

Detection of the natural α decay of tungsten

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The natural α -decay of ^{180}W has been unambiguously detected for the first time. The α peak is found in a (γ , β , and neutron)-free background spectrum. This has been achieved by the simultaneous measurement of phonon and light signals with the CRESST (Cryogenic Rare Event Search with Superconducting Thermometers) cryogenic detectors. A half-life of $T_{1/2} = (1.8 \pm 0.2) \times 10^{18}$ years and an energy release of $Q = [2516.4 \pm 1.1(\text{stat.}) \pm 1.2(\text{sys.})]$ keV have been measured. New limits are also set on the half-lives of the other naturally occurring tungsten isotopes.

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I. INTRODUCTION

The α decay of the naturally occurring isotopes of tungsten (W) has been the subject of experimental search for many decades. Tungsten is an interesting element because α decay is energetically allowed for all five naturally occurring isotopes. Mass excess measurements [1] show that the available decay energy Q for all these isotopes is low (<3 MeV) and that the Q values lie in the same energy range as β and γ decay due to natural chains (see Table I). Therefore, background suppression and possibly event-by-event discrimination against γ 's and β 's, which may obscure the α signal, are crucial issues for the detection of such rare events.

While ^{182}W , ^{183}W , ^{184}W , and ^{186}W are expected to have half-lives well above 10^{32} years, the isotope ^{180}W is expected to have a half-life near 10^{18} years, and is close to observability with present techniques. Most recently, the Kiev-Firenze Collaboration, operating CdWO_4 scintillation detectors in the Solotvina mine reported a “first indication of a possible α decay of ^{180}W ” with a half-life of $T_{1/2} = 1.1^{+0.8}_{-0.4}(\text{stat.}) \pm 0.3(\text{sys.}) \times 10^{18}$ years [5]. It is well known [6] that the scintillation yield for α particles is lower than that for β or γ particles of the same energy. After pulse shape analysis, an α -peak at ~ 300 keV was interpreted as ^{180}W α decay. Due to the low energy resolution [full width at half maximum (FWHM) = 110 keV] it was impossible to exclude alternative explanations for the peak, hence, the result was treated as an indication. They also set a 90% C.L. lower limit of $T_{1/2} \geq 0.7 \times 10^{18}$ years. Also the ROSEBUD collaboration, operating a 54 g CaWO_4 scintillating bolometer at the Can-

franc Underground Laboratory, obtained a limit of $T_{1/2} \geq 1.7 \times 10^{17}$ years at 90% C.L. [7].

The high sensitivity and excellent energy resolution of low-temperature detectors (see for example [8,10]) and the great efforts made to suppress and understand radioactive background in dark matter searches and double- β decay experiments make such an apparatus sensitive to rare nuclear decays. For example, the α decay of ^{209}Bi (considered as the heaviest stable isotope) has been recently detected by a French group developing low-temperature bolometers for dark matter direct detection [11]. CRESST (Cryogenic Rare Event Search with Superconducting Thermometers) is a low background cryogenic facility primarily devoted to the direct detection of Weakly Interactive Massive Particle (WIMP) dark matter particles via their scattering by nuclei. Such nuclear recoils resulting from WIMP interaction can be dis-

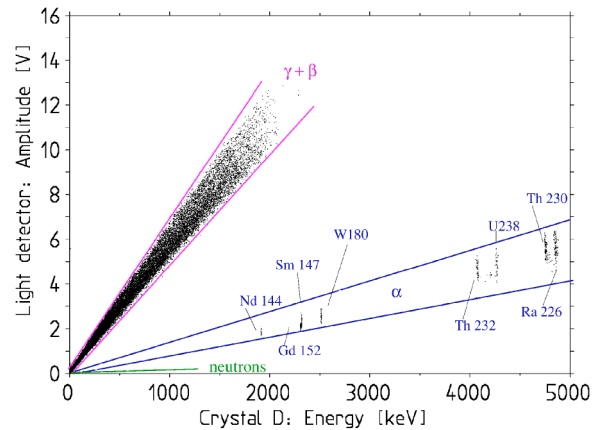


FIG. 1. Pulse amplitude in the light channel vs energy in the CaWO_4 crystal D from run 28. Three clearly separated event populations appear. The solid lines border the three different bands: the upper band is due to γ 's and β 's, the lowest one to neutrons and the middle one is due to α 's. Each peak in the α band has been identified (see text) and is here labeled accordingly.

