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First measurement of the beta spectrum of ^{241}Pu with a cryogenic detector

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ABSTRACT

The LNE-LNHB is developing metallic magnetic calorimeters, a specific type of cryogenic detectors, for beta spectrometry. The aim is the determination of the shape factors of beta spectra. Our latest detector has been designed to measure the spectrum of ^{241}Pu, a pure beta emitter with an endpoint energy of 20.8 keV. In this paper, the detection principle of metallic magnetic calorimeters is explained and a detailed description is given of the realization of the detector enclosing a ^{241}Pu source inside the detector absorber. A spectrum resulting from our first measurement is shown and compared with a theoretical spectrum.

1. Introduction

We are exploring the potential of metallic magnetic calorimeters (Enss et al., 2000; Fleischmann et al., 2005) for beta spectrometry, with the aim of determining the shape factors of beta spectra. We are particularly interested in the spectra of pure beta emitters decaying via forbidden transitions, whose spectra are difficult to calculate and often experimentally not well known. Beta spectra are generally difficult to measure with lithium drifted silicon (Si:Li) detectors or electrostatic or magnetic spectrometers due to low detection efficiency, non-linear response of the detector, or energy loss of the electrons in the source or in the dead layer at the surface of Si:Li detectors.

Metallic magnetic calorimeters (MMCs) are cryogenic detectors (LTD-12, 2008; LTD-13, 2009), usually operated at temperatures between 10 and 50 mK. We embed the beta emitter inside the detector absorber so as to realize a solid angle of 4π. So even if beta particles loose (part of) their energy in the source, the energy remains contained in the detector and will be detected. By choosing the absorber dimensions according to the maximum energy of the beta spectrum, a detection efficiency $> 99\%$ can be realized over the entire spectrum, starting from a threshold at around 1% of the endpoint energy. Such a low threshold can be achieved with carefully designed MMCs for endpoint energies from a few keV (the lowest existing) up to about 1 MeV. It has been demonstrated that the linearity of MMCs can be better than 0.5% over two orders of magnitude in energy (Rodrigues et al., 2008). We believe that MMCs are a very powerful means to reliably determine the shapes of beta spectra.

Our latest detector has been designed to measure the spectrum of ^{241}Pu, a pure beta emitter with an endpoint energy of 20.8 keV (Audi, 2003). This nuclide is present in nuclear reactor cores and nuclear waste. As for most pure beta emitters, its activity is commonly measured by liquid scintillation counting. This technique requires the precise knowledge of the spectrum shape in order to determine the detection efficiency. The spectrum of ^{241}Pu is not very well known. The results presented here make us confident that MMCs will contribute to a better knowledge of this and other beta spectra. Finally, precise determination of shape factors and comparison with calculated spectra may contribute to a better theoretical understanding of beta decay.

2. Detection principle of metallic magnetic calorimeters

In a metallic magnetic calorimeter, the energy of a particle is measured in the form of a temperature rise. Within a wide energy range, this temperature rise is proportional to energy. The detector is composed of a metallic absorber (mostly gold) in tight thermal contact with a paramagnetic thermometer (gold containing a dilute concentration of erbium, Au:Er). A magnetic field is applied to magnetize the thermometer. The energy deposited by a particle in the detector absorber is transferred to the metal's conduction electrons, resulting in a temperature rise of the absorber and consecutively of the thermometer. The consequence is a decrease of the thermometer's magnetic moment. This change in magnetic moment is measured very precisely by a SQUID magnetometer. A SQUID (superconducting quantum interference device) is a magnetic flux-to-voltage converter realized in thin film technology and is particularly well matched to the measurement of the small change of magnetic moment of the thermometers of MMCs. After each particle interaction, the detector returns to its initial temperature via a weak thermal link to a heat

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sink (thermal bath). Hence, the detector does not integrate an incident power, but determines the energy of each individual particle. The pulse shape of the individual signals can be approximated in first order by an exponential rise time of 1 to a few microseconds, given essentially by the diffusion time of the heat within the detector, and an exponential decay time of the order of a millisecond, corresponding to the return of the detector to its equilibrium temperature.

The magnitude of the temperature rise is inversely proportional to the detector's heat capacity, which decreases with its temperature. Therefore at very low temperature the amplitude of the signal becomes relatively large, whereas the thermal noise is very low. Metallic magnetic calorimeters, usually being operated at temperatures below 50 mK, offer very high signal-to-noise ratios.

