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# Development of total decay energy spectrometry of $\alpha$ -emitting radionuclides using Metallic Magnetic Calorimeters

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## Abstract

Total decay energy spectrometry with cryogenic detectors is a promising technique for radionuclide analysis of  $\alpha$ -emitting nuclides. The radioactive sample is embedded in the detector absorber and the total decay energy for each disintegration is measured as a temperature elevation. We are developing this technique with Metallic Magnetic Calorimeters (MMCs).

The main condition of this technique is a detection efficiency close to unity. However, some  $\alpha$ -emitting nuclides emit intense  $\gamma$ -rays that can partially escape from the absorber. So a feasibility study for several nuclides has been carried out based on Monte Carlo simulations of the detection efficiency and numerical calculations of the expected energy resolution to identify the radionuclides that can potentially be measured. Furthermore, an MMC prototype has been built and tested. The total decay energy spectrum of Po-210 was measured and a FWHM energy resolution of 1.25 keV at 5.4 MeV was obtained and a Gaussian width at half maximum of 0.827 (5) keV. The baseline FWHM energy resolution is 207 eV, consistent with the resolution obtained on low energy L X-rays in the same spectrum at 14 keV.

**Keywords** Total decay energy spectrometry • Q spectrometry • Metallic Magnetic Calorimeters • Po-210

## 1 Introduction

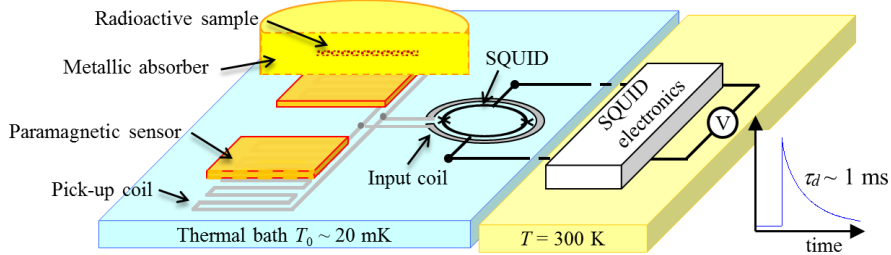
The spectrometry of the total decay energy (or  $Q$  spectrometry) of  $\alpha$ -emitting nuclides is a promising alternative to  $\alpha$  spectrometry [1,2]. It consists in enclosing the radioactive sample in the spectrometer absorber in order to absorb all the particles emitted during the radioactive decay and to measure the summed energy, i.e. the characteristic  $Q$ -value of  $\alpha$ -emitting

nuclides present. Compared with the complex spectra from  $\alpha$  spectrometry, the corresponding  $Q$  spectrum becomes very simple. For a mixed radionuclide spectrum only one peak appears per radionuclide and these are generally spaced by at least a few tens of keV. The count rate per peak is equal to the activity of the corresponding nuclide. However, three conditions are indispensable: an energy resolution better than the energy differences of the  $Q$ -values of the nuclides present, a detection efficiency close to unity and a source allowing for quasi complete thermalization of the decay energy. Cryogenic detectors are suitable for  $Q$  spectrometry because they are able to provide very high energy resolution and it is convenient to enclose sources in their absorbers for a high detection efficiency [1-3].

In this work we will show the expected performances of MMCs for  $Q$  spectrometry in terms of calculated energy resolution and simulated detection efficiency for 25  $\alpha$ -emitting nuclides<sup>1</sup>. In addition, the performance of an MMC prototype will be presented for the measurement of a Po-210 source.

## 2 Detection principle of $Q$ spectrometry with MMCs

The radioactive sample is enclosed in a metallic absorber with heat capacity  $C_a$ , itself strongly thermally coupled with a planar paramagnetic sensor with heat capacity  $C_s$  [4]. The sensor is inductively coupled with a meander shaped pick-up coil connected with another pick-up coil to form a superconducting closed circuit. A supercurrent flows in the circuit and locally magnetizes the sensor. The circuit is connected with the input coil of a current sensor SQUID to form a flux transformer.



**Fig. 1** Schematic of the metallic magnetic calorimeter (MMC) and its read-out circuit.

<sup>1</sup> The  $\alpha$ -emitting nuclides were chosen based on not having too short a half-life compared with the experiment duration of between a few hours and several days. Very long half-life nuclides would probably be problematic due to the large amount of material required to provide sufficient activity, but the problem of source preparation is not considered in this paper.