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TABLE I. Representative Isotopic Composition according to the IUPAC, best single measurements on isotopic abundance from individual references, and decay energies for the isotopes of natural tungsten.

Isotope	Abundance (%)		Q (keV)	
	IUPAC [3]	Ref. ^a [4]	Ref. [1]	Ref. [2]
^{180}W	0.12(1)	0.1198(2)	2516(5)	2508(4)
^{182}W	26.50(16)	26.4985(49)	1774(3)	1771.8(2.2)
^{183}W	14.31(4)	14.3136(6)	1682(3)	1680.0(2.2)
^{184}W	30.64(2)	30.6422(13)	1659(3)	1656.2(2.2)
^{185}W	28.43(19)	28.4259(62)	1123(7)	1124(7)

^aConsidered by the IUPAC (International Union of Pure and Applied Chemistry) to be the best single measurements.

criminated from electron background (caused by photons or electrons) by measuring phonons and scintillation light simultaneously. The energy detected via phonons in a cryogenic detector is, to first order, independent of the nature of the particle. There is, however, a significant difference in scintillation yield for nuclei and electrons of the same energy. The fraction of energy released in the nuclear interaction channel, negligible for photons and electrons, becomes important for α 's and heavy ions and is dominant for WIMPs and neutrons. As a result WIMP- and neutron-induced recoils give considerably less scintillation light than electrons of the same energy, while α particles can be clearly identified because they interact partly with both nuclei and electrons. This results in a pure α spectrum, i.e., one without contributions from β , γ , and neutron events as in the middle band of Fig. 1. Finally, we remark that the energy measured following an internal α decay in a cryogenic detector corresponds to the sum of the energies of the α particle and of the recoiling nucleus, i.e., the total decay energy Q .

In this paper we present clear evidence for the α decay of ^{180}W from early runs of the CRESST II dark matter detectors, originating from the tungsten in the CaWO_4 crystals used as the dark-matter target.

II. EXPERIMENTAL SET-UP

CRESST is an ultralow background facility operating at the Gran Sasso underground laboratories. Detailed descriptions can be found in [8,9]. CRESST has developed very

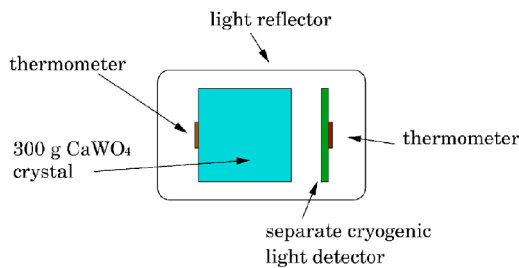


FIG. 2. Schematic of a detector module. It consists of a scintillating 300 g CaWO_4 crystal (phonon channel) and a Si wafer (light channel), both read out by a SPT. The setup is surrounded by a reflective foil.

sensitive cryogenic detectors consisting of a dielectric target crystal with a superconducting phase transition thermometer (SPT) evaporated onto one surface. Particle interactions in a cryogenic detector produce phonons that propagate to the surface where they heat the electron system in the SPTs. These thermometers, biased in the middle of their superconducting-to-normal transition near a temperature of 10 mK, transform the temperature rise of their electrons into a relatively large increase of the resistance of the film. The resistance change of the low impedance thermometer is then measured via a superconducting quantum interference device (SQUID). The resistance increase is a measure for the energy deposited.

