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Measurement of Low Temperature Specific Heat of Crystalline TeO_2 for the Optimization of Bolometric Detectors

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The optimization of bolometric detectors, like those that will be developed for the rare event experiment CUORE, requires a complete knowledge of the detector's thermal parameters. Since the CUORE detecting elements will consist of TeO_2 crystals, we have measured the specific heat of this material down to 60 mK with the thermal relaxation method. Previous available data were taken at temperatures higher than 0.6 K. Our results are clearly consistent with a lattice dominated specific heat. The Debye temperature, evaluated to be (232 ± 7) K, is in excellent agreement with the elastic constant values measured by other authors. The knowledge of the Debye temperature allows a simple prediction of the pulse amplitude of presently working bolometers.

INTRODUCTION

Much interest in underground physics is at present devoted to the search for rare events. The proposed Cryogenic Underground Observatory for Rare Events (CUORE)[1] is an apparatus, based on cryogenic detectors, mainly devoted to the study of the neutrinoless Double Beta Decay (DBD),[2] although it can also perform other important experiments such as Dark Matter and low-energy nuclear physics searches. From the technical point of view, it represents an important step towards a new generation of large mass cryogenic detectors, with total masses in the ton scale. CUORE has been proposed by an international collaboration involving groups from the Lawerence Berkeley Laboratory, the University of Florence, the

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University of Leiden, the University of Insubria, the University of Milan, the University of South Carolina, the University of Zaragoza, the Gran Sasso National Laboratories and the Legnaro National Laboratories.

Neutrinoless DBD is a rare nuclear process forbidden by the standard electroweak model. Its observation would imply lepton number non-conservation. From the rate of neutrinoless DBD one can extract an average neutrino Majorana mass, subjected however to considerable uncertainties coming from the nuclear matrix element calculation.

Direct measurements of this process are based on the detection of the two emitted electrons and can be divided into two categories: those in which the source is different from the detector and those in which the detector itself is made with a $\beta\beta$ active material. CUORE is an experiment of this last type where the $\beta\beta$ active material is ^{130}Te. It consists of an expansion of the presently running Milano Double Beta Decay experiment performed at the underground INFN Gran Sasso Laboratories. In this experiment, the detector consists of a tower of 20 elements. Each element, working as an individual device, consists of a 340 g TeO_2crystal. The total sensitive mass is 6.8 kg, the largest bolometric set-up ever operated in the world.[3]

CUORE is in the Research, Development and Design stage at the moment.[1] It will consist of an array of 1020 closely packed independent elements. Each element[4] will consist of a $5 \times 5 \times 5$ cm^3 TeO_2 crystal (with a mass of 760 g) and a Neutron Transmutation Doped (NTD) germanium thermistor[5] and will be cooled down to a temperature of about 10 mK.

As far as the operation of the single element is concerned, satisfactory preliminary results have been obtained: an array with four $5 \times 5 \times 5$ cm^3 TeO_2 crystals has been successfully tested. One of the crystals has shown a resolution similar to that of the best Ge diodes for high-energy gamma rays and definitely superior (4.2 keV at FWHM) for alpha-particles.[4] Moreover the energy threshold achieved ($\sim$5 keV) aims at other interesting applications like search for WIMPs and Solar Axions.

The knowledge of the specific heat of TeO_2 at very low temperatures is necessary to formulate a thermal model for the detector, able in principle to explain the results so far obtained and to help in the design of future devices. The aim of the work described in this paper was to measure the specific heat of the TeO_2 crystals used for CUORE construction, at temperatures as close as possible to the foreseen operation temperature ($\sim$10 mK). In the literature,[6,7] TeO_2 heat capacity data are reported for temperatures down to 0.6 K. At such temperatures, the heat capacity of the addendum (that part of the set-up necessary to perform the measurements, such as thermometers, heaters and holding parts) is often larger than that of the specimen, introducing an important error source. On the contrary,

we were able to realize an addendum with a negligible heat capacity in comparison with that of the specimen in the temperature range of interest.

