Search for α decay of naturally occurring tungsten isotopes

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(Submitted 3 May 1995)
Pis'ma Zh. Éksp. Teor. Fiz. **61**, No. 11, 869–873 (10 June 1995)

An experiment has been carried out at the Solotvin underground laboratory with cadmium tungstenate and zinc tungstenate scintillation crystals. The results put a lower limit of 10^{17}–10^{19} yr on the lifetime of naturally occurring tungsten isotopes with respect to α decay.

Theoretically, all five naturally occurring isotopes of tungsten are unstable with respect to α decay,[1,2] but this decay has yet to be reliably observed for any of them.[3–8] According to the calculations of Refs. 1 and 2, only the α decay of ^{180}W could be detected at a reasonable sensitivity of the measurement procedure ($T_{1/2} \approx 10^{18}$ yr). Studies of the other isotopes are of interest only for observing any anomalous effects which are not predicted by the existing theory and which are capable of increasing the probability of this decay by many orders of magnitude (Table I).

In the search for a possible α activity of tungsten which we are reporting here, we made use of measurements of the background of scintillation crystals of cadmium tungstenate ($CdWO_4$, with a volume of 56.8 cm^3 and a mass of 452 g, and $^{116}CdWO_4$ with 11.4 cm^3 at 91.5 g) carried out as part of a study of the 2β decay of ^{116}Cd (Ref. 11) and of the β decay of ^{113}Cd (Ref. 12). In addition, we measured the background of a small crystal of zinc tungstenate, $ZnWO_4$ (with a volume of 0.6 cm^3 and a mass of 4.5 g).

All these studies were carried out at the Solotvin underground laboratory, in a salt mine at a depth of more than 1000 meters water equivalent. The flux of cosmic-ray muons is suppressed by a factor of 10^4 at this depth. Because of the low radioactive contamination of salt, the natural γ background in the salt mine is lower by a factor of 30–70 than in other underground laboratories, surrounded by ordinary materials.[13]

The low-background apparatus used in the measurements is described in detail in Refs. 11 and 12. There is passive shielding consisting of oxygen-free copper (5–8 cm), mercury (8–10 cm), lead (23 cm), and polyethylene (24 cm). The cadmium tungstenate crystals were monitored from two sides by separate FÉU-110 photomultipliers through plastic optical waveguides 25 cm long. The special shape of these waveguides prevented essentially any degradation of the energy resolution of the detectors. The resolution was 10.7% at an energy of 662 keV for the $CdWO_4$ crystal (4 cm in diameter, 4.5 cm long), while it was $\approx 13\%$ for the $^{116}CdWO_4$ and $ZnWO_4$. The spectrometers were subjected to a periodic energy calibration with the help of ^{137}Cs, ^{207}Bi, ^{232}Th, and ^{241}Am sources.

TABLE I. α decay of naturally occurring isotopes of tungsten.

Isotope	Abundance[9]	Q_α, keV (Ref. 10)	Theory $T_{1/2}$ yr (Ref. 1)	Experiment: $\lim T_{1/2}$, yr Ref. 6	Present study
^{180}W	0.13(4)%	2514(5)	2.5×10^{18}	1×10^{15}	7.4×10^{16}
			7.5×10^{17} (Ref. 2)	9×10^{14} (Ref. 8)	
^{182}W	26.3(2)%	1773.5(29)	1.6×10^{33}	2.1×10^{17}	8.3×10^{18}
^{183}W	14.3(1)%	1681.9(28)	6.3×10^{38}	1.1×10^{17}	1.9×10^{18}
					$1.0\times10^{19}(1/2^-\rightarrow1/2^-)$
^{184}W	30.67(15)%	1658.3(28)	3.2×10^{36}	2.5×10^{17}	4.0×10^{18}
				3.0×10^{17} (Ref. 7)	
^{186}W	28.6(2)%	1123(7)	2.5×10^{57}	2.3×10^{17}	6.5×10^{18}

The detection system used in these experiments consists of a microcomputer, a tape drive, and a CAMAC crate with an analog-to-digital converter (ADC) and a timer. Since the emission time of the $CdWO_4$ and $ZnWO_4$ scintillators is 10–20 μs, some special electronic units were used in all the measurements. These units integrated the signals from the photomultiplier anodes over a time ≈40 μs and generated the short output pulses required for normal operation of the ADCs. Scintillation signals from photomultiplier noise pulses were rejected in the same electronic units by the technique of pulse shaped discrimination. The effect was to reduce the detection threshold of the $CdWO_4$ to 30–40 keV.