## Development of total decay energy spectrometry with MMCs

After a radioactive decay, the emitted particles are absorbed in the absorber and nearly the entire  $Q$ -value energy  $E_Q$  is converted to heat. The absorber and the sensor are rapidly in thermal equilibrium with a time constant  $\tau_r$ . The temperature rise,  $\delta T = E_Q / (C_s + C_a)$ , produces a change of magnetization  $\delta m$  measured as a current variation  $\delta i$  in the flux transformer and converted to a voltage change by the SQUID. The detector recovers its temperature equilibrium with the thermal bath at temperature  $T_0$  with a time constant  $\tau_d$  given by the thermal conductance of a weak metallic thermal link between the sensor and the bath.

### 3 Simulated and calculated performances of MMCs

#### 3.1 Monte Carlo simulations of the detection efficiency

A detection efficiency close to unity means nearly complete absorption of the energies of the emitted particles per decay. The particles emitted during the  $\alpha$  decay are approximatively: a 5-MeV  $\alpha$  particle, a 100-keV recoil nucleus, electrons mainly below a few tens of a keV, and X-ray and  $\gamma$ -ray photons up to a few hundred keV. While about 10  $\mu\text{m}$  of gold are sufficient to absorb the  $\alpha$  particle, the recoil nucleus and the electrons, larger thicknesses are needed to absorb the photons. Because the absorber volume must be kept as small as possible to minimize the heat capacity  $C_a$  and to achieve the best energy resolution, the minimal thicknesses of gold absorbers for detection efficiencies of 99% and 99.9% were determined by Monte Carlo simulations with the Penelope 2014 code [5].

Since the solid angle is  $4\pi$ , the emitted particles per decay are detected in coincidence, so the cascade of the particles and their energies must be taken into account in the efficiency simulation. The subroutine PenNuc has been used to generate the cascade used as input in the Monte Carlo code [6]. The efficiency per decay was simulated for 16 absorber thicknesses between 20  $\mu\text{m}$  and 6 mm. In the simulated geometry, the source with an active area of 1  $\text{mm}^2$  is placed in the middle of the absorber. The cylindrical absorber has a diameter of 2 mm, except for thick absorbers for which the diameter is given by source diameter plus the absorber thickness in order to limit the escape of particles from the side of the absorber.

Table 1 shows the minimum thicknesses, obtained by interpolation or extrapolation of the simulated results, for the different radionuclides to reach a detection efficiency of 99% and 99.9%. For U-235, Pa-231, Ra-223 and Cm-243, the absorber dimensions become very large with gold absorbers thicker than 1 mm and with heat capacities larger than 10 nJ/K. For Ra-223,

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the alternative could be to measure one of the daughters in secular equilibrium, i.e. Po-215 for which absorbers of only 20  $\mu\text{m}$  thickness are needed. For Po-209, we can notice that an efficiency of 99% is obtained with only 20  $\mu\text{m}$  thick absorber, but an efficiency of 99.9% cannot be reached with 6 mm thick absorbers because of the 261 keV  $\gamma$ -ray with an intensity of 0.25%.

Table 1. Gold absorber thicknesses and energy resolutions calculated at 20 mK for a given detection efficiency for different actinides.