We measured the heat capacity of the TeO_2 in the 0.06–0.28 K temperature range by means of the thermal relaxation method.[8] As we shall show, a single time constant approximation was adequate in treating the data.[9]

EXPERIMENTAL PROCEDURE

The sample was a TeO_2 optical quality crystal provided by the Shanghai Quinghua Nonmetal Material Co. The volume was about $2 \times 2 \times 3\ cm^3$, corresponding to a mass of 75.493 g. It was sustained by four pure tin cylinders, which kept the crystal blocked inside a copper frame, as shown in Fig. 1. The length of the cylinders was chosen to compensate the thermal contraction of the crystal. The frame was in good thermal contact with the mixing chamber of a dilution refrigerator. A RuO_2 thermometer measured the temperature T_0 of the frame. A thermal shield at the temperature of the mixing chamber was used.

A gold film (thickness $s = 1000$ Å, residual resistance $R \sim 9\ \Omega$) was deposited on the crystal in order to obtain a heater with a negligible heat capacity. We have carefully checked that the film resistance was constant along the temperature range of the specific heat measurements. A calibrated[(1)] NTD #31 Ge sensor, glued on the crystal with a minimum amount of GE-varnish in order to minimize the heat capacity of the addendum, was used as a thermometer. The dimensions of our sensor are $2.95 \times 1.15 \times 0.3\ mm^3$. The two opposite faces ($1.15 \times 0.3\ mm^2$) have undergone a B ion implantation and two depositions: Pd (200 Å) and Au (4000 Å). This thermistor was calibrated in the range 0.05 K–0.3 K.

NbTi wires, 25 μm diameter, realized the electrical connections for the thermometer and the heater on the TeO_2 crystal. The wires (two for each resistance) were stretched along the opposite sides of the copper frame (Fig. 2). Aluminum capillaries (0.2 mm outer diameter, 0.1 mm inner diameter, length 2 mm) clamped together the NbTi wires and the gold wires (25 μm diameter), connecting both the NTD sensor and the gold deposition.

In Table I, the contributions to the heat capacity C_{sp} of the addendum are shown. A factor 1/3 was attributed to the heat capacity contribution of the elements linking the crystal to the frame. It can be shown[8] in fact that, when considering the finite heat capacity of the thermal link, it contributes

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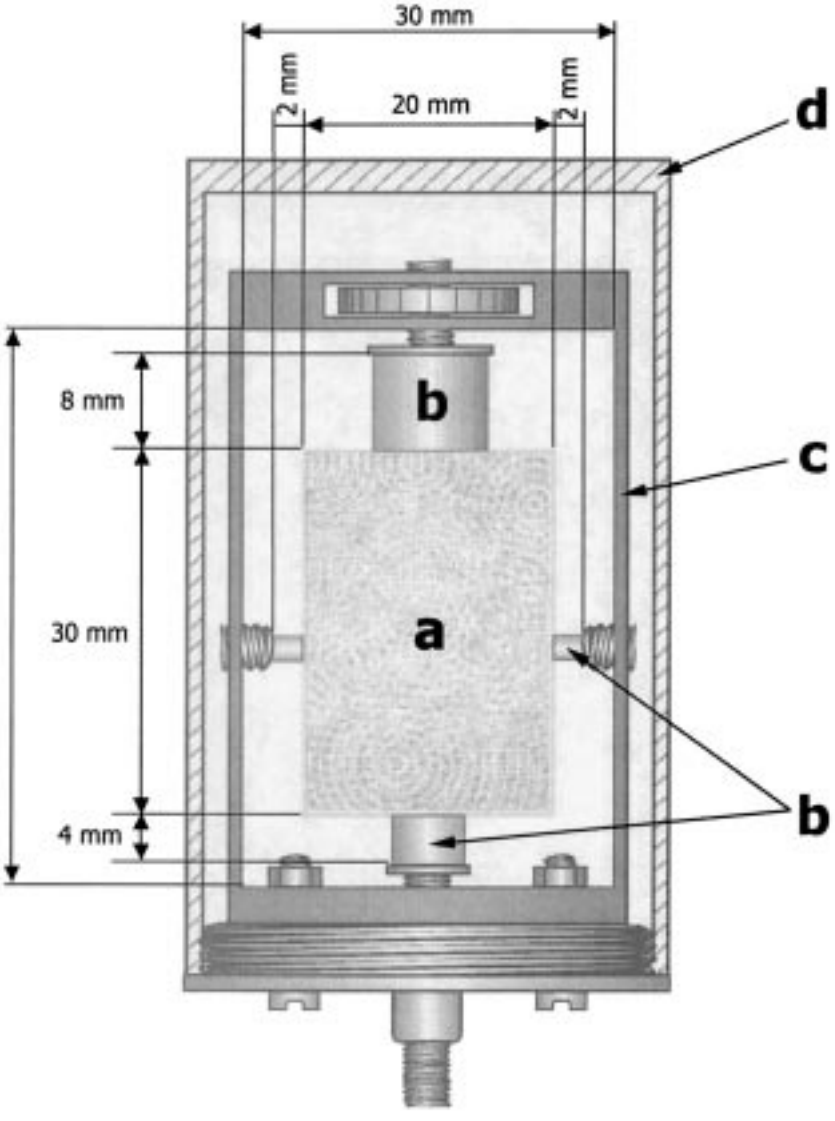