The setup for the simultaneous detection of scintillation light and phonons is described in [9] and shown schematically in Fig. 2 for one absorber module. It consists of two independent detectors, each one with its own SPT with SQUID readout. The main detector consists of a ~ 300 g cylindrical CaWO_4 crystal ($\varnothing = 40$ mm, $h = 40$ mm). CaWO_4 is a well-known scintillator with high light yield and very broad emission spectrum peaked at 420 nm. It is characterized by a high index of refraction ($n = 1.92$) and a decay time of 17 μs at 77 K. The scintillation light produced in each target crystal is detected via an associated calorimeter consisting of a silicon wafer of $(30 \times 30 \times 0.45)$ mm³ volume

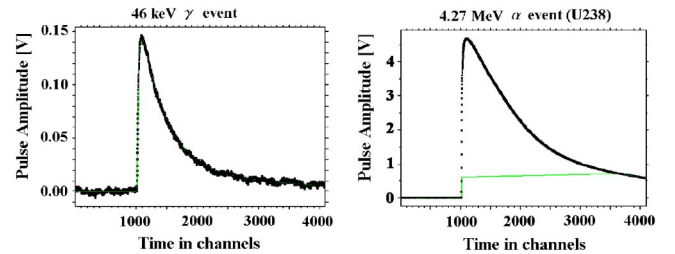


FIG. 3. Fit of two events from crystal D (run 28) with a template obtained by averaging pulses from the 122 keV peak. The fit is truncated at 0.7 V. For pulses of amplitude below this value the fit procedure is standard (left plot). For pulses with an amplitude exceeding this value, the higher part of the pulse shape (above the horizontal dotted line in the right plot) is distorted due to saturation. Therefore, the fit is truncated, i.e., fit only to data points below 0.7 V, and the amplitude of the pulse is reconstructed from the template.

TABLE II. Results from the ICPMS analysis at the Gran Sasso facility on rare earths, hafnium, and osmium. For the semiquantitative analysis the calibration curve is determined with a multielement standard, which may be different from the one to be analyzed. On some elements, in particular on samarium, a quantitative analysis was performed. In this case the calibration is obtained using known standards of the element to be analyzed. All the samples are dissolved in a HNO_3 solution in the microwave oven with different exposures for samples I and II.

Natural element	Crystal B		Crystal E		
	Semiquantitative analysis		Quantitative analysis		Semiquantitative analysis
	Sample I (ppb)	Sample II (ppb)	Sample I (ppb)	Sample II (ppb)	(ppb)
Nd	555 $\pm$ 167	773 $\pm$ 232	683 $\pm$ 137	994 $\pm$ 199	1100 $\pm$ 330
Sm	<22	11 $\pm$ 3.3	9 $\pm$ 2	13 $\pm$ 3	<6
Eu	<8	4 $\pm$ 1.2	4 $\pm$ 1	5 $\pm$ 1	<2
Gd	2898 $\pm$ 869	4084 $\pm$ 1225			1000 $\pm$ 300
Er	1328 $\pm$ 398	1877 $\pm$ 566			940 $\pm$ 283
Dy	<15	<3			<3
Hf	<22	<4			<3
Os	<29	<5			<4

with a 20 nm thick SiO_2 layer on both surfaces. In CaWO_4 only a few percent of the absorbed energy is transformed into light. To minimize light losses the whole module is thus enclosed in a highly reflective foil.

In the data to be presented here, three different CaWO_4 crystals (referred to as crystals B, D, and E) were used in the Gran Sasso setup for a total of four different CRESST runs (22, 23, 27, 28). For detector operation, the temperature of the thermometer is controlled by a dedicated heater. Additionally the heater is used to inject test pulses that monitor the long-term stability of the detector [9]. In particular, in the runs 27 and 28 a large-voltage pulse was produced, reaching the saturation region of the superconducting transition specifically to monitor the detector response above 1 MeV. This pulse was sent, at a rate of 0.5 Hz, throughout the measuring period. A record length of 4096 samples and a time base of 40 μs , resulting in a time window of 164 ms for recording an event, was chosen for all the runs presented.

III. OFF-LINE ANALYSIS

CRESST detectors are optimized to be most sensitive in the energy region relevant for dark-matter direct detection (<200 keV). In this energy range the detector response is mapped out with electric heater test pulses, and the 122 keV line from a ^{57}Co source provides the calibration of the test pulses in terms of a γ -equivalent energy [8]. A pulse-shape template is obtained by averaging pulses from the 122 keV line. This template is used in a fit to the actual signal pulses in order to accurately determine the signal amplitude. This is valid as long as the detector response is in a linear regime. If the response becomes nonlinear, then the pulse shape changes and the same template cannot be used over the full energy range.

