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First test of an enriched $^{116}$CdWO$_4$ scintillating bolometer for neutrinoless double-beta-decay searches

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Abstract For the first time, a cadmium tungstate crystal scintillator enriched in $^{116}$Cd has been successfully tested as a scintillating bolometer. The measurement was performed above ground at a temperature of 18 mK. The crystal mass was 34.5 g and the enrichment level $\sim$82%. Despite a substantial pile-up effect due to above-ground operation, the detector demonstrated high energy resolution (2–7 keV FWHM in 0.2–2.6 MeV $\gamma$ energy range and 7.5 keV FWHM at the $^{116}$Cd double-beta decay transition energy of 2813 keV), a powerful particle identification capability and a high level of internal radio-purity. These results prove that cadmium tungstate is a promising detector material for a next-generation neutrinoless double-beta decay bolometric experiment, like that proposed in the CUPID project (CUORE Upgrade with Particle IDentification).

1 Introduction

Neutrinoless double-beta ($0\nu 2\beta$) decay is a hypothetical nuclear transformation that changes the lepton number by two units when a candidate even-even nucleus emits two electrons with no neutrino in the final state. The observation of $0\nu 2\beta$ decay would testify lepton number non-conservation and the presence of a Majorana term in neutrino masses, and give information on the neutrino-mass absolute scale along with the ordering of the neutrino-mass eigenstates [1–3]. It should be stressed that many effects beyond the Standard Model can contribute to the $0\nu 2\beta$ decay rate [4–7].

In contrast with the two-neutrino mode ($2\nu 2\beta$), experimentally observed in eleven isotopes with half-lives in the range $10^{18}$–$10^{24}$ years (see reviews [8–10] and references therein) and allowed in the Standard Model, the $0\nu 2\beta$ decay has not been detected yet. The most sensitive experiments give only half-life limits on the level of $T_{1/2} > 10^{25}$–$10^{26}$ years, which correspond to constraints on the effective Majorana neutrino mass around $\langle m_\nu \rangle < 0.1$–1 eV, in the degenerate hierarchy region of the neutrino mass eigenstates (see reviews [7,10–13] and the recent KamLAND-Zen result [14]). The goal of the next-generation $0\nu 2\beta$ experiments is to probe the inverted hierarchy region of the neutrino mass, which requires a sensitivity of $\langle m_\nu \rangle < 0.05$–0.02 eV. This neutrino mass scale corresponds to half-lives $T_{1/2} \sim 10^{27}$–$10^{28}$ years even for the nuclei with the highest decay probability [1,2]. The attainment of such a high sensitivity requires the construction of a detector containing a large number of $2\beta$ active nuclei ($10^3$–$10^4$ moles of isotope of interest), extremely low (ideally zero) radioactive background, high detection efficiency (obtainable in the calorimetric approach “source = detector”) and ability to distinguish the effect searched for (in particular, as high as possible energy resolution). Taking into account the extremely low decay probability and the difficulties of the calculations of the nuclear matrix elements [1,2], the experimental program should include a few candidate nuclei.

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The technique of low temperature scintillating bolometers looks very promising to satisfy the above mentioned requirements [15–17]. The nucleus $^{116}$Cd is one of the most attractive candidates thanks to one of the highest energy release ($Q_{\beta} = 2813.50(13)$ keV [18]), comparatively large natural isotopic abundance ($\delta = 7.512(54)$% [19]), applicability of centrifugation for cadmium isotope enrichment in a large amount, and availability of cadmium tungstate crystal scintillators (CdWO$_4$).

Cadmium tungstate crystals are routinely produced on an industrial basis and are among the most radiopure and efficient scintillators, with a long history of applications in low counting experiments to search for double-beta decay [20–24] and investigate rare $\alpha$ [25] and $\beta$ decays [26–28]. Recently, high-quality radiopure CdWO$_4$ crystal scintillators were developed from deeply-purified cadmium samples enriched in the isotopes $^{106}$Cd [29] and $^{116}$Cd [30] with the help of the low-thermal-gradient Czochralski crystal-growth technique [31]. These enriched scintillators are currently and successfully used in the $0\nu \beta \beta$ decay experiments with $^{106}$Cd [32,33] and $^{116}$Cd [34,35]. Important advantages of the low-thermal-gradient Czochralski method are a high yield of the crystal boules ($\approx 87$%) and an acceptable low level of irrecoverable losses of enriched cadmium ($\approx 2$%). Thus, production of high quality radiopure cadmium tungstate crystal scintillators from enriched isotopes is already a well developed technique. Starting from the beginning of nineties of the last century, CdWO$_4$ was intensively tested first as a pure scintillator [26] and then as a scintillating bolometer with a high performance in terms of energy resolution, particle discrimination ability and low radioactive background [15,36–38].

