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Performance of a large TeO$_2$ crystal as a cryogenic bolometer in searching for neutrinoless double beta decay

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ABSTRACT: Bolometers are ideal devices in the search for neutrinoless Double Beta Decay ($0\nu$DBD). Enlarging the mass of individual detectors would simplify the construction of a large experiment, but would also decrease the background per unit mass induced by $\alpha$-emitters located close to the surfaces and background arising from external and internal $\gamma$'s. We present the very promising results obtained with a 2.13 kg TeO$_2$ crystal. This bolometer, cooled down to a temperature of 10.5 mK in a dilution refrigerator located deep underground in the Gran Sasso National Laboratories, represents the largest thermal detector ever operated. The detector exhibited an energy resolution spanning a range from 3.9 keV (at 145 keV) to 7.8 keV (at the 2615 $\gamma$-line of $^{208}$Tl) FWHM. We discuss the decrease in the background per unit mass that can be achieved increasing the mass of a bolometer.

KEYWORDS: Cryogenic detectors; Calorimeters

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1 Introduction

Neutrinoless Double Beta Decay (0νDBD) is a rare nuclear process hypothesized to occur in certain nuclei. If observed, it would give important information about the properties of the neutrino and the weak interaction. Double Beta Decay searches [1–3] gained critical importance after the discovery of neutrino oscillations and many experiments are concluding R&D and other are now under construction.

Thermal bolometers are ideal detectors for this survey because they can be composed by most of the more interesting 2β-emitters and, fundamental for next generation experiments, they show an excellent energy resolution. The Cuoricino experiment [4], which constituted of an array of 62 TeO$_2$ (750 g) crystal bolometers, demonstrated the power of this technique and established the basis for the CUORE experiment [5], which will operate 988 TeO$_2$ crystals of the same size. In addition to $^{130}$Te, 2β-scintillating bolometers [6] based on $^{116}$Cd [7], $^{100}$Mo [8, 9], and $^{82}$Se [10] were recently operated with success.

In such experiments, increasing the mass of the individual detector module can be extremely helpful, for several reasons. First, it improves the Peak-to-Compton ratio for γ-ray interactions, enabling not only better identification of environmental background but also decreasing the continuum induced by Compton and multi-Compton scattering. Second, the reduction in the surface-to-volume ratio reduces the background per unit mass from surface impurities [11]. Moreover, the total 2β-efficiency (related to the full containment of the 2 e$^-$ emitted in the decay) of the detector will slightly increase. Last, a large-mass experiment inevitably requires the use of an array of detectors, so a larger individual detector corresponds to a lower number of readout channels, and a simpler setup.
2 Growth of a 2.13 kg TeO$_2$ crystal

The TeO$_2$ crystal studied in this work was grown using the modified Bridgman method, described in detail in previous articles [12, 13]. The growth system (furnace, crucible movement and temperature controllers, etc) was the one used for the large-scale crystal production for CUORE experiment. Several improvements were needed therefore in order to accommodate a larger crucible in a furnace which was designed for the production of crystals of typically $5 \times 5 \times 5$ cm$^3$. The main challenge in this case, where the aim was to grow the largest possible crystal, was to maintain an adequate thermal compensation during all growth stages, especially in the final one when the heat flow is maximal. The thermal compensation is needed to guarantee a flat or slightly convex liquidus-solidus (LS) interface which is a compulsory condition for the obtainment of a good crystal perfection along the whole ingot. One of the peculiarities of the growth method applied was that the seed side of the crucible remained outside the furnace cavity (i.e. in open air) during all stages of growth, so that the control of the LS interface was very challenging. Moreover, in the final stage, almost all the crucible lays in open air which results in a huge thermal radiation and consequent need of thermal compensation. In the particular case of growing a very large crystal the thermal compensation issue was solved by using different thermal compensation rates during the growth of an approximately 2.5 kg TeO$_2$ ingot, considerably larger than the standard ingot used for regular TeO$_2$ crystal production (typically 1.5 kg). In the final phase of cutting and polishing the as-grown ingot, a compromise was chosen in order to get the maximum weight and a reasonable standard shape and quality. In particular 2 of the crystal corners were discarded, corresponding to roughly 2 cm$^3$. The crystal thus obtained shows a slightly truncated-pyramidal shape with a rectangular section. The dimensions of the boundary sections are $54.7 \times 59.6$ mm$^2$ and $54.0 \times 58.2$ mm$^2$. The length is 111.3 mm and its total weight is 2.133 kg.