Typically, the pulse shape for the CaWO_4 detectors can be described by $\tau_{\text{rise}}=1.1$ ms and $\tau_{\text{decay}}=30$ ms. However, the temperature rise caused by α decays is usually beyond the dynamic range of the SPT and for precise energy determination the signal duration rather than the pulse height must be used. Therefore, two analysis methods, described in [12], were developed in order to reconstruct the higher energies (MeV region), for which the detector response is nonlinear.

In the first method (Method A), a template from pulses in the linear region (for example, the 122 keV template from the calibration) is used to fit all measured pulses with an amplitude below the voltage at which the response becomes nonlinear. For all pulses with an amplitude exceeding that voltage, the fit is truncated and the upper part is reconstructed from the template. This is shown in Fig. 3. The resulting amplitude spectrum is then linear by construction.

If the pulse duration is too long, then the information from the decaying part of the pulse is outside the recording time window and the method fails. To overcome this limitation a second method was developed (Method B). The template from the linear region is fitted with a function following the model described in [16]. This assumes an exponential rise and two exponential decay components. Once the parameters of the model function are determined for the linear region, a mathematical description of the changes in pulse shape in the nonlinear range is needed. At higher energies the pulse shape varies according to the nonlinearities of the superconducting phase transition. These changes can be described via successive approximation to the model function. The higher-order correction coefficients are extracted by fitting templates of different energies up to 2.31 MeV (see [12]), enabling the energy spectrum to be reconstructed up to the highest recorded α signal (α - α cascade at ~ 15 MeV).

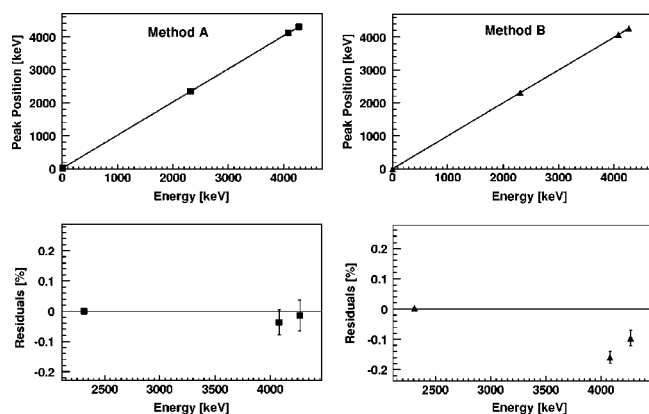


FIG. 4. Top: Fitted peak position vs known energy. The energy scale for the y-axis is fixed by the ^{147}Sm peak, while the position of the ^{232}Th and ^{238}U peaks results from a simple linear extrapolation of the calibration for both methods A and B. Bottom: Residuals of the fitted position from the expected position. For method A, the ^{232}Th peak results at (4081.3 ± 1.7) keV [$\Delta E = (28 \pm 4)$ keV (FWHM)], and the ^{238}U at (4269.4 ± 2.2) keV. For method B, the peaks lie at (4076.3 ± 0.8) keV [$\Delta E = (14.1 \pm 1.7)$ keV (FWHM)], and (4265.9 ± 1.1) keV, respectively. The calibration of the amplitude spectrum resulting from method A is much more accurate; however, the resolution is less compared with method B.

IV. BACKGROUND

Due to the powerful discrimination technique CRESST II detectors offer, contaminations from natural decay chains and other α -unstable isotopes have been identified by their α decays with a sensitivity of $\sim 1 \mu\text{Bq/kg}$. The results of this analysis have shown very good agreement with the results obtained using different techniques commonly employed to determine the presence of impurities in crystals. These include ICPMS¹ (Inductively Coupled Plasma Mass Spectrometry), HPGe² (High Purity Germanium γ spectroscopy) and x-ray luminescence³ techniques.

Evidence for contaminations due to α -unstable rare-earth elements has been found in all the measured detectors. Rare-earth doped crystals are commonly produced for laser applications, therefore, these impurities are likely to have been introduced during the production of the crystals. In particular, an α peak at 2.31 MeV has been detected and identified as the α decay of ^{147}Sm . This isotope is naturally occurring with 15% isotopic abundance and is known to be a pure α -unstable radionuclide with $T_{1/2} = 1.06 \times 10^{11}$ years. The intensity of this peak varies for the different detectors, implying different levels of contamination in each crystal. Crystal B showed a count rate of (1.7 ± 0.1) counts/h, which translates into (13 ± 1) ppb of natural samarium in the crystal. The rate on the same peak measured with crystal E corresponds to a contamination of (5.5 ± 0.4) ppb of natural samarium.