Fig. 1. Schematic view of the mounting of the TeO_2 crystal: (a) TeO_2 crystal, (b) tin cylinders, (c) copper frame, (d) thermal shield.

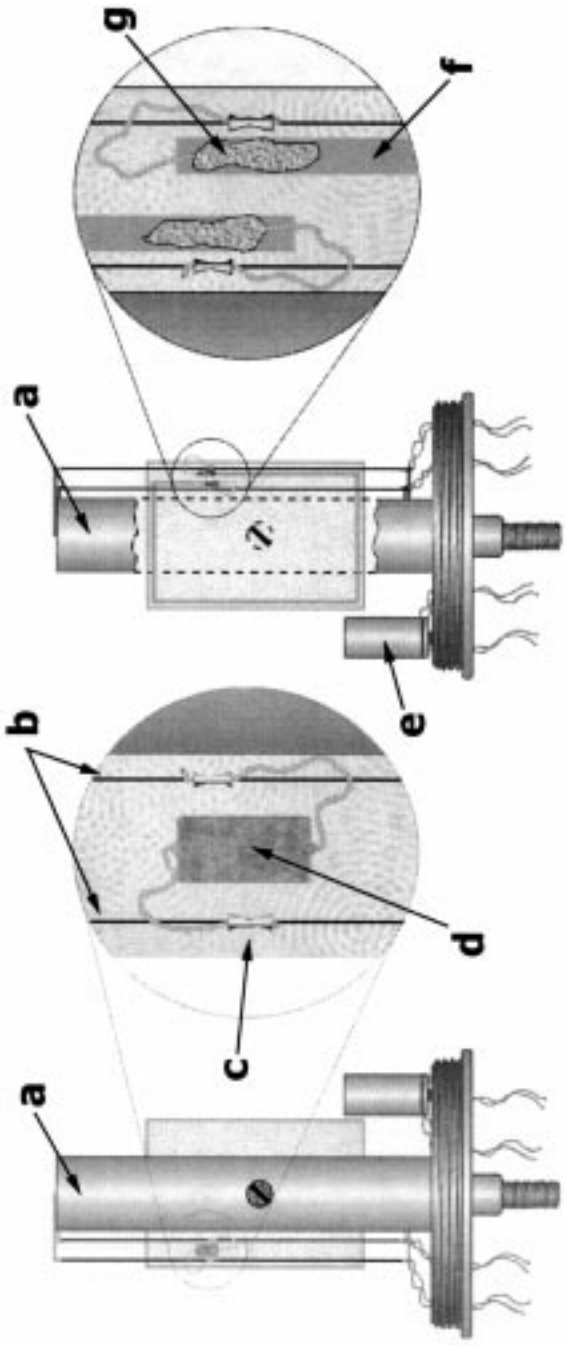

Fig. 2. Electrical connections of the NTD thermistor (left) and of the gold film heater (right): (a) copper frame, (b) NbTi wires, (c) Al capillaries, (d) Ge thermistor, (e) thermometer, (f) gold deposition, (g) indium soldering.