Actinide	Decay Data		For an efficiency of 99%			For an efficiency of 99.9%		
	$Q$ value (keV)	Half-live	Au absorber thickness ( $\mu\text{m}$ )	Absorber capacity (nJ/K)	FWHM (eV)	Au absorber thickness ( $\mu\text{m}$ )	Absorber capacity (nJ/K)	FWHM (eV)
Th-232	4082	$1.40 \times 10^{10}$ y	20	0.0897	27	28	0.125	31
U-238	4270	$4.47 \times 10^{09}$ y	20	0.0897	27	31	0.139	33
U-236	4573	$2.34 \times 10^{07}$ y	20	0.0897	27	34	0.152	34
<i>U-235</i>	4678	$7.04 \times 10^{08}$ y	3920	107	1713	<b>6440</b>	<b>399</b>	<b>5370</b>
U-234	4858	$2.46 \times 10^{05}$ y	20	0.0897	27	38	0.171	37
Ra-226	4871	1600 y	20	0.0897	27	356	1.60	113
Np-237	4958	$2.14 \times 10^{06}$ y	784	3.52	173	1972	19.5	490
<i>Po-209</i>	4979	115 d	20	0.0897	27	<b>&gt; 6000</b>	-	-
Pu-242	4985	$3.73 \times 10^5$ y	20	0.0897	27	31	0.139	33
<i>Pa-231</i>	5150	32670 y	<b>5400</b>	<b>248</b>	<b>3511</b>	<b>8920</b>	<b>987</b>	<b>12605</b>
Pu-239	5245	24100 y	20	0.0897	27	23	0.102	29
Pu-240	5256	6561 y	20	0.0897	27	31	0.137	33
Po-210	5407	138.4 d	20	0.0897	27	20	0.0897	27
U-232	5414	70.60 y	20	0.0897	27	44	0.197	39
Am-243	5439	7367 y	548	2.46	142	1448	9.72	314
Cm-246	5477	4723 y	20	0.0897	27	38	0.170	36
Th-228	5520	1.913 y	20	0.0897	27	442	1.98	127
Pu-238	5593	87.74 y	20	0.0897	27	37	0.167	36
Am-241	5638	432.6 y	234	1.05	91	814	3.65	177
Cm-244	5902	18.11 y	20	0.0897	27	42	0.190	38
Ac-225	5935	10 d	880	3.950	185	<b>7400</b>	<b>586</b>	<b>7677</b>
<i>Ra-223</i>	5979	11.43 d	<b>6280</b>	<b>372</b>	<b>5035</b>	<b>10360</b>	<b>1500</b>	<b>18896</b>
<i>Cm-243<sup>a</sup></i>	6169	28.9 y	<b>6400</b>	<b>392</b>	<b>5282</b>	<b>7920</b>	<b>706</b>	<b>9152</b>
Cm-242	6216	162.9 d	20	0.0897	27	39	0.176	37
Cf-252	6217	2.65 y	20	0.0897	27	41	0.183	38
Po-215 <sup>b</sup>	7526	1.78 ms	20	0.0897	27	20	0.0897	27

<sup>a</sup>The simulated efficiencies of Cm-243 do not take into account the small electron capture branch of 0.3%.

<sup>b</sup>Po-215 has a short half-life compared with the measurement duration, but it is in equilibrium with Ra-223 and can be used to quantify it.

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### 3.2 Expected energy resolutions

Once the required absorber thicknesses for given detection efficiencies have been determined, the corresponding energy resolutions are calculated from the expected signal to noise ratios (SNR). Here the broadening of the energy resolution due to the creation of defects in the absorber from the recoil nuclei or the  $\alpha$ -particles is not taken into account, a contribution of about 1 keV is expected for noble metals [7]. The SNR depends on many parameters related to the thermodynamic properties of the sensor and the absorber, the thermal conductances (time constants), the read-out circuit and the noise sources. For a given absorber heat capacity and a given thermal bath temperature ( $T_0 = 20$  mK), the optimum parameters that maximize the SNR can be derived by rather complex numerical calculations [4]. To simplify the calculations, the same SQUID parameters are used for all the absorbers<sup>2</sup>. In addition, complete numerical optimizations have shown that the best energy resolution is obtained when the sensor heat capacity is close to the one of the absorber [4], thus the sensor area has been scaled in order to equalize the heat capacities,  $C_a = C_s$ . Then the meander pick-up coil area is scaled to be equal to the sensor area, while the meander strip line dimensions (pitch and width) are kept constant, and the meander inductance is proportional to the sensor area (2.8 nH/mm<sup>2</sup>). For such a configuration, the optimum permanent current is between 60 and 70 mA.

The pulse rise time and decay time constants are fixed to 50  $\mu$ s and 1 ms respectively. In the numerical calculations, the SQUID noise, the sensor one-over- $f$  noise and the noise from the thermodynamic fluctuations are taken into account. The calculated FWHM energy resolutions are presented in Table 1. For all radionuclides, the resolution is smaller than the difference between their  $Q$ -values and the  $Q$ -values of adjacent nuclides. However, some require very large gold absorbers, and the resulting energy resolution is no longer advantageous compared with semiconductor detectors.