ICPMS analyses were performed in the Gran Sasso facility on crystals B and E giving results, shown in Table II in

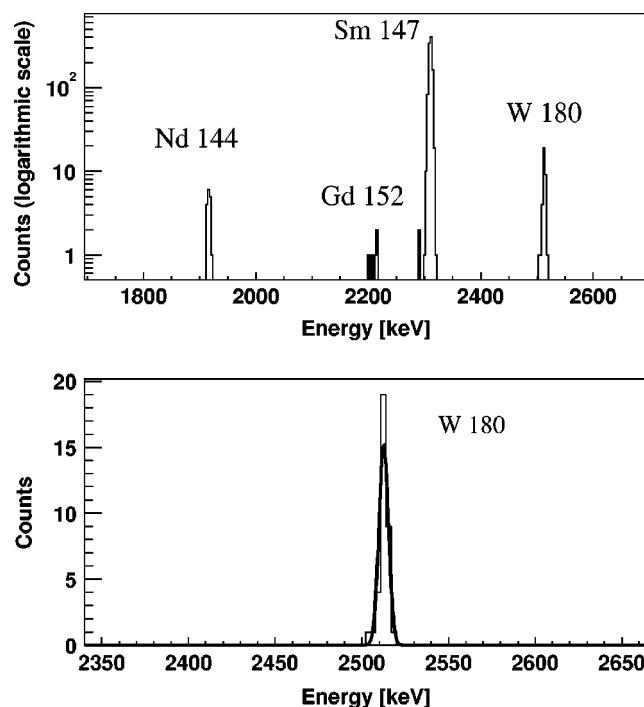


FIG. 5. Alpha events measured in Run 28. In the top figure the energy spectrum between 1.7 and 2.7 MeV is shown with 3 keV bins. On the vertical axis, the number of counts are displayed on a logarithmic scale. In the bottom figure the fit to the ^{180}W α decay is shown. No background counts are present in a wide-energy interval around the peak.

agreement with our analysis of the α -peak intensities. Evidence of ^{144}Nd [$Q=1905.1$ keV, isotopic abundance (i.a.) = 23.8%, $T_{1/2} = 2.29 \times 10^{15}$ years] and ^{152}Gd ($Q=2205$ keV, i.a.=0.2%, $T_{1/2} = 1.08 \times 10^{14}$ years) [18] α decay has also been observed, and the results are in accordance with the ICPMS analysis. Furthermore, x-ray luminescence analysis on crystal B showed the presence of the characteristic emission peak of Gd and Er [17], the two most abundant rare-earth contaminants (see Table II).

Finally, alternative candidates, such as ^{174}Hf ($Q = 2496$ keV, i.a.=0.162%, $T_{1/2} = 2.0 \times 10^{15}$ years) and ^{186}Os

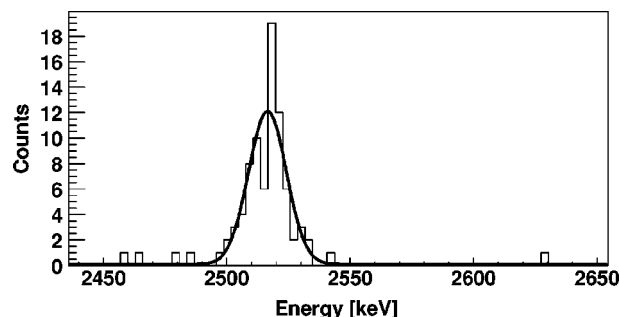


FIG. 6. Sum of the four different runs, each analyzed independently with method A. The resulting peak is fitted giving an energy of (2516.4 ± 1.1) keV with resolution FWHM = (18 ± 2) keV. Few background events due to a surface contamination of the Ag foil used in run 27 are visible. The scintillating foil used in runs 22, 23 and 28 efficiently vetoes all these events (see text).