We remark that the overall quality of the obtained crystal could have been improved, with a custom larger furnace. But the cost of such a new installation was not affordable at this stage of R&D.

3 Experimental details

The TeO$_2$ crystal bolometer is secured by means of eight S-shaped PTFE supports mounted on Cu columns (figure 1). The S-shape of the Teflon supports ensures that with the decrease of the temperature the crystal is clamped tighter, due to the fact that the thermal contraction of PTFE is larger than TeO$_2$. The temperature sensor is a $3 \times 3 \times 1$ mm$^3$ neutron transmutation doped Germanium thermistor, identical to the ones used in the Cuoricino experiment [4]. It is thermally coupled to the crystal via 9 glue spots of $\sim$0.6 mm diameter and $\sim$50 $\mu$m height. In addition, a $\sim$300 k$\Omega$ resistor made of a heavily doped meander on a 3.5 mm$^3$ silicon chip, is attached to each crystal and acts as a heater to stabilize the gain of the bolometer [14, 15]. The 50 $\mu$m gold wires ball-bonded on thermistor and heater are crimped into 0.65 mm copper tubes (“male” pin) inserted into larger copper tubes (“female” pin) glued (electrically insulated) on a copper plate. Twisted constantan wires having a diameter 60 $\mu$m (not shown in figure 1) are crimped in similar Cu tubes on the opposite end of the female connectors and carry the electrical signal up to the cryostat’s Mixing
Chamber, where a custom wiring brings the electrical signal up to the front-end electronics, located just outside the cryostat.

The detector was operated deep underground in the Gran Sasso National Laboratories in the CUORE R&D test cryostat. The details of the the cryogenic facility and its electronics can be found elsewhere [16–19].

Heat pulses, produced by particle interactions in the TeO$_2$ crystal are transduced into voltage pulses by the NTD thermistor, and are then amplified and fed into a 16 bit NI 6225 USB ADC unit. The entire waveform (“raw pulse”) of each triggered voltage pulse is sampled and acquired. The amplitude and the shape of the voltage pulse is then determined by an off line analysis which uses the Optimum Filter (O.F.) technique [5, 20]. The signal amplitude is computed as the maximum of the optimally filtered pulse, while the signal shape is evaluated on the basis of several different parameters: the rise time ($\tau_{\text{rise}}$) and the decay time ($\tau_{\text{decay}}$); the rise time is computed as the time difference between the 10% and the 90% of the trailing edge while the decay time, is computed as the time difference between the 90% and the 30% of the leading edge.

4 Detector Performaces

The detector was operated at a temperature of $\approx 10.5$ mK. The corresponding working resistance of the thermistor was $65 \, \text{M} \Omega$. The main characteristics of the detector are reported in table 1.
Table 1. Main parameters of the crystal bolometer. The second column represents the theoretical resolution given by the Optimum Filter. The last column represents the absolute signal read out across the thermistor. The electronics has a six-pole Bessel filtering stage at 12 Hz.

<table>
<thead>
<tr>
<th>R</th>
<th>FWHM (O.F.)</th>
<th>T\text{rise}</th>
<th>T\text{decay}</th>
<th>Signal</th>
</tr>
</thead>
<tbody>
<tr>
<td>[MΩ]</td>
<td>[keV]</td>
<td>[ms]</td>
<td>[ms]</td>
<td>[µV/MeV]</td>
</tr>
<tr>
<td>65</td>
<td>3.7</td>
<td>29</td>
<td>95</td>
<td>24</td>
</tr>
</tbody>
</table>

Figure 2. Average thermal pulse obtained for the 2.1 kg crystal under study (light red). For comparison we also plot the average pulse obtained with 750 g TeO$_2$ crystal (dark blue) under similar conditions (temperature and resistance). The signals are normalized to the rising edge. The inset shows that the 2.1 kg crystal has a predominant fast decay and a second, very slow decay constant. Also the rise time of the large crystal results ≈2 times faster with respect to the smaller crystal.