## 4 Experimental performances for $Q$ spectrometry with MMCs

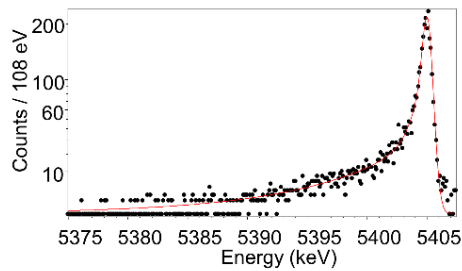
In order to study the performances of the MMCs independently of the source quality, a  $Q$  spectrum was measured with a Po-210 source with an activity of 0.2 Bq self-deposited on a silver foil and embedded between two silver layers of 4 mm<sup>2</sup> x 41  $\mu$ m. Such a deposit is very thin and should not degrade the energy resolution, since the quality of the source deposit is known to

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<sup>2</sup> Two stage SQUID from Magnicon GmbH with an input coil inductance of 27 nH

potentially degrade the energy resolution [2]. In a first approach, the absorber was directly glued onto a sensor of  $1.3 \times 1.3 \text{ mm}^2$ . However, with such a configuration, the pulses cannot be measured properly because the expected pulse height of  $18 \Phi_0$  for an energy absorption of  $5.4 \text{ MeV}$   $Q$ -value, combined with a rise time of  $4.2 \mu\text{s}$ , was not compatible with the SQUID electronics slew rate of about  $1 \Phi_0/\mu\text{s}$ . Therefore, the rise time has been adapted by adding two stacks of  $1 \text{ mm}^2$  of ( $3 \mu\text{m}$  Stycast +  $3 \mu\text{m}$  Au) between the absorber and the sensor that reduces the thermal conductance. The rise time was increased to  $46 \mu\text{s}$  and pulses of  $18 \Phi_0$  were successfully measured. The  $Q$  spectrum measured at  $11 \text{ mK}$  is shown in Fig. 2.

The FWHM of the Po-210  $Q$  peak is  $1.25 \text{ keV}$ , however a left tail is clearly present. The peak was fitted with a Gaussian function with two left tails to take into account the tail and the Gaussian broadening. The resulting Gaussian width at half maximum is  $827 (5) \text{ eV}$ . The detector has also been characterized with an external Cm-244  $\alpha$ -source, but the source quality was not sufficient to obtain similar energy resolutions on the alpha peaks. The L X-rays emitted by the Cm-244 allow us to measure the energy resolution in the low energy range; FWHM of  $185$  and  $194 \text{ eV}$  are observed respectively on  $L\alpha_1$  ( $14.3 \text{ keV}$ ) and  $L\beta_1$  ( $18.3 \text{ keV}$ ) which are consistent with the baseline energy resolution of  $207 \text{ eV}$ . The latter is five times worse than the calculated baseline energy resolution of this detector. However, the experimental noise was limited by the acquisition card; taking into account the excess noise leads to a factor two between the calculated and the measured baseline resolutions. Even a resolution two times worse than calculated is largely sufficient to measure the radionuclides in Table 1.



**Fig. 2** Experimental  $Q$  spectrum of Po-210 (*black dots*) and the fit with a Gaussian with two left tails (*continuous red line*). (Color figure online)

The non-linearity has been characterized from  $14.3 \text{ keV}$  to  $5.8 \text{ MeV}$  using various X-ray and  $\alpha$ -particle lines. A small deviation of  $2.2\%$  is observed at  $5.8 \text{ MeV}$ , meaning a calibration at  $\sim 5 \text{ MeV}$  is necessary to allow correct radionuclide identification for the foreseen applications. The

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calibration could be achieved with an external  $\alpha$ -particle source or by embedding an  $\alpha$ -emitting tracer.

In summary, we have demonstrated that MMCs can be potentially used to quantify a large number of  $\alpha$ -emitting nuclides by  $Q$  spectrometry, meaning that high energy resolution, combined with a detection efficiency close to unity, can be obtained. Nevertheless, an important point remains to be studied: the source preparation and the nature of the radioactive deposit in the absorber which can both have a major impact on the energy resolution and must be considered in future investigations.

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