¹Gran Sasso Laboratory and Durham University [13].

²Modane Underground Laboratory and Gran Sasso Underground Laboratory [14].

³Durham University [15].

TABLE III. Half-life for the α decay of ^{180}W obtained from the present work. The results are given independently for the four different runs and for the total exposure. For the runs 27, 28, and the total exposure, the number of counts is determined by a Gaussian fit to the peak. Errors are given at 1σ . For run 22 and 23, due to the small number of counts, the total number of events contained within $\pm 3\sigma$ of the expected energy position is given.

Crystal	Run number	Exposure (kg days)	Number of counts	Half-life (years)
D	28	12.268	35 ± 4.3	$T_{1/2} = (1.7 \pm 0.2) \times 10^{18}$
D	27	11.745	28.5 ± 5.6	$T_{1/2} = (1.9 \pm 0.4) \times 10^{18}$
E	23	3.467	9	$T_{1/2} \geq 1.1 \times 10^{18}$ (90% C.L.)
B	22	1.14	4	$T_{1/2} \geq 6.8 \times 10^{17}$ (90% C.L.)
All Crystals		28.62	75.5 ± 8.7	$T_{1/2} = (1.8 \pm 0.2) \times 10^{18}$

($Q=2822$ keV, i.a.=1.58%, $T_{1/2}=2.0 \times 10^{15}$ years) [18], were considered to cause the 2.31 MeV peak. They were discarded, however, due to the unreasonably high atomic concentration needed to produce the detected rate.

V. CALIBRATION

Once unambiguously identified, ^{147}Sm was used to calibrate the full α spectrum. This allowed a precise identification of the U-Th peaks, which were then used to determine the accuracy of the calibration for both reconstruction methods A and B. In Fig. 4, the linearity of the response functions for methods A (top-left figure) and B (top-right figure) is shown for the data of run 28. On the x -axis the central values of the tabulated energies [1] of the ^{147}Sm ($Q=2310.5 \pm 1.1$ keV), ^{232}Th ($Q=4082.8 \pm 1.4$ keV), and ^{238}U ($Q=4270 \pm 3$ keV) peaks are given. On the y -axis their fitted peak position is plotted. The energy scale for the y -axis is fixed on the ^{147}Sm peak at 2310.5 keV, while the position of the ^{232}Th and ^{238}U peaks results from a simple linear extrapolation of the calibration. At the bottom of the figure the residuals of the fitted position with respect to the expected position are shown. The calibration of the amplitude spectrum resulting from method A is much more accurate, but less information can be used to reconstruct the signal amplitude compared to method B. This results in a slightly worse energy resolution $\Delta E = (12.9 \pm 0.3)$ keV (FWHM) on the ^{147}Sm peak, and it fails to completely reconstruct the higher part of the U-Th α spectrum. Method B, on the other hand,

using a larger amount of information from the pulses than method A, gives a much better resolution [$\Delta E = (6.7 \pm 0.1)$ keV (FWHM) on the ^{147}Sm peak], but the approximation of the pulse-shape description introduces a higher systematic error on the linear extrapolation of the calibration. In the following, method B has been used to fully reconstruct and identify the entire α spectrum, while method A was applied as an independent check on the peak position of the ^{180}W evidence. In particular, method A is used to extract the measured energy for the ^{180}W Q value.

VI. RESULTS

Crystal D showed the best performance as a detector [energy resolution: $\Delta E = 1.8$ keV (FWHM) for 122 keV γ 's, and $\Delta E = (6.7 \pm 0.1)$ keV for 2.31 MeV α 's, using method B], and it has the longest exposure (11.745 kg days in run 27 and 12.268 kg days in run 28). In run 28 the performance of the module was improved by replacing the Ag reflective foil used in run 27, which showed a ^{210}Pb contamination, by a highly reflective (99% at 420 nm) scintillating polymeric foil. It is these data that we discuss in full detail. A peak was observed at 2512.8 ± 0.4 (stat.) ± 4.3 (sys.) keV with a resolution $\Delta E = (6.5 \pm 0.7)$ keV (FWHM). This is shown in Fig. 5. The position in energy obtained by reconstructing the spectrum with method A is (2516.5 ± 1.4) keV. A half-life $T_{1/2} = (1.7 \pm 0.2) \times 10^{18}$ years is obtained. This is consistent with the value from run 27 on the same crystal and on crystals B and E, as shown in Table III. The consistency of the results

TABLE IV. Lower limits (90% C.L.) on the half-lives of the non-observed α -decays of the other four naturally occurring tungsten isotopes, obtained with a total exposure of 16.875 kg days. For comparison the best previously published limits are also shown [5,7].