for 1/3 of this value to the total value obtained by the thermal relaxation method. Note that the electron heat capacity of the NTD #31 sensor was derived from the electron heat capacity of the NTD #12 thermistor (close to #31 as doping level), since it was reported that differences in electron heat capacity for so similar sensors are negligible.[10] C_{sp} was between 10^{-10} and 10^{-8} J/K, due to several contributions as reported in Table I.

In order to control that the calibration of the thermometer did not change because of the mounting procedure and gluing, in a second run a good thermal contact (a copper strip of thermal impedance about 10^4 K/W at 0.1 K) was made between the crystal and the copper frame. Within the experimental errors, we did not find changes in the calibration curve of the thermometer.

RESULTS

Thermal Conductance of the Sample to the Bath

The thermal conductance between the TeO_2 crystal and the heat sink was measured by supplying a power P to the heater. When thermal equilibrium was reached, the temperature T of the crystal was recorded. By varying the power supplied to the crystal and keeping the temperature T_0 constant (around 0.05 K), we obtained a set of data $P(T)$, with T values extending up to about 0.3 K. The $P(T)$ curve was then differentiated with respect to T in order to obtain the conductance G.

The fit of the values obtained is:

$$G(T) = (12.3 \pm 0.3) \times T^{(2.54 \pm 0.01)}\,\mu\text{W/K}$$

It can be observed that the bulk thermal conductance of the tin cylinders[11] and of the NbTi wires[12] are respectively about two orders of magnitude higher and four order of magnitudes lower than the measured $G(T)$. Therefore, the main contribution to G is the thermal conductance of the contacts Cu/Sn and Sn/TeO_2. Moreover, the exponent between 2 and 3 has been already reported for measurements of contact thermal resistances between solids.[13]

Measurement of the Heat Capacity

Heat capacity was measured in the 0.06–0.28 K temperature range using a relaxation method.[6] A small power supplied by the heater rose the temperature T of the crystal above T_0 by typically ~ 10 mK. When the thermal equilibrium was reached, the heating power was switched off and the exponential decay of the crystal temperature was recorded by means of

TABLE I

Contributions to the Heat Capacity C_{sp} of the Addendum. In the Last Column, the Heat Capacities at 0.1 K are Reported

Material	Volume (mm^3)	$C = aT + bT^3$ (μJ/K mm^3)		C (μJ/K) $T = 0.1$ K
		a	b	
Au D. C. Alsop *et al.*, *Appl. Opt.* **31**, 6673 (1992)	1.93×10^{-2}	6.8×10^{-2}	4.5×10^{-2}	1.32×10^{-4}
Pd D. C. Alsop *et al.*, *Appl. Opt.* **31**, 6673 (1992)	0.102×10^{-2}	1.1	1.1×10^{-2}	1.12×10^{-4}
NTD31 P. H. Keesom and G. Seidel, *Phys. Rev. B* **133**, 33 (1959) E. Aubourg *et al.*, *J Low Temp. Phys.* **93**, 289 (1993)	1.61	6.02×10^{-4}	3.0×10^{-3}	1.02×10^{-4}
Ge-Varnish R. B. Stephens, *Phys. Rev.* **8**, 2896 (1973)	0.381	7.031×10^{-3}	2.104×10^{-2}	2.76×10^{-4}
In H. R. O'Neal *et al.*, *Phys. Rev. A* **137**, 748 (1965)	0.215	—	7.77×10^{-2}	1.67×10^{-5}
Al N. E. Phillips, *Phys. Rev.* **114**, 676 (1959)	0.263	—	2.83×10^{-1}	7.44×10^{-5}
Sn H. R. O'Neal *et al.*, *Phys. Rev. A* **137**, 748 (1965) Y. S. Touloukhian *et al.*, *Thermophysical Properties of Matter*, Plenum (1970)	57	—	1.478×10^{-2}	2.81×10^{-4} ($\frac{1}{3}$ contribution)
NbTi T. Bishoff *et al.*, *Cryogenics* **22**, 131 (1982)	$4 \times 2.36 \times 10^{-2}$	—	7.03×10^{-2}	2.21×10^{-6} ($\frac{1}{3}$ contribution)