The aforementioned results played a crucial role in including CdWO$_4$ in the list of the possible candidates for the CUPID project [39,40]. In this context, the first bolometric test of an enriched $^{116}$CdWO$_4$ scintillating bolometer – here reported – adds a crucial missing piece of information in view of the full implementation of the cadmium tungstate technology for $0\nu \beta \beta$ search. It should be stressed that reproducing the results achieved with materials of natural isotopic composition with enriched crystal scintillators is not trivial. Indeed, the procedures of purification of enriched isotopes and the growth of crystals from enriched materials are severely constrained by the strong requirements of a high yield in developing ready-to-use crystals and minimal losses of the costly enriched materials. These requirements may affect negatively the bolometric performance and the intrinsic background, which need to be specifically studied for bolometers containing enriched isotopes. Among the three candidates that are very attractive for the scintillating bolometer technology, i.e. $^{106}$Mo, $^{82}$Se and $^{116}$Cd, positive tests on enriched materials were performed before this work only in the first two cases [41,42]. The results here described on $^{116}$Cd complete the investigation of these isotopes and enhance the merits of the $^{116}$CdWO$_4$ technology.

### 2 Test of a $^{116}$CdWO$_4$ scintillating bolometer

A sample of enriched $^{116}$CdWO$_4$ crystal scintillator was cut from the wide part of the growth cone of a 1.9 kg crystal boule [30] (see Fig. 1 in Ref. [43], where the boule and cut parts are shown). The crystal mass and size are respectively 34.5 g and $28 \times 27 \times 6$ mm, and the isotopic concentration of $^{116}$Cd is 82%. The light detector (LD) consists of a high-purity germanium wafer ($\varnothing 44 \times 0.175$ mm) produced by Umicore. The scintillator and the Ge wafer were fixed in individual copper frames by using PTFE pieces and brass/copper screws. The inner surface of the detector holder was covered by a reflecting foil (Vikuiti™ Enhanced Specular Reflector Film) to improve the scintillation light collection. A neutron transmutation doped (NTD) Ge thermistor with a mass of $\approx 50$ mg was glued on the $^{116}$CdWO$_4$ crystal by six spots of epoxy (Araldite®) to register the temperature pulses induced by the absorption of particles in the $^{116}$CdWO$_4$ crystal. An approximately three-times-smaller NTD Ge thermistor was attached to the LD with the aim to reduce the added heat capacity and to increase the LD sensitivity. Both bolometers were supplied with a silicon chip on top of which a heavily doped meander was formed by donor ion implantation. The meander resistance is stable down to millikelvin temperature and was used as a heater [44] to inject periodically fixed amounts of thermal energy for the detector stabilization. The partially assembled $^{116}$CdWO$_4$ scintillating bolometer and the LD are shown in Fig. 1.

The low-temperature tests of the $^{116}$CdWO$_4$ scintillating bolometer were performed in a cryogenic laboratory of the CSNSM (Orsay, France) by using a dry high-power dilution refrigerator [45] with a $4$ K stage cooled by a pulse-tube.

![Fig. 1 Photograph of the 34.5 g $^{116}$CdWO$_4$ scintillating bolometer assembled on a copper plate covered by a reflecting foil (left) together with the Ge-based light detector (right). See the text for the details](image-url)
The sample holder is mechanically decoupled from the mixing chamber by four springs to reduce the acoustic noise caused by the cryostat vibrations. The outer vacuum chamber of the refrigerator is surrounded by a passive shield made of low radioactivity lead (10 cm minimum thickness) to suppress the environmental γ background. The shield mitigates the pile-up problem typical for above-ground measurements with macro-bolometers, given the slow response of these devices (tens or even hundreds of milliseconds). For the same reason, we have used a relatively small 116CdWO4 sample aiming to reduce the counting rate of the environmental γ background.

