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Beta spectrometry with metallic magnetic calorimeters

M. Loidl *, M. Rodrigues, C. Le-Bret, X. Mougeot

LNE—Laboratoire National Henri Becquerel, CEA Saclay, DRT/LIST/DM2I, Gif-sur-Yvette 91191, France

HIGHLIGHTS

- The beta spectrum of ^{63}Ni was measured with metallic magnetic calorimeters.
- Dried sources are not adequate, electroplated sources yield accurate spectra.
- The atomic exchange effect was confirmed down to very low energy (200 eV).

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ABSTRACT

Metallic magnetic calorimeters are a specific type of cryogenic detectors that have been shown to enable precise measurement of the shape of low energy beta spectra. The aim of their use at LNHB is the determination of the shape factors of beta spectra. The beta source is enclosed in the detector absorber, allowing for very high detection efficiency. It has turned out that the type of source is of crucial importance for the correctness of the measured spectrum. Spectra of ^{63}Ni measured with several sources prepared by drying a $NiCl_2$ solution differ from one another and from theory, whereas spectra measured with electroplated sources are reproducible and agree with theory. With these latter measurements we could confirm the atomic exchange effect down to very low energy (200 eV).

1. Introduction

Since several years the LNHB is developing metallic magnetic calorimeters (MMCs) for beta spectrometry. The aim is the precise determination of shape factors. Metallic magnetic calorimeters (Enss et al., 2000; Fleischmann et al., 2005) are a specific type of cryogenic detectors that may be adapted to soft and hard X-ray, alpha, beta, and optical photon detection. For beta spectrometry, the source is enclosed in the detector absorber. Since the solid angle is 4π sr and the absorber size can be chosen as a function of the endpoint energy of the beta emitter, a detection efficiency of practically 100% can be realized for the whole spectrum, starting from a very low energy threshold typical for these detectors.

Several years ago, the spectrum of ^{241}Pu was measured with a MMC at LNHB (Loidl et al., 2010). A discrepancy between the experimental and a theoretical spectrum raised the question whether this discrepancy reflects a distortion of the experimental spectrum whose reason would need to be revealed, or is due to an insufficiency of the calculation of the theoretical spectrum: ^{241}Pu decays via a first forbidden, non-unique transition, and the spectra of forbidden, non-unique transitions are not straightforward to calculate. Consequently, the decision was taken to validate the potential of MMCs for beta spectrometry by measuring the spectrum of an allowed transition that can be calculated more reliably for comparison with the experimental spectrum. The pure beta emitter ^{63}Ni was chosen; its half life is 98.7 (24) years and the endpoint energy 66.980 (15) keV. This low energy beta emitter was chosen also because in this energy range the potential energy loss due to escape of Bremsstrahlung photons from the detector can be safely neglected, as has been confirmed by Monte Carlo simulation. This makes the validation of the experimental method more reliable because no correction for this energy loss needs to be applied. The aim of this development remains of course the determination of the shape factors particularly for beta emitters decaying via forbidden transitions, whose spectra are difficult to calculate and often experimentally not well known, and up to endpoint energies around 1 MeV. For the energy range ~500 keV to several MeV, a new beta spectrometer based on semiconductor detectors is currently being implemented at LNHB (Bisch et al., 2013).

2. Metallic magnetic calorimeters for beta spectrometry

Like most cryogenic detectors, metallic magnetic calorimeters (MMCs) are thermal detectors: the energy E of each individual particle is measured as a temperature rise $\Delta T = E/C$ where C

* Corresponding author. Tel.: +33 1 69084277; fax: +33 1 69082619.
E-mail address: martin.loidl@cea.fr (M. Loidl).

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denotes the total heat capacity of the detector. Since the specific heat of most materials strongly decreases with temperature, the signal ΔT is maximized when operating the detector at very low temperature: usually 10–50 mK. At the same time, in this temperature range thermodynamic fluctuation noise and other thermal noises are very small, hence a very high signal-to-noise ratio of MMCs is achievable, an essential condition for low threshold and high energy resolution. A thermal link connects the detector to a heat sink kept at constant temperature in order to restore the initial temperature after each particle interaction. The signal decay time τ is determined by the thermal conductance G of the link: $\tau = C/G$. Decay times of a few hundred μs to several ms are typical, allowing for count rates of order 10 s^{-1}.

Mostly MMCs have a metallic particle absorber, often made of gold, and the temperature rise is measured by a metallic paramagnetic thermometer in good thermal contact with the absorber. The name "*metallic* magnetic calorimeters" refers to the metallic nature of the thermometer that is necessary for a fast response. The thermometer is composed of gold and a small concentration of erbium (of order 0.1%), and its magnetization in the presence of a magnetic field (typically a few mT) strongly varies with temperature. This variation upon a particle interaction and the consecutive temperature rise is measured with a SQUID (Superconducting Quantum Interference Device), a very low noise magnetometer also operating at cryogenic temperatures.