A very important parameter of the detectors used in a search for α decay is the energy-dependent ratio of the light yields for α and β particles (the so-called α/β ratio). Since the values found for this ratio with the help of an external α source are afflicted with large errors, which are difficult to eliminate (because of defects of the surface layer, self-absorption of α particles, etc.), this ratio was measured by a method involving α decay of nuclides from the ^{232}Th, ^{235}U, and ^{238}U families, which are present in trace amounts in the crystals used.[11] For this purpose we developed and used a method for analyzing the temporal distributions of background events.[11] This method involves, for example, a search for the following chain of two α decays which lead to the ^{232}Th family:

$$^{220}\text{Rn}(E_\alpha=6.29\ \text{MeV};\quad T_{1/2}=55.6\ \text{s})\rightarrow{}^{216}\text{Po}(E_\alpha=6.78\ \text{MeV};\quad \text{T}_{1/2}=0.15\ \text{s})\rightarrow{}^{212}\text{Pb}.$$

This analysis yielded the following approximate function to describe the behavior of the α/β ratio as a function of the energy of the α particles (E_α, MeV) for the $CdWO_4$ and $^{116}CdWO_4$ crystals: $\alpha/\beta=0.14(3)+0.009(5)E_\alpha$. A similar function was found for the $ZnWO_4$ crystals.

Figure 1 shows the background spectra of the $CdWO_4$ crystal (4 cm in diameter, 4.5 cm long) and the $^{116}CdWO_4$ crystal (2.8 cm in diameter, 1.8 cm long), found in measurements over 433 and 951 h, respectively. The abrupt rise below an energy of 350 keV here is the spectrum of the quadruply forbidden β decay of ^{113}Cd [$T_{1/2}=(9.3\pm1.9)\times10^{15}$ yr, threshold energy of 316 keV; Ref. 10]. The concentration of this isotope in the enriched crystal is[11] 2.15(20)%, while that in a naturally occurring crystal is[9] 12.22(4)%. The decay half-life and the shape of the β spectrum of ^{113}Cd, which are

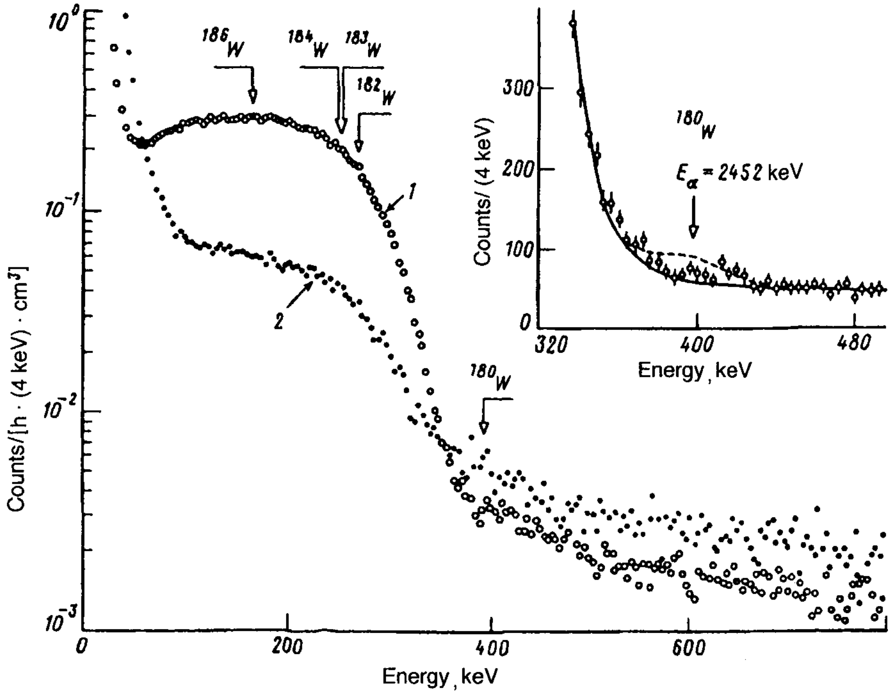

FIG. 1. Spectra of the background of the crystals. 1—$CdWO_4$ (4 cm in diameter, 4.5 cm long, measurement time of 433 h); 2—$^{116}CdWO_4$ (2.8 cm, 1.8 cm, 951 h). The inset shows part of the spectrum of the $CdWO_4$ detector and an approximation of it. The area under the α peak of ^{180}W (460 counts), which has been ruled out at a 90% confidence level, corresponds to $\lim T_{1/2}(^{180}W) = 7.4\times 10^{16}$ yr.

important in the construction of a model for the background, were refined in the measurements we mentioned earlier involving a $CdWO_4$ crystal:[12] $T_{1/2} = (7.7\pm 0.3)\times 10^{15}$ yr.