3. Experimental setup

A ^{241}Pu source has been sandwiched between two 12 μm thick gold foils serving as absorber. A much smaller thickness of gold ($<$1 μm) would be sufficient to stop all beta particles. But in a very thin foil the heat diffusion would be slower. This could lead to a dependence of the pulse rise times on the position of the particle interaction within the detector, and consequently to an inhomogeneous response of the detector. The source was deposited as a small drop (0.5 mg) of a plutonium nitrate solution on one of the gold foils. Prior to the deposit, we have carried out a chemical separation of ^{241}Pu from ^{241}Am, whose alpha emission would have strongly perturbed the measurement. The source activity is approximately 8 Bq. To delimit the Pu deposit, the edges of the gold foil had previously been covered with a silicon glue (CAF 4, Rhodia, France) that was removed after drying of the solution without leaving any trace. The source was then sandwiched with a second gold foil and the two foils were diffusion welded at 400 °C. The final size of the absorber is 0.76 mm × 0.66 mm × 25 μm.

The thermometer is a cuboid (50 μm × 50 μm × 15 μm) of Au:Er (erbium concentration 0.9% in mass). It was fixed to the absorber by ultrasonic cold welding (wedge bonding), thus assuring a metallic contact and a very efficient heat transfer. The detector (absorber+thermometer) was placed on the SQUID chip, with the thermometer being placed inside the SQUID sensing loop. The thermal link consists in a gold wire (diameter 5 μm, length 1.1 mm) wedge bonded to the absorber and to the copper detector holder. For energy calibration, an external ^{55}Fe source was used. A schematic of the detector setup is shown in Fig. 1.

The magnetic field required to magnetize the Au:Er sensor is initially being created with a superconducting coil and then maintained by a superconducting lead ring without injection of an electric current from outside the cryostat. This avoids parasitic heating of the cryostat and allows keeping the magnetic field very stable.

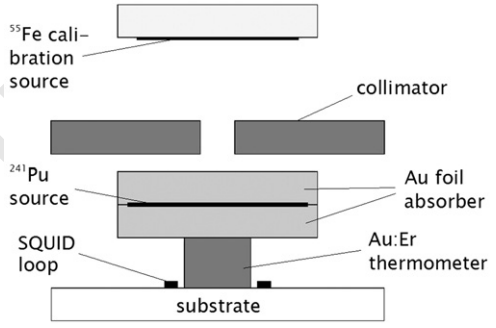

Fig. 1. Schematic of the MMC enclosing a ^{241}Pu source and exposed to an external ^{55}Fe calibration source.

We are using a two-stage SQUID setup with the first SQUID reading out the thermometer and the second acting as a first stage amplifier for the signal delivered by the readout SQUID. The detector together with the readout SQUID was operated in a dilution refrigerator at a regulated temperature of 16 mK. The amplifier SQUID is placed on a dedicated stage of the refrigerator and operated at 1.4 K. This temperature was also regulated in order to guarantee a stable gain.

4. Comparison of experimental and theoretical spectrum

Data were recorded continuously during 61 h at a sampling rate of 200 kHz. All data analysis including triggering was done offline. The pulses had a rise time of about 10 μs and a decay time (1/e) of 3.5 ms. Pulse amplitudes at the output of the SQUID amplifier ranged up to 275 mV for the endpoint of the beta spectrum, at a white noise level of 2.8 μV/Hz$^{1/2}$. After digital filtering, this translates to an energy resolution, as determined for the Mn Kα line, of 29 eV (FWHM) at 5.9 keV. Applying a dead-time of 25 ms and cuts on pulse shape, and setting the (software) trigger threshold at 300 eV, the total number of counts is 1.27×10^6. The beta spectrum (after subtracting the Mn Kα and Kβ lines) contains 1.13×10^6 events. In the following we compare the experimental spectrum with a theoretical spectrum.

4.1. Theoretical beta spectrum of ^{241}Pu

We have calculated the beta spectrum of ^{241}Pu using the programme SPEBETA (Cassette, 1992). The beta decay of ^{241}Pu is a first forbidden, non-unique transition. SPEBETA calculates the spectrum like for an allowed transition and does not take into account radiative corrections. At this stage, we did not introduce any shape factor to the calculation. Since the aim of our work is to determine shape factors, we do not want to use any shape factor from literature. Therefore we expect some discrepancy between the experimental and the theoretical spectrum, in particular at low energy. We consider the result of this experiment as preliminary and will use a forthcoming measurement to determine a shape factor.

4.2. Calibration by the Mn Kα line

In a first attempt, we used the Mn Kα line for the energy calibration of the beta spectrum. Fig. 2 shows the experimental and the theoretical spectrum. The theoretical spectrum has been normalized with respect to the experimental spectrum in the energy range 8–0 keV, where both spectra show the best agreement. Towards higher energies, the experimental and the theoretical spectrum diverge starting from about 12 keV. This is due to a shift of the endpoint of the experimental spectrum with respect to the literature value used for the calculation of the theoretical spectrum.