A low-noise electronics based on DC-coupled voltage-sensitive amplifiers [46] and located inside a Faraday cage was used in the experiment. The 116CdWO4 and the LD NTD sensors were biased with currents of 4.2 and 25 nA, respectively. The bias current was injected through two load resistors in series with a total resistance of 200 MΩ for both channels. The stream data were filtered by a Bessel filter with a high frequency cut-off at 675 Hz and acquired by a 16 bit ADC with 10 kHz sampling frequency.

Most of the measurements were performed with the sample holder temperature stabilized at 18.0 mK. However, the 116CdWO4 detector was approximately 2 mK warmer due to a not reached temperature equilibrium between the mixing chamber and the detector itself, because the scintillating bolometer was mounted to the mechanically-decoupled holder by means of brass rods, non-optimal for thermalization. Therefore, the NTD-Ge-thermistor resistances (RNTD) at the working temperature had a clear trend to increase. For instance, the resistance of the heat channel thermistor changed from an initial ∼0.4 MΩ value to a final ∼1 MΩ during the two-week background run. It is worth noting that the sample-holder temperature reached 9.6 mK during a short test with unregulated temperature, and the corresponding NTD-Ge-thermistor resistance of the 116CdWO4 bolometer went quickly up to 1.6 MΩ with a tendency to further increase. We expect that a better thermal coupling and operation at lower temperatures would enable much higher detector performance (see the next Section). In this regard, we remark that the CUPID experiment is expected to be performed at ∼10 mK base temperature, which is in fact the value used in Cuoricino and CUORE-0, predecessors of the CUORE experiment.

We accumulated 59.6 h data with a 232Th source (consisting of a 15.2 g thoriated tungsten rod containing 1% of Th), and 190.1 h of background-only measurements, which altogether constitute 249.7 h life time. The 116CdWO4 detector was calibrated by means of the γ quanta from the environmental radioactivity (mainly emitted by 214Pb and 214Bi radionuclides from the 238U chain) and in calibration run by γ quanta from the 232Th source (mainly 228Ac and 208Tl, daughters of 232Th). The rear side of the LD was permanently irradiated by a weak 55Fe X-ray source. In addition, an optic fiber was mounted inside the cryostat to transmit LED light pulses to the LD every 30 s, which can be also used for calibration/stabilization purposes.

### Table 1

| Detector | RNTD (MΩ) | SNTD (nV/keV) | FWHM_{Bsl} (keV) | τ_R (ms) | τ_D (ms) |
|----------|-----------|---------------|-----------------|----------|----------|
| LD       | 0.12      | 258           | 0.6             | 1.3      | 4.7      |
| 116CdWO4 | 1.0       | 135           | 1.5             | 5.1      | 28.5     |

#### 3 Results and discussion

The collected data were processed off-line by applying the optimum filtering procedure [47] and several pulse-characterizing parameters were evaluated for each recorded signal: the pulse amplitude, the rise-(τR) and decay-(τD) times, several pulse-shape indicators, and the DC baseline level of the pre-triggered part (over 0.15 s). In addition, the energy resolution of the filtered baseline noise (FWHM_{Bsl}) and the amplitude of the signal (SNTD) for a given deposited energy were estimated for each data set (1–3 days of measurements). Some of these parameters, characterizing the performance of the 116CdWO4 scintillating bolometer and the LD, are given in Table 1.

Taking into account the expected high light yield of cadmium tungstate at low temperatures (e.g., ∼17 keV/MeV [38]), we have chosen a light detector with a relatively modest performance, as it is visible from Table 1. Therefore, we were not able to separate clearly the 55Fe X-ray doublet (at 5.9 and 6.5 keV) from the noise due to the poor energy resolution (FWHM_{F_{55}} ≈ 0.7 keV). However, the LD time characteristics (τR and τD of the scintillation signals) are similar to that of devices instrumented with small-size NTD Ge sensors (e.g., see the performance of a first batch of six LDs preliminary tested for the CUPID-0 detector array with Zn^{62}Se scintillating bolometers [42]).