The most remarkable feature of this large bolometer is the signal shape. In particular, the decay time shows two unexpected features: it abruptly decreases to ~8% of the signal height and then shows an extremely long decay constant. An example of this behaviour is presented in figure 2, along with the average thermal pulse\(^1\) of a Cuoricino (750 g) TeO$_2$ crystal for comparison. In a very simplified model of a bolometer the system should exhibit only one decayconstant instead of the two clearly observable in figure 2. This decay constant is given by $\tau = C/G$, where $C$ represents the heat capacity of the crystal ($\propto$ mass) and $G$ is the thermal link to the heat sink (dominated by the PTFE supports). Besides this simple model, there are several mechanisms that can largely modify this decay constant. Crystal-lattice imperfections (slightly present in this crystal) will increase the Debye heat capacity, giving rise to an increase of the decay constant (probably the long one observed). The same holds for inclusions (impurities) in the crystal. On the other hand, impurities can also act as "trapping centers" for phonons, inducing further time constants in the system.

\(^1\)The average thermal pulse, i.e. the shape of a pulse in absence of noise, is computed from a proper average of a large number of raw pulses.
Within the Cuoricino experiment a large fraction of the 750 g TeO$_2$ crystals did show two decay constants \cite{21},\footnote{Refer to this reference for a complete treatment of thermal bolometers modelling.} even if this effect was not so evident as in the case of this large crystal.

Considering that the mean decay time observed in the Cuoricino detector was $\sim$250 ms, under the assumption of “perfect” crystals, and given the fact that the PTFE supports used in Cuoricino were identical in shape and number, one would have expected a decay time of $2.13/0.75 \times 250$ [ms] $\approx 700$ ms, instead of 95 ms effectively measured for our crystal.

A fast decay constant is normally observed in crystal absorbers in which the crystal structure exhibits some defects. In the specific case of TeO$_2$ crystals, this behaviour was observed in the enriched ones (operated in Cuoricino) which, being the only visibly “imperfect” TeO$_2$ tested up to now, show a pulse shape \cite{21} very similar to the one observed in our large crystal under study.

Another important point, that comes out from the complete analysis of the Cuoricino data, is a strict correlation between the background $\alpha$-line of $^{190}$Pt and the presence of a second decay constant. Given the natural abundances of Pt isotopes, this effects start to be “visible” once the natural Pt content inside the crystal starts to be at the level of $10^{-7}$ to $10^{-6}$ g/g.

As stated in \cite{13} the surfaces of the ingots are properly cut in order to avoid Pt contaminations from the crucible. Moreover the CUORE crystals undergo a double growth in order to increase the purity of the material. This was not the case of the crystal of the present work.

The second characteristic that is normally associated with non perfect crystal lattices (often resulting also in poly-crystalline structures) and with impurities is a reduction in the absolute signal height. The observed value of 24 $\mu$V/MeV is rather small when compared with the mean value of $\sim 150$ $\mu$V/MeV observed in Cuoricino at a similar working resistance (i.e. temperature) \cite{22}. We believe that this unusual thermal response can be definitely ascribed to some imperfections of the crystal and -more probably- to a large contamination in Pt arising from the compromise of having the maximum weight and a reasonable standard shape and quality as described in section 2.

These imperfections/inclusions can also imply a degradation in the energy resolution of the device as well, even if this cannot be easily evaluated. The degradation can be qualitatively described in terms of position effects due to localized imperfections/inclusions in which the interacting particle could thermalize slightly differently. As example, in Cuoricino the mean FWHM energy resolution of the enriched crystals (evaluated at 2615 keV) was $\approx 15$ keV, while the resolution of the same-sized ($3 \times 3 \times 6$ cm$^3$) natural crystal was $\approx 9$ keV \cite{4}.