The best way to determine the endpoint of the experimental spectrum is a Kurie et al. (1936), plot where the quantity

$$K=\sqrt{\frac{n(W)}{\tilde{p}WF(W)}}$$

is plotted versus energy. Here, n is the number of counts at an interval of the normalized electron energy $W=1+E/(m_ec^2)$ $\tilde{p}=\sqrt{W^2-1}$ is the normalized electron momentum, and F is the

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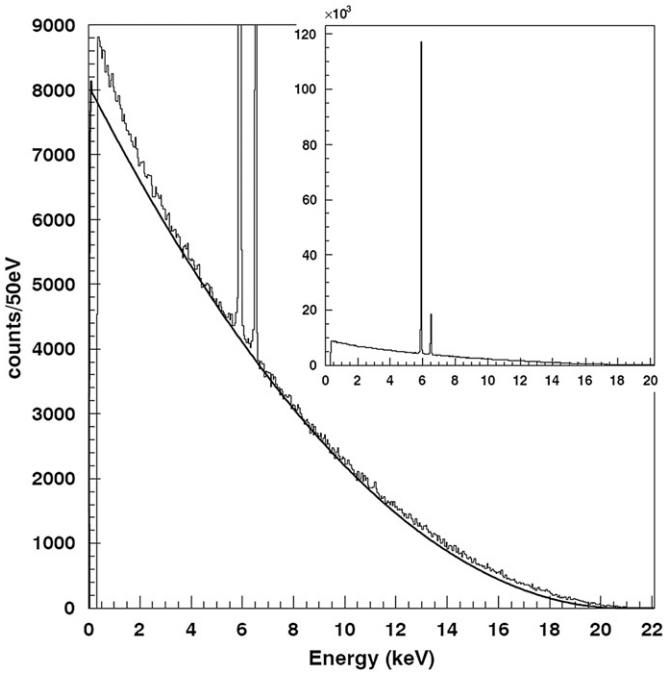

Fig. 2. Experimental and theoretical spectrum of ^{241}Pu, using the Mn Kα line for energy calibration. The inset shows the full spectrum with the unclipped Mn Kα and Kβ lines.

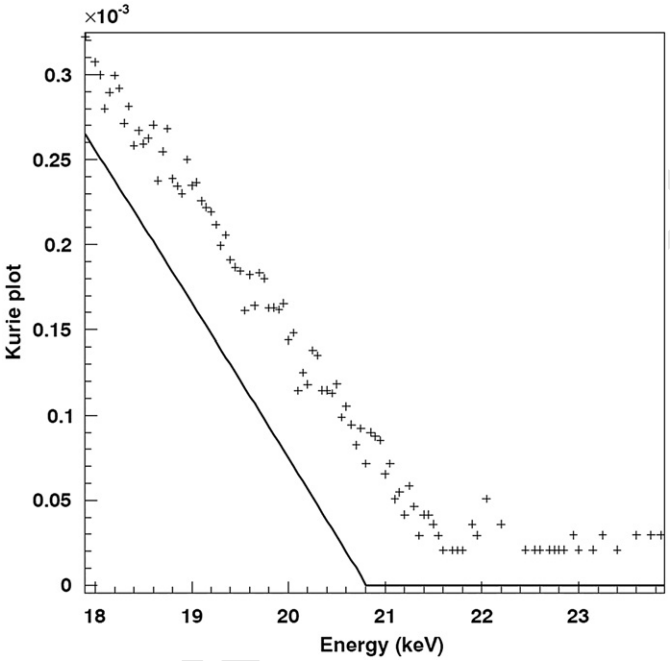

Fig. 3. Kurie plot of the region around the endpoint of the experimental (crosses) and the theoretical (solid line) spectrum of ^{241}Pu, using the Mn Kα line for energy calibration.

Fermi function. In this representation the beta spectrum appears as a straight line whose intersection with the energy axis indicates the end point energy.

In Kurie plot of the region around the endpoint of both our experimental and the theoretical spectrum (Fig. 3), the endpoint of the experimental spectrum appears at about 21.6 keV, whereas the literature value is 20.8 (2) keV (Audi, 2003). As mentioned in Section 4.1, we did not use any shape factor, but this has no influence on the endpoint energy.