Since there are no structural features in the background spectra which clearly indicate an α activity of tungsten, these results were used to estimate a limiting probability for this process. A limit on the decay half-life, $\lim T_{1/2}$, was calculated from the formula $\lim T_{1/2} = \ln 2\epsilon t N_n / \lim S_e$, where N_n is the number of nuclei of the isotope of interest, ϵ is the total detection efficiency, t is the measurement time, and $\lim S_e$ is the limiting number of events of the effect whose hypothesized presence in the experimental spectrum can be ruled out at the given confidence level. Estimates of the latter quantity were calculated by the maximum-likelihood method in light of the recommendations of the Particle Data Group.[14] It was assumed that the experimental spectrum of the $CdWO_4$ and $^{116}CdWO_4$ crystals is described over the range 100–600 keV by a sum of three functions, two corresponding to background (the β spectrum of ^{113}Cd and an exponential function), while the third corresponds to the expected effect (a Gaussian curve). The efficiency of the α-particle detection in the crystals is essentially 100%. The position of the center of gravity of the Gaussian corresponds to the energy of the α particles for each isotope of W, but it is varied in view of the uncertainties in the coefficients in the energy dependence of the α/β ratio given above. A set of parameters (and the errors in them) corresponding to the maximum probability for observation of the measured spectrum (within the framework of the model adopted) can then be found by maximizing the likelihood function. As adjustable parameters in making this fit, we used the amplitude and constant of the exponential function and the area under the α peak.

Applying this procedure to the spectra of the $CdWO_4$ crystal (for ^{180}W) and the $^{116}CdWO_4$ crystal (for the other isotopes of W), and taking account of the measurement time and the number of nuclei (9.9×10^{20} ^{180}W, 4.00×10^{22} ^{182}W, 2.18×10^{22} ^{183}W, 4.65×10^{22} ^{184}W, 4.35×10^{22} ^{186}W), we find the following limits on $T_{1/2}$ for the α decay of the tungsten isotopes (at a confidence level of 90%):

$$\lim T_{1/2}(^{180}W)=7.4\times 10^{16}\ \text{yr}, \quad \lim T_{1/2}(^{182}W)=8.3\times 10^{18}\ \text{yr},$$

$$\lim T_{1/2}(^{183}W)=1.9\times 10^{18}\ \text{yr}, \quad \lim T_{1/2}(^{184}W)=4.0\times 10^{18}\ \text{yr},$$

$$\lim T_{1/2}(^{186}W)=6.5\times 10^{18}\ \text{yr}.$$

As an example, the inset, in linear scale, in Fig. 1 shows an approximation of part of the background spectrum of $CdWO_4$ detector in the region of the α decay of ^{180}W. The area under the α peak here (eliminated with a 90% level) is 460 counts and corresponds to $\lim T_{1/2}=7.4\times 10^{16}$ yr.

We also attempted to observe an α transition of ^{183}W to a metastable level ($T_{1/2}=18.7$ s, energy of 375 keV) of the ^{179}Hf nucleus. A characteristic of this transition is the presence of a second event, delayed with respect to the emission of the α particle. The distribution of the length of the intervals between them should correspond to $T_{1/2}=18.7$ s. With this goal in mind, we used a time-analysis method to study the results of measurements over 526 h of the background of the zinc tungstenate ($ZnWO_4$) crystal (1.4 cm in diameter, 0.4 cm long, with 1.24×10^{21} ^{183}W nuclei). In the time window of 0.1–60 s (which includes $\approx$90% of the decays of ^{179m}Hf), we did not observe any pair of events in the measured spectrum which satisfied all the criteria for α decay of ^{183}W to the metastable level of ^{179}Hf. The efficiency of the detection of this transition was calculated by the Monte Carlo method with the help of the GEANT 3.15 software package.[15] The result was 0.45. Since the number of events of the effect ruled out is 2.3 (according to Ref. 14), we find the following limit on $T_{1/2}$ for the α decay of ^{183}W to the metastable level of ^{179}Hf (confidence level of 90%):

$$\lim T_{1/2}(1/2^- \rightarrow 1/2^-)=1.0\times 10^{19}\ \text{yr}.$$

All the limits found on $T_{1/2}$ are listed in Table I. Comparison with results found previously shows that this study has made progress amounting to a factor $\approx$20–50 for each tungsten isotope. This comparison also reveals that the sensitivity of the method is very close to the level which would be necessary in order to observe α decay of ^{180}W. It is hoped that this level can be exceeded in a new experiment, presently being prepared, by reducing the background further and by increasing the volume of the detector.

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Translated by D. Parsons