In figure 3 we present the calibration spectrum obtained with the 2.1 kg crystal. All the observed low-energy lines are due to internal contaminations of metastable Te isotopes activated through fast neutron interactions which occurred during shipping (15 hours via airplane, from Shanghai to Rome). The most intense low-energy lines are 88 keV ($^{127m}$Te), 105 keV ($^{129m}$Te), 145 keV ($^{125m}$Te), 247 keV ($^{123m}$Te) and 294 keV ($^{121m}$Te). The lines at 570, 1064 and 1770 keV are due, instead, to the presence of another sample, namely a $5 \times 5 \times 5$ cm$^3$ BGO crystal, located a few cm away in the same setup. $^{207}$Bi is a typical BGO contamination and emits these particular three $\gamma$-lines. Finally, the 1461 keV line is due to environmental $^{40}$K contamination, while the 2615 keV line arises from the external $^{232}$Th source. The FWHM energy resolutions evaluated on the most intense lines are presented in table 2.
Figure 3. The 2.1 kg crystal’s calibration spectrum. The most prominent low-energy lines are due to internal contaminations of $^{121m}\text{Te}$, $^{123m}\text{Te}$, $^{125m}\text{Te}$, $^{127m}\text{Te}$ and $^{129m}\text{Te}$, activated during the shipment by airplane. Due to the high counting rate a weak $^{232}\text{Th}$ source was used, in order to avoid pile-up. The corresponding $^{208}\text{Tl}$ 2615 keV $\gamma$-line is highlighted. The 1461 keV line is due to $^{40}\text{K}$ environmental contamination, while the lines at 570, 1064 and 1770 keV are due to the presence of a large BGO crystal, that show a contamination in $^{207}\text{Bi}$.

Table 2. FWHM energy resolutions (in keV) evaluated from the calibration spectrum of figure 3. $^\dagger$The resolution is evaluated on the right (Gaussian) tail of the peak (see figure 4).

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>Resolution (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>145 keV</td>
<td>3.9±.3</td>
</tr>
<tr>
<td>570 keV</td>
<td>4.7±.4</td>
</tr>
<tr>
<td>1461 keV</td>
<td>6.6±.3</td>
</tr>
<tr>
<td>2615 keV</td>
<td>7.8±.7</td>
</tr>
<tr>
<td>5407 keV</td>
<td>7.8±.2 $^\dagger$</td>
</tr>
</tbody>
</table>

Despite the small value of the absolute signal (24 $\mu$V/MeV) the energy resolutions we obtained are only slightly worse with respect to the ones obtained for the CUORE Crystals [23]: (4.1±.4) keV @1461 keV, (5±1)keV @2615 keV, and (5.1±1.2) @5407 keV. The absolute signal height, in fact, does not play a fundamental role in the energy resolution, unless it becomes comparable with the noise level of the electronic chain. During our measurement the noise due to the electronic chain (5±10 nV Hz$^{-1/2}$) is negligible and can be estimated (for this detector) as 1.2 keV FWHM.

All the TeO$_2$ crystals produced so far [23] show a contamination in $^{210}\text{Po}$. This contamination does not represent a serious problem for DBD searches since it has a “relatively short” decay time ($T_{1/2}$=138 days) and is an almost pure $\alpha$-decay. Furthermore, it seems that $^{210}\text{Po}$ is normally
homogeneously distributed in the bulk so that in fact it represents a natural calibration and stabilization line. In this crystal we observed a decay rate of the order of 0.026 Bq. The total acquisition rate of the detector during measurement was 0.13 Hz. This relatively high rate (both in the β and α regions) combined with the extremely long tail of the thermal pulses results in a sort of “permanent” pile-up. As an example, from figure 2 it can be evaluated that ∼7 sec after a 210Po decay the baseline is still at 1.3% of its maximum value. Very naively one can calculate that at this point the bolometer still has to “dissipate” an amount of heat (i.e. energy) of roughly 1.3%×5407 keV ≈ 70 keV. This value is rather high when compared with the energy resolution presented in table 2.

In effect, then, iso-energy events will be randomly distributed over a decreasing tail (mostly induced by 210Po) whose amplitude variation can represent a large fraction with respect to the one induced by a random event. This will slightly affect the energy resolution. The results presented in work were obtained by discarding events occurring within 3 seconds after the signal induced by a 210Po decay. This cut imply a dead time of the order of 5%. A small improvement of the energy resolution can be obtained by increasing this dead window, but the values are, in any case, statistically compatible with the ones presented in table 2.