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1. Here the rise-time is defined as the time interval between 10 and 90% of the maximum amplitude of the signal for the rising edge, while the decay-time corresponds to the time interval between 90 and 30% of the maximum amplitude of the signal for the decaying edge.

2. Here we define “light yield” as the ratio between light and heat signal amplitudes (converted into detected energy), which of course is lower than the absolute light yield of CdWO4.
by the 34.5 g 116CdWO4 bolometer in a composition [15,26,36–38]. This confirms that CdWO4 is bolometers produced from cadmium with natural isotopic composition and can be used to enhance the energy resolution of the heat channel [38]. The improvement is shown in Fig. 4, where the FWHM values of the most intensive γ peaks before and after applying the anticorrelation correction are presented. It is evident from Fig. 4 that the achieved improvement is quite modest (around 10%) in contrast to the results of Refs. [36,38]. This may be explained by a higher uniformity of the light collection from our smaller sample, which is expected to make the light-heat anticorrelation less significant. The energy resolution can be improved further in an underground cryostat shielded against environmental γ radiation.

The data of the heat-light coincidences can be transformed into the so-called Q-plot shown in Fig. 5. The projection of the points on the γ-axis can be used to evaluate the light...
reported for CdWO$_4$-based scintillating bolometers. In particular, approximately twice lower values were obtained in Ref. [38] (however, the crystal used in that study was an order of magnitude larger in volume). This excellent result is obtained by the twice-larger area of the LD, the overall compact geometry of the arrangement (which enhances the light collection), a high optical transmittance of the material [30], and a low self-absorption of the scintillation photons in our relatively thin sample. By using the LYS, one can also estimate the quenching factors for $\alpha$’s and nuclear recoils as 0.175(3) and 0.084(3), respectively.

To evaluate the discrimination power (DP) between $\gamma$($\beta$) and $\alpha$ event distributions, the LY data shown in Fig. 5 were used within the 2.6–7 MeV energy range and the 4–38 keV/MeV LY interval (cutting most of the pile-up events in the vicinity of the $\alpha$ clusters). The obtained distributions were fitted by Gaussian functions to estimate their mean values ($\mu_{\gamma(\beta)}, \mu_{\alpha}$) and standard deviations ($\sigma_{\gamma(\beta)}, \sigma_{\alpha}$). After defining

$$\text{DP} = (\mu_{\gamma(\beta)} - \mu_{\alpha})/\sqrt{\sigma_{\gamma(\beta)}^2 + \sigma_{\alpha}^2},$$

as usually done for scintillating bolometers [17], we obtain DP $= 17 \pm 1 \text{(stat.)}^{+3}_{-2} \text{(syst.)}$ in an energy interval which includes $Q_{2\beta}$ of 116Cd. The systematic uncertainty is related to the LY and pulse-shape cuts. This high value for the DP, which can be even improved in underground conditions, is compatible with a full suppression of $\alpha$-induced background in the $0\nu2\beta$ decay ROI of 116Cd.

The radioactive contamination of the 116CdWO$_4$ crystal was estimated by using the energy spectrum of the $\alpha$ events, presented in Fig. 6. The events were selected under the condition that the associated LY be below 10 keV/MeV. The peaks of 238U, 234U, and 210Po were identified in the data. The $\alpha$ events outside the energy regions expected for U/Th with their daughters can be explained by a surface pollution of the 116CdWO$_4$ detector or/and of the surrounding construction materials (which did not undergo an accurate cleaning process). Therefore, we have estimated the specific activities of the nuclides, while for other members of the U/Th process). Therefore, we have estimated the specific activities of the nuclides, while for other members of the U/Th chains only limits were obtained by using the procedure recommended by Feldman and Cousins [50]. The estimations of the 116CdWO$_4$ crystal scintillator radioactive contamination are presented in Table 2. Data on radioactive contamination of the 116CdWO$_4$ crystal No. 1 described in Ref. [34] are also reported.