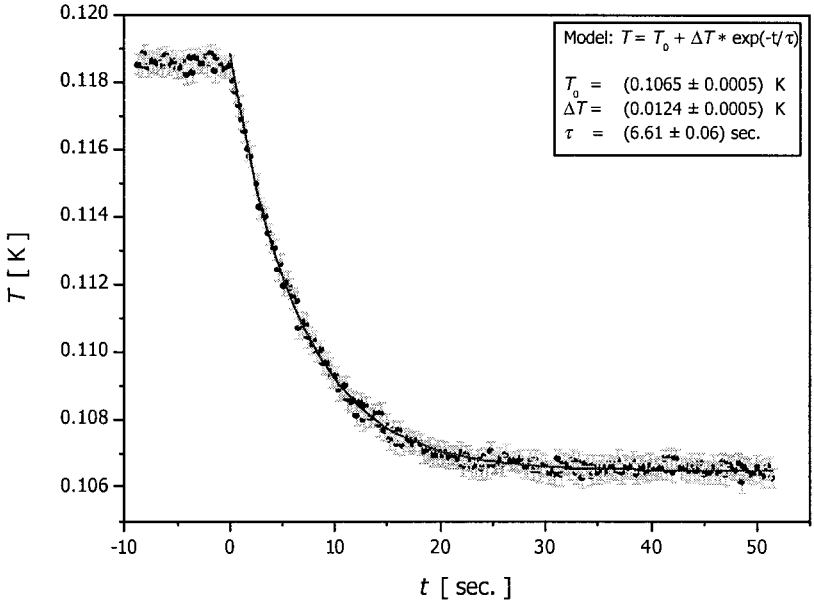

Fig. 3. Example of the exponential decay of the crystal temperature after removal of the heating power.

a LR700 bridge, at a rate of 5 sample/s. An example of $T(t)$ is reported in Fig. 3. A single relaxation time constant was always observed. We repeated the same procedure at different values of T_0 between 0.06 and 0.28 K, in order to obtain a set of thermal discharges.

The value of the heat capacity was obtained as τG. These values were then corrected for the contribution of the addendum C_{sp}. Heat capacity data are shown in Fig. 4, in which the ratio C/T is reported as a function of T^2. Note that the contribution of the addendum to the total heat capacity is between 1.85 and 0.3% over the whole temperature range. From a linear fit to the data, we obtained for the coefficient:

$$a = (2.21 \pm 0.03) \times 10^{-4}\ \mathrm{J/K^4}$$

Since C had a cubic dependence on T and G was mainly due to the contact resistance, τ was almost constant over the measurement temperature range and close to 10 s.