The peak due to the 210Po decay is presented in figure 4. It can be noted that the peak presents a long non-Gaussian tail on its left side. We do not have a clear explanation for this feature, even though we think it could be partially related to the high event rate of the detector. The tail could also be induced by an anomalous 210Po concentration close to the surfaces of the crystal. This is rather difficult to evaluate but it cannot be excluded considering what was discussed in section 2. The peak is fitted with the Crystal Ball function [24], which is commonly used to model various lossy processes in high-energy physics. The function consists of a Gaussian core portion and a power-law low-end tail, below a certain threshold.

Figure 4. The α+recoil peak due to internal contamination of 210Po. The fit, performed with the Crystal Ball function, is shown in red.
5 Background considerations

In this section we want to study, through MC simulations, the decrease in background per unit mass that can be achieved with larger crystal bolometers. In this view, assuming we can properly operate 2.13 kg crystal bolometers, we want to evaluate the background level in the ROI (Region Of Interest) of a large crystal with respect to a standard 750 g CUORE crystal.

We point out that for the simulations we considered a crystal of $7.1 \times 7.1 \times 7.1$ cm$^3$ (corresponding to a mass of 2.13 kg) because, in a possible future experiments with larger crystals, the choice will fall on regular shapes crystals (i.e. cubic). For the sake of completeness the differences in the simulated background level between a 2.13 kg cubic crystal and the 2.13 kg crystal of this work are rather similar, being such differences between 3 and 20% (depending on the distance of the source).

The main sources of background for TeO$_2$ bolometers [11] are due to:

- $\alpha$-emitters located on or close to the surface of a detector.
- Environmental $\gamma$-emitters, mostly arising from $^{232}$Th decay chain.

Both contributions can be reduced by increasing the size of the crystal.

Surface contamination represents the a major source of background for the Cuoricino and CUORE experiments. Since this background is proportional to the active surface of the crystal, while the DBD signal is proportional to the mass, decreasing the surface-to-volume ratio will result in an increase of the signal over background ratio.

If for example we compare a $7.1 \times 7.1 \times 7.1$ cm$^3$ TeO$_2$ crystal to a $5 \times 5 \times 5$ cm$^3$ CUORE crystal we find the background per unit mass will be smaller by a factor given by the ratio of their sides, $7.1/5= 1.42$. Though this may not seem like a large number, it must be taken into account that the sensitivity of a DBD experiment is directly proportional to $(\text{Detector Mass/Background})^{1/2}$.

The second background effect originates from the environmental $\gamma$ radioactivity. Before discussing the relationship between the size of the crystal detector and the background level due to environmental $\gamma$ radiation, it is useful to briefly describe the main contributions to this background in the energy region 2500–2600 keV, since the DBD peak of $^{130}$Te is expected at 2528 keV.

The background in this region is largely dominated by the $\beta$ decay of $^{208}$Tl, belonging to the $^{232}$Th chain. Several high-energy $\gamma$'s are emitted within this transition, the dominant being the 2615 keV $\gamma$-line, emitted with a Branching Ratio (BR) of 99%. Due to the extremely high transition energy of the decay (5001 keV), a cascade of other high-energy $\gamma$'s are emitted simultaneously, the main ones being at 277 keV, 583 keV and 860 keV.

In order to understand the background, we distinguish two different mechanisms, both involving the 2615 keV $\gamma$ line:

1. multi-Compton events of the 2615 keV $\gamma$-line of $^{208}$Tl.
2. multi-Compton events of the 2615 keV $\gamma$-line, with the simultaneous interaction of a second $\gamma$ in the crystal.

Both of these contributions vary according to the size of the detector. In the first case, an increase in the crystal size increases the probability that a $\gamma$, after several Compton interactions,
releases all of its energy inside the crystal, with a consequent improvement of the peak-to-multi-Compton ratio.

In the second case, however, with a larger crystal there is an increased probability for two γ’s to interact simultaneously in the crystal thus increasing the background level. This contribution strongly depends on the distance between the source and the crystal.