For beta spectrometry, the beta emitter is enclosed inside the absorber: it can either be deposited on a metal foil constituting half of the absorber; it is then covered by a foil forming the second half of the absorber. Or it can be deposited on a separate thin metal foil that is then sandwiched between the two absorber foils. In order to tightly enclose the source, the metal foils are diffusion welded to each other. The minimum absorber thickness necessary to fully stop all beta particles is determined by Monte Carlo simulation. This way, the detection efficiency is virtually 100% between the detection threshold and the endpoint of the spectrum. The area of the absorber foils depends on the area required to deposit the beta emitter. Mostly 1 mm^2 is sufficient.

Care must be taken to exclude all possible sources of distortion of the experimental spectra, like the following:

(a) Detector non-linearity
The linearity of the MMCs, including electronics and data acquisition system, has been tested and is very good, see Section 4.1.

(b) Pile-up
The count rate, i.e. source activity, must be chosen in dependence of the signal decay time in order to keep pile-up at a very small level. It can be further reduced by application of dead time and careful pulse shape analysis. Any remaining unresolved pile-up would not be noticed within the spectrum and could cause spectrum distortion, but it would show up beyond the endpoint of the spectrum. In the measurements presented in this paper it is negligible.

(c) Energy loss by escape of Bremsstrahlung photons
This issue has been checked by Monte Carlo simulation. For the low energy beta emitters presented in this paper it is negligible, but for higher energy emitters (maximum energy several hundred keV) it must be corrected for.

(d) Atomic excitation
A small part of the beta energy can be expended by excitation of the electrons of the daughter atom: shake-up or shake-off. This effect can distort the spectrum if the source is external to the detector. By enclosing the source in the absorber, this fraction of the beta energy is also detected in the form of heat and integrated in the total measured energy.

(e) Self-absorption
Partial loss of beta energy in the source is one of the most common causes of spectrum distortion in beta spectrometry. With the source enclosed in the absorber of a MMC, one would expect that this part of energy will also be detected in the form of heat. However our measurements have revealed that this is not always true, see Section 4.1. The influence of the source turned out to be the most critical issue in beta spectrometry with MMCs.

3. Exchange effect

The exchange effect is an atomic effect that is mostly not taken into account in the calculation of beta spectra, although for some nuclides it has a stronger influence than the screening effect, the nuclear size effect or the radiative corrections. It corresponds to the case that a beta electron is not directly emitted to the continuum, but created into a bound orbital of the daughter atom, accompanied by the simultaneous emission of a bound electron from the same orbital to the continuum. The exchange effect has an influence on the spectrum shape: it enhances the global beta emission probability, with the enhancement increasing towards low energies.

The first theoretical description of the exchange effect, taking into account only the 1s electrons, predicted a reduced emission probability at low energy (Bahcall, 1963). A more complete treatment, including higher orbitals, came to the contrary conclusion that the emission probability increases at low energies (Harston and Pyper, 1992). Experimental evidence has been reported only twice up to now (Lowry et al., 1993; Angrave et al., 1998). These measurements were also carried out with cryogenic detectors (of different type than MMCs), but with energy thresholds of several keV where the influence of the exchange effect is still relatively small. Both of them observed en enhancement of the emission probability at low energies. MMCs can probe the exchange effect down to much lower energies.

The evaluation of the exchange effect has been implemented in the code BetaShape, developed at LNHB, which in its present form calculates the shape of allowed and unique forbidden transitions. Details on the formalism and computational approach for the calculation of the exchange effect are given in Mougeot et al. (2012).

4. Measurements of the beta spectrum of ^{63}Ni

4.1. Dried sources

Several MMCs were made with ^{63}Ni sources prepared by drying a small drop of $NiCl_2$ solution. The linearity of the detectors was checked using the gamma and X-rays of a combination of ^{55}Fe, ^{109}Cd and ^{241}Am sources. A linear fit was applied to the measured positions of X- and gamma-ray lines as a function of the tabulated line energies. In the energy range 5.9–88 keV covered by the photon sources the maximum deviation of the measured line positions from the linear fit was 0.1% and no trend of the deviations with energy was observed. So the detectors can be considered as linear. For the final spectra only the 59.5 keV gamma line of ^{241}Am was used for energy calibration.

Fig. 1 shows several of the experimental spectra together with theoretical spectra calculated with and without the exchange effect using the code BetaShape. All experimental spectra differ from one another and from theory. It is clear, however, that the agreement with the theoretical spectrum including the exchange effect is much better than neglecting the exchange effect. The reason for the differences between the experimental spectra is not clear. The most likely explanation appears to be that part of the