DISCUSSION

As we can see from Fig. 5, the heat capacity we measured is consistent with White's data[7] above 2 K. Below that temperature, our data are

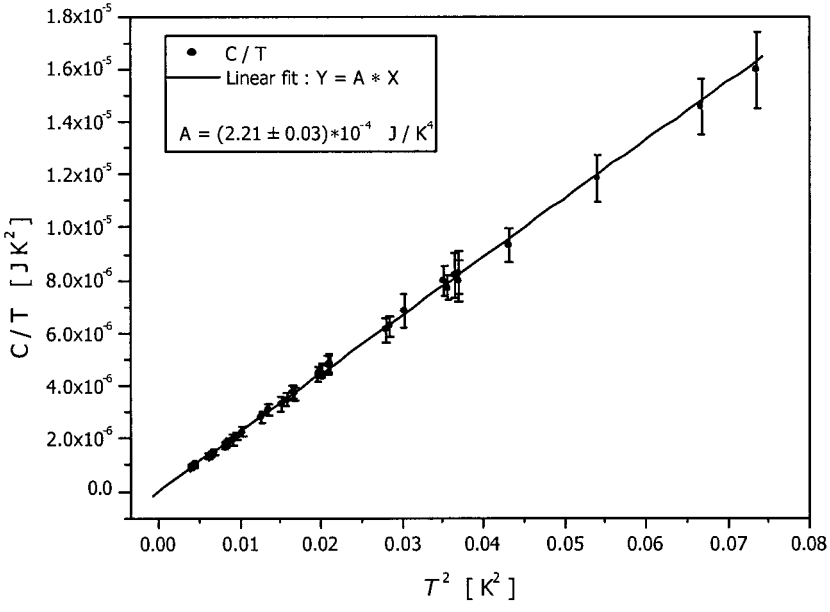

Fig. 4. Heat capacity data, showing the evident linearity of C/T versus T^2.

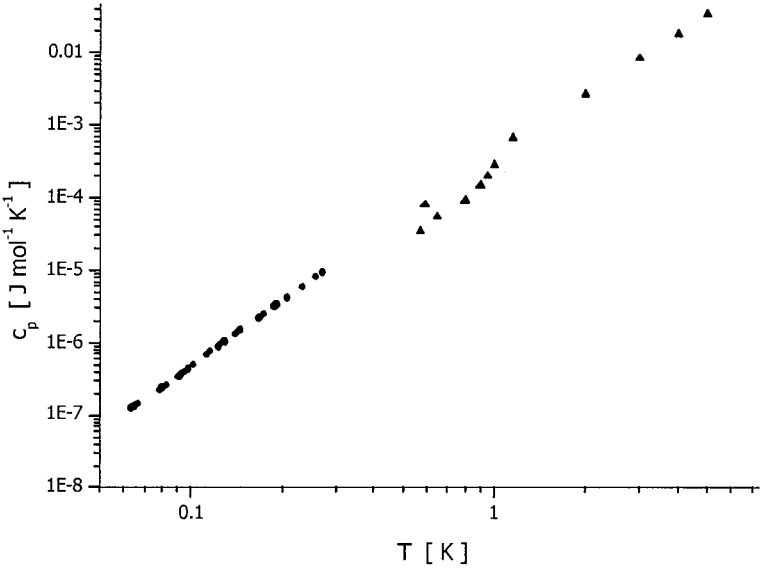

Fig. 5. Comparison of White's measurements (triangles) with our data (full circles).

slightly higher than White's data, but our measurements are made with a negligible contribution of the addendum, while in White's experiment the heat capacity of the addendum increases to more than double that of the small specimen below 3 K.

From the slope of the linear fit of C/T vs. T^2, it is straightforward to extract the limiting value of the Debye temperature. We obtained $\theta_D = (232 \pm 7)$ K. All values reported in the literature for θ_D of TeO_2 are decreasing with decreasing T and the lowest value reported is the White's value 265 ± 10 K, extrapolated from heat capacity data at about 1 K. Our value is the lowest ever reported for θ_D and it is in excellent agreement with the value calculated from the extrapolation at $T = 0$ K of Ohmachi and Uchida's[14] and Schweppe's[15] ultrasonic measurements of elastic stiffness constants c_{ij}. In fact, using the Betts, Bhatia and Horton's approximation[16] to evaluate θ_D for a tetragonal crystal, including all tetragonal harmonics up to those of degree six, we calculated $\theta_D = (232 \pm 8)$ K. The error on the elastic estimation of θ_D is mainly due to the extrapolation of the elastic constant values at low temperatures.