In order to better evaluate the background per unit mass, several Monte Carlo simulations (GEANT4) have been run in which the distance between the source and crystals of different size was varied. In order to have enough statistics (especially for large distances) we defined the ROI as the energy interval between 2500 keV and 2600 keV. We simulated point-like sources placed at various distances from a 5×5×5 cm³ and a 7.1×7.1×7.1 cm³ TeO₂ crystals. In order to better understand the contribution due to the two above mentioned mechanisms, we simulated two different sources: a complete ⁴⁰⁴⁰⁴⁰²³²-Th decay chain, corresponding to a real physical case and a second one consisting of a single 2615 keV γ-line emission. In this way the contribution of the two mechanism as a function of the distance between crystal and source can be disentangled.

In figure 5 we show the results of the simulations. We plot the ratio between the number of counts (within the ROI) per unit mass obtained with a 5×5×5 cm³ crystal with respect to the same for a 7.1×7.1×7.1 cm³ crystal, as a function of the distance between the source and the crystals.

As it can be seen, the background in the ROI for sources very close or very far from the crystal is significantly lower for larger crystals. For intermediate distances, were the coincidences dominate, this difference is less significant. This can be explained in a simple way. When the point-like source is very close to the crystal surface, the coincidence probability does not depend strongly on crystal dimensions since it covers almost half of the solid angle. Once the distance of the source from the detector increases and becomes of the same size of the crystal dimensions then the difference in the coincidence probability of the two crystals reaches its maximum, just due to a solid angle effect. When the distance from the crystal further increases, the difference in the coincidence probability decreases again and becomes negligible. We also performed simulations considering a diffused ⁴⁰⁴⁰⁴⁰²³²-Th source within a “support material” (such as Cu, widely used for thermal detectors) and varying the distance of this support from the crystal. Apart from the time needed for the simulation to accumulate enough events in the ROI, the ratio of the background per unit mass is very close to the one reported in figure 5.

We also performed some simulations considering ²¹⁴Bi, the most troublesome radionuclide of the ²³⁸U chain. In this case we obtained only very small differences between crystals of different size. This is probably due to the fact that in ²¹⁴Bi β-decay there is an enormous number of γ’s emitted simultaneously. In this case, the term due to coincidences dominates with respect to the Multi-Compton term, resulting in a very small difference in the background per unit mass. For the sake of completeness it should be remarked that for experiments based on DBD emitters whose transition energy does not exceed 2615 keV, the main source of environmental radioactivity is dominated by ²³²Th trace contaminations. This is simply due to the fact that the BR of ²¹⁴Bi into high energy γ’s is ~0.15% for the ²³⁸U decay chain, while in the case of ²⁰⁸Tl the BR into the 2615 keV line is 36% for the ²³²Th decay chain.

We conclude by pointing out that the efficiency of containing the 2 e⁻ of the 0νDBD will increase from 87.4% for a 0.75 kg crystal to 91.5% for a 2.13 kg crystal.
Figure 5. Ratio of the events per unit mass of a 5x5x5 cm$^3$ crystal with respect to a 7.1x7.1x7.1 cm$^3$ crystal evaluated in the ROI, as a function of the distance between crystal and source. The black points are obtained simulating a point-like “real” $^{232}$Th source, while the red points relate to a single 2615 keV $\gamma$-line source. The error bars are due to statistical error and correspond to one $\sigma$-level. The lines through the data points are only to guide eyes.

6 Conclusions

We successfully tested a 2.1 kg TeO$_2$ crystal as a thermal bolometer, the largest such detector to date. Despite the presence of imperfections in the crystal, the detector energy resolution in the $0\nu$DBD region is the same as that obtained in the Cuoricino experiment. The advantages of using larger mass crystals for $0\nu$DBD decay searches was simulated and discussed. We strongly believe that our technique is capable of operating multi-kg crystal detectors (composed by different $\beta\beta$-emitters) with the required energy resolution in the 2÷3 MeV energy window. There is large room for improvement in crystal quality and thus in pulse shape and energy resolution as well. The current sample came from a routine production, so better-quality large crystals could be obtained if dedicated furnaces were used.

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