the spectrum contains 5.65 million events. The energy resolution is practically energy-independent, about 100 eV (FWHM) up to 384 keV, the highest energy gamma line of a $^{133}$Ba source used for energy calibration and check of the linearity. Comparing the measured and the tabulated line energies between 31 keV and 384 keV shows no larger deviations than 70 eV, less than the energy resolution, and no obvious trend.

At PTB, a $^{99}$Tc source was prepared with a micro-drop dispenser directly on a 90 µm thick gold foil formatted to an array of absorber elements with lateral dimensions of about 1.6 mm and 0.7 mm, shown in Fig. 2a. An identical foil was diffusion welded onto the first foil with the dried radioactive material. One of the larger source/absorber assemblies with an expected activity of about 5 Bq was selected and glued onto a matching MMC. The heat capacity of the absorber assembly is 545 pJ/K at 20 mK. The observed pulses had a rise time (10% - 90%) of 31 µs and a decay time (1/e) of 4.6 ms. The data acquisition took 42 h and the resulting spectrum consisted of 0.5 million events with an energy threshold of about 5 keV. A $^{57}$Co source was used for energy calibration and the 122 keV gamma line showed an energy resolution of 600 eV (FWHM). The larger absorber and total heat capacity of the setup, as well as experimental problems with the temperature stability of the thermal bath explain the degraded energy resolution and threshold compared to the measurement performed at CEA. Figure 32 shows a superposition of both experimental spectra. It is clear that the spectrum shape is strictly the same. This spectrum shape will be a valuable input for the improvement of the theoretical calculation for this type of transition, 2nd forbidden non-unique. A theoretical spectrum calculated with the current version of the code BetaShape is also shown in figure 32. The improvement of this code, in particular for higher order forbidden, non-unique transitions, is also part of the MetroBeta project.
References