It should be noted that the 116CdWO$_4$ sample No. 1 was cut from the same crystal boule, however our sample was closer to the beginning of the boule. Therefore, the hint of a lower specific activity of 238U and 210Po in the present sample (see Table 2) can be explained by segregation of uranium and lead (210Po being originated by 210Pb) in the CdWO$_4$ crystal growth process. As it was observed in Refs. [30,51,52] the radioactive contamination of the crystal boule by 228Th.
segregation of the radioactive impurities in the CdWO₄ crystal. It significantly improves the boule contamination. These features indicate a strong growth by potassium, radium, and thorium, exceeding the boule. Besides, the contamination of the residuals after the crystal growth by potassium, radium, and thorium exceeds the boule. The radioactive contamination of the 116CdWO₄ crystal scintillator. Data on the radioactive contamination of the 116CdWO₄ crystal No. 1 [34] are given for comparison.

| Chain | Nuclide (sub-chain) | Activity (mBq/kg) |
|-------|---------------------|-------------------|
|       |                     | This work          | No. 1 [34]    |
| 232-Th | 232-Th              | ≤0.13              |               |
| 228-Ra | 228-Ra              | ≤0.07              | 0.031(3)      |
| 238-U  | 238-U               | 0.3(1)             | 0.5(2)        |
| 234-U  | 234-U               | 0.26(9)            |               |
| 230-Th | 230-Th              | ≤0.07              |               |
| 226-Ra | 226-Ra              | ≤0.07              | ≤0.005        |
| 210-Po | 210-Po              | 0.23(8)            | 0.6(2)        |
| 235-U  | 235-U               | ≤0.13              |               |

4 Conclusions

A cadmium tungstate crystal scintillator with a mass of 34.5 g, enriched in 116Cd to 82 %, was tested over ~250 h at 18 mK as a scintillating bolometer in an above-ground cryogenic laboratory. The 116CdWO₄ detector exhibits an energy resolution of the order of ~2–7 keV FWHM for 0.2–2.6 MeV γ quanta. The expected value at the 116Cd double-beta decay transition energy (2813 keV) is ~7.5 keV FWHM. This result is inferior to that obtained with Ge-diodes for 0ν2β decay search in the candidate 76Ge (ΔE_FWHM ∼ 3 keV at Q(76Ge) = 2039 keV [53]) or with other bolometric materials like TeO₂, used to investigate the 0ν2β candidate 130Te (ΔE_FWHM ∼ 5 keV at Q(130Te) = 2527 keV [54]), but the energy resolution is remarkably high and can be improved as discussed above. We observed an almost complete discrimination between β(γ) and α events (a discrimination power of ~17 was achieved in the 2.6–7.0 MeV region). These promising results were obtained in spite of a significant pile-up effect related to the above-ground location of the set-up.

We have found that the energy-to-voltage conversion and the time characteristics of the 116CdWO₄ signals are similar to those observed earlier with CdWO₄-based bolometers not produced from enriched material and sharing an akin detector design. The light yield observed in the present investigation is about twice higher (31 keV/MeV for γ quanta) than that given in the literature for CdWO₄ scintillating bolometers thanks to the high optical quality of the enriched scintillator and an efficient collection of the scintillation light in the detector module.

The radioactive contamination of the 116CdWO₄ crystal by 238U, 234U, and 210Po is estimated to be on the level of ~0.3 mBq/kg each, which is lower than that in the 116CdWO₄ crystal samples cut from the same crystal boule farther away from the growth cone (from which the studied sample was obtained). This observation indicates a segregation of uranium and lead in the CdWO₄ crystals growth process. For other α emitters belonging to the U/Th chains only limits on the level of 0.07–0.13 mBq/kg were obtained.

The present work demonstrates that 116CdWO₄ scintillating bolometers represent one of the most promising technologies for a next-generation bolometric experiment aiming at exploring the inverted hierarchy region of the neutrino mass, as discussed in the CUPID project.

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Fig. 6 Energy spectra of α events accumulated by the 34.5 g 116CdWO₄ bolometer over 250 h of data taking. The energy scale corresponds to γ energy calibration. The bin width is 10 keV. The ~6–7% shift of the α peaks from the nominal Qα values is caused by a thermal quenching (see details in Ref. [38]).
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