Measurements of θ_D for TiO_2, a crystal with the same structure as TeO_2, reported consistent values for the Debye temperature resulting from calorimetric and elastic measurements,[17] excluding the presence of excitations other than acoustic lattice vibrations.

IMPLICATIONS ON DETECTOR PERFORMANCES

The measured value for the Debye temperature of TeO_2 can be used to estimate the expected pulse amplitude for a given energy deposition in the presently running 340 g detectors. This evaluation can be compared with the amplitude experimentally observed and some interesting considerations can be performed.

In order to evaluate *a priori* the pulse amplitude expected for the detectors under discussion, one should have developed a complete thermal model for the device, and some critical assumptions must be made around the mechanism of pulse formation.

A naive approach consists of considering the detector as a monolithic device from the thermal point of view, characterized by a total heat capacity dominated by the TeO_2 crystal heat capacity. Furthermore, for sake of simplicity one assumes that the energy deposited in the crystal is instantaneously thermalized in the whole detector, including the NTD Ge thermistor. Under these hypotheses, the signal amplitude ΔV across the thermistor is given by the following formula, which represents an upper limit for pulse height in case of full thermalization:

$$\Delta V = I(\partial R/\partial T)\, E/C$$

where I is the current flowing in the thermistor, $\partial R/\partial T$ the thermistor sensitivity, E the deposited energy and C the total detector heat capacity at the operating temperature. In the case of the 20 detector array developed by the Milano group, most of the detectors are biased with a current of 8.5×10^{-11} A with an average thermistor resistance of about 130 MΩ, corresponding to an operating temperature of 12.1 mK. In these conditions, $(\partial R/\partial T)$ is typically 90 MΩ/mK. An energy deposition of 1 MeV would determine a temperature increase of 9.2×10^{-5} K, considering a Debye temperature of 232 K for TeO_2. This would give a pulse amplitude of 703 μV. The corresponding average experimental amplitude is 364 μV. The comparison between the two values suggests some consideration:

• The observed pulse amplitude is of the correct order of magnitude if full thermalization is assumed. It is well known however that the phonon spectrum generated by a ionizing particle impinging on a dielectric crystal at low temperatures is out of thermal equilibrium, since initially the phonons have typically the Debye energy. Only if the phonon thermalization time is fast enough with respect to the thermal time constants of the detector, full instantaneous thermalization can be assumed (as in our case), and the subsequent pulse evolution is determined by the heat flow through the various detector elements. It is then meaningful to attribute a temperature to each detector element and the pulse formation can be considered essentially thermal.

• The signal is about half of the value given by the optimistic estimation reported above. This is not surprising. In a full thermal model, the detector should be divided into three thermal stages: the TeO_2 crystal, the lattice of the NTD thermistor and the electrons of the NTD thermistor; finite thermal conductances connect these stages among them and with the heat sink. The deposited energy is thermalized in the TeO_2 crystal, while the signal consists of a variation of the *electron* temperature of the thermistor. Since the energy flowing from the TeO_2 crystal to the thermistor electrons is partially lost towards the heat sink, the signal temperature variation can only be less than the original temperature variation of the crystal.

• Since the observed amplitude is reasonably close to the expected one, no dramatic improvements in detector performances can be expected with respect to the present ones (at least in terms of pulse amplitude). In order to get substantially higher pulses, the philosophy of the detector design should be deeply changed, aiming at non-thermal phonon collection and therefore at much faster sensors than NTD thermistors. (The present rise time is around 50 ms). The present energy resolution is however sufficient for CUORE purpose.

In conclusion, we observe that a very naive thermal model, joined to an independent experimental determination of TeO_2 specific heat, is able to predict pulse amplitudes within a factor two. This result is a stimulus for the experimental determination of the other poorly known thermal parameters of the detector (like the NTD electron specific heat), in order to calibrate a more complete and predictive model of the device.

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