Isotope	Half-life (years)		
	This work	Previous [5]	Previous [7]
^{182}W	$T_{1/2} \geq 7.7 \times 10^{21}$	$\geq 1.7 \times 10^{20}$	$\geq 2.5 \times 10^{19}$
^{183}W	$T_{1/2} \geq 4.1 \times 10^{21}$	$\geq 0.8 \times 10^{20}$	$\geq 1.3 \times 10^{19}$
^{184}W	$T_{1/2} \geq 8.9 \times 10^{21}$	$\geq 1.8 \times 10^{20}$	$\geq 2.9 \times 10^{19}$
^{186}W	$T_{1/2} \geq 8.2 \times 10^{21}$	$\geq 1.7 \times 10^{20}$	$\geq 2.7 \times 10^{19}$

from different crystals tends to confirm that the signal originates from the ^{180}W decay and not from unknown impurities, which may be expected to vary for each individual crystal, as seen in Sec. IV. Furthermore, ICPMS measurements performed on these two crystals reported in Table II exclude alternative explanations for the tungsten peak, such as ^{174}Hf .

Finally, the four measurements were added, resulting in a spectrum for a total exposure of 28.62 kg days. The data from each run have been analyzed with method A, calibrated individually, and then summed. This results in half-life of $T_{1/2} = (1.8 \pm 0.2) \times 10^{18}$ years, which is consistent with the previously published limits. Including the uncertainty on the position of the ^{147}Sm line, a peak of energy $[2516.4 \pm 1.1 (\text{stat.}) \pm 1.2 (\text{sys.})]$ keV is found with a resolution $\Delta E = (18 \pm 2)$ keV (FWHM), as shown in Fig. 6. This result is consistent with expectations based on the mass difference of ^{180}W and ^{176}Hf from Ref. [1], but lies 2σ away from the latest update [2] (see Table I). We note, however, that most of the input data used to evaluate the atomic masses consists of relative measurements that set a relation in mass or energy among two or more nuclei. In the diagram showing the relations among input data (Fig 1 of Ref. [2]), ^{180}W and ^{176}Hf are only weakly connected and an underestimation of the errors is plausible. Our result represents the missing direct connection between ^{180}W and ^{176}Hf .

Limits on the half-lives for the nonobserved α decays of the other four naturally occurring tungsten isotopes were also calculated. For this purpose the data from run 27 could not be used. In that run, a Ag foil was used to surround the crystal, while in the other three runs the scintillating polymeric foil was used. Any contamination of α -unstable isotopes, such as ^{210}Po on the surface of the foil or on the crystal, may result in α events of degraded energy from the U-Th chain energy, going down to ~ 100 keV. These

were observed in run 27 with a rate of 0.087 counts/100 keV/kg/day both between ~ 500 keV and the ^{144}Nd peak at 1.9 MeV, and between the ^{180}W and ^{232}Th peaks. The polymeric foil used in all the other runs did not show any surface contamination. Furthermore, if the scintillating side of the foil faces directly the crystal, as in run 28, then it efficiently vetoes all the surface events due to the light emitted by the polymeric foil itself. The limits obtained from the sum of runs 22, 23, and 28 are reported in Table IV.

VII. CONCLUSIONS

We have observed the natural α decay of ^{180}W with a half-life of $T_{1/2} = (1.8 \pm 0.2) \times 10^{18}$ years and a Q -value of $[2516.4 \pm 1.1 (\text{stat.}) \pm 1.2 (\text{sys.})]$ keV. In addition, the lack of any signal from the other tungsten isotopes sets new limits on their half-lives. These results offer an improvement of roughly a factor of 50 over the best previously published limits. Finally, we would like to note the power and potential of this cryogenic technique, which here enabled us to achieve a very high sensitivity in an application to nuclear physics.

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