Up to now, we have no explanation for the shift of the endpoint in the experimental spectrum. Using both the Mn Kα and Kβ lines for calibration shifts the experimental endpoint towards lower energies by 25 eV. So the measured non-linearity between these two lines cannot explain the shift by +800 eV. From the total heat capacity of the detector (1.8×10^{-11} JK^{-1} at 16 mK), we can estimate the non-linearity between 5.9 and 20.8 keV. The endpoint should be shifted to lower energies by ~0.8% (170 eV) rather than to higher energies. In a next measurement, we shall use a calibration source emitting photons near the endpoint.

One might consider a difference of the response of the MMC to photons (calibration source) and to electrons (^{241}Pu). Previous measurements of the beta spectra of ^{36}Cl (Rotzinger et al., 2008) and ^{63}Ni (Lausberg, 2008) with MMCs, which also relied on energy calibration by external photon sources, showed no discrepancy between the measured and the expected endpoint to within the uncertainties of the measurements. Also, we see no physical mechanism that would lead to a reduced response of the MMC to photon interactions with respect to its response to beta particles. In this experiment we could rather expect the opposite, as explained in the last paragraph of this paper. Finally, one may consider that the literature value of the endpoint energy could be incorrect.

Below 3 keV, both spectra diverge substantially. Given the fact that the theoretical spectrum is calculated without any shape factor, this discrepancy is not surprising. The energy threshold of the experimental spectrum, corresponding to the trigger level, is 300 eV.

4.3. Calibration by the endpoint

For a better comparison of the experimental and theoretical spectra at higher energies, we have in a second attempt used the endpoint as the only calibration point. The resulting spectrum is shown in Fig. 4 together with the theoretical spectrum. Both spectra show very good agreement between the Mn Kβ line and

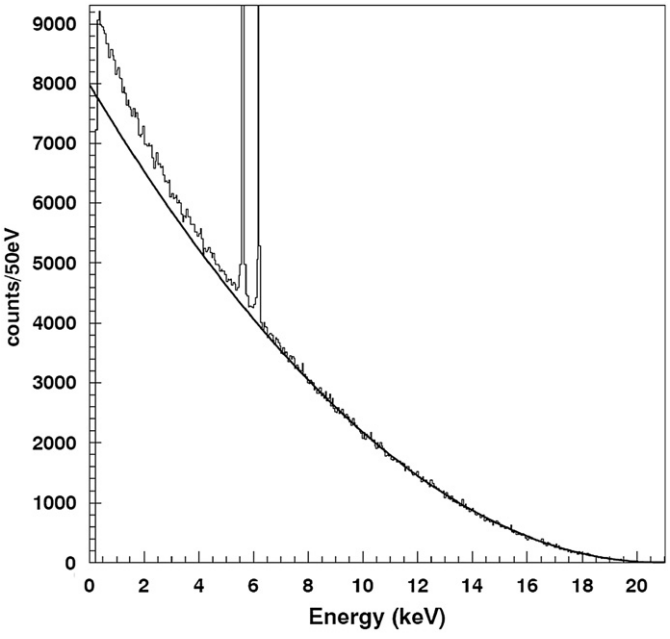

Fig. 4. Experimental and theoretical spectrum of ^{241}Pu, using the endpoint for energy calibration.

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the endpoint. The discrepancy between the two spectra at low energies is, however, more pronounced than when using the calibration by the Mn Kα line.

5. Possible influence of the source quality on the experimental spectrum

In a previous experiment we have carried out an absolute activity measurement of ^{55}Fe (Loidl et al., 2008). In that experiment, where the ^{55}Fe source was also enclosed in the absorber of a MMC, we have observed a substantial difference in the response of the MMC to X-ray photons and Auger electrons. We assumed that X-rays are absorbed in the gold foil forming the detector absorber, whereas Auger electrons loose at least part of their energy in the $FeCl_3$ crystals forming the source. The transfer of the heat produced in the source crystals to the gold foil was slow, the pulse rise times for photon detection and for electron detection differed by at least a factor two. At the same time there seemed to be a mechanism leading to a partial loss of detected energy for the Auger electrons.

The ^{241}Pu solution from which we made the source for the measurement of the beta spectrum contained a substance that was needed for the chemical separation of ^{241}Pu from ^{241}Am. Therefore the thickness of the deposit after drying was of the order of micrometres. Virtually all beta particles loose their entire energy within the thickness of the source. Given the result of the ^{55}Fe measurement, one has to consider whether the ^{241}Pu spectrum is not distorted by a partial loss of energy of the beta particles. This might also explain the discrepancy of the experimental and theoretical spectra at low energies. However, it seems unlikely that a distortion of the spectrum would only occur at low energies whereas both spectra are in good agreement within two third of the energy range. Our confidence in the measured spectrum is supported by the fact that, in contrast to the ^{55}Fe experiment, the pulse shapes are the same for X-rays and for beta particles. The energy transfer from the source to the gold absorber seems to be much more rapid and homogenous than in the ^{55}Fe experiment.

Nevertheless, the quality of the source may have a minor impact on the measured spectrum. We are planning a new measurement of the beta spectrum of ^{241}Pu with the plutonium nuclei being implanted in the gold absorber. In this configuration, the energy of the beta particles is directly deposited in the gold absorber. We are also intending to measure the total emission spectrum of ^{55}Fe with an implanted ^{55}Fe source. This should clarify if the response of a MMC is intrinsically the same for electrons and photons.

6. Conclusion

To our knowledge, we have performed the first measurement of the beta spectrum of ^{241}Pu with a cryogenic detector. The source was enclosed inside the gold absorber of a MMC. The measured spectrum was compared with a theoretical spectrum. We have observed a shift of the endpoint for which we have no explanation. In a forthcoming measurement we will use additional X-ray lines for energy calibration. We also observe a discrepancy between the two spectra at low energies. We attribute this discrepancy to the calculation of the theoretical spectrum without any shape factor. Since the quality of the source may have an influence on the shape of the measured spectrum, we plan to re-measure the spectrum of ^{241}Pu with a source implanted into the detector absorber.

Appendix: Discussion

(John Hardy): I wonder if you have looked at where the Q value from the Audi tables came from.

(Martin Loidl): I started to search for the literature but I did not found the most interesting publications, as for example one Ph.D. thesis written in Russian and I could not find it and some other publications that I still have not been able to come across.

(John Hardy): Do you know what they did? I mean what they may have done is to measure an endpoint energy with a lot worse system than you have.

(Martin Loidl): It is possible. I think that most measurements are based on a magnetic spectrometer. But I do not know any details. I found one publication where they measured only a part of the energy range up to 9 kV and then extrapolated to a value which is compatible with the previously published values.

(John Hardy): I mean, as you correctly pointed out if you have a source problem then what you get is a lower Q value and I would have said then that the lower Q value is likely wrong, I mean that the Audi tables are only as good as the input and I have found many cases where they are off by lots more than those error bars, so before you condemn your detector and throw it away I would suggest that maybe you throw away the Audi tables instead.

(Martin Loidl): Yes, but even better is to look for a better source, that is where I have more confidence and to use additional calibration. Still one comment on one of the slides, at one moment I was asking myself if our detector may have a different response for electrons and for X-rays even though I did not find any physical evidence that would support this and the answer is here in that there is just one quite sharp lines that contains Auger electron and X-rays events, so I think the response is strictly the same.

(Peter Volkovitsky): Could it be some kind of coincidence detection like summation of the beta spectrum and Auger electrons that would shift the spectrum?

(Martin Loidl): I do not believe so, because we looked very carefully at pile up and summing of any events. We applied some strict dead time and I do not believe so.

(Stefaan Pommé): Yes you were mentioning pile up but you had a reasonable count rate, millions of counts in a few days?

(Martin Loidl): The source was 8 Bq and the decay time of the pulses was 3.5 msec. So we had a little bit of pile up. We had a number of events in the raw spectrum when we just simply filtered and gave a first look to the spectrum there were a few events scattered above here.

(Stefaan Pommé): About 1%.

(Martin Loidl): No much less, much less.

(Stefaan Pommé): Because you have 8 Bq and 3 or 4 msec, so if you multiply 8 by 3.4 and divide by a thousand, it is more than one percent?

(Martin Loidl): I think that a part of the primary pile up disappeared in filtering because we artificially shortened the pulse length.

(Stefaan Pommé): So you can find out a pulse upon another pulse from decay.

(Martin Loidl): And the rest were killed by dead time.

(Stefaan Pommé): But the true piled up ones cannot be killed by dead time, they will show up.

(Martin Loidl): Yes, but then we would have gone up to quite high energy and we did not have that.

(Stefaan Pommé): OK, it was just an idea.

(Martin Loidl): At the end point, maybe the right or wrong endpoint, there was nothing.

(Eduardo García-Toraño): If you measure beta emitters of higher energy, I understand that the absorber thickness

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must be greater. Does this affect the time characteristics of your pulses?

(Martin Loidl): Not a lot, the rise time will be a little bit longer, the decay time is determined by the thermal conductance of the thermal link so we can, to some extent, adjust the decay time, so I think we can always adjust the reasonable decay time of the pulses. Of course we must make a much thicker absorber, we have already done a measurement of ^{36}Cl some years ago and it worked quite well.

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