Precision half-life measurement of the 4-fold forbidden electron capture of $^{50}$V

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A sensitive search of the 4-fold forbidden non-unique beta decay of $^{50}$V has been performed. A total exposure of 185.8 kg · d has been accumulated. A reliable half-life value with the highest precision so far of $(2.29 \pm 0.25) \cdot 10^{15}$ years of the electron capture decay of $^{50}$V into the first excited state of $^{50}$Cr could be obtained. A photon emission line following the 4-fold forbidden beta decay into the first excited state of $^{50}$Cr could not be observed, resulting in a lower limit on the half-life of the beta decay branch of $1.7 \cdot 10^{15}$ years. This is barely in agreement with a claimed observation of this decay branch.

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I. INTRODUCTION

In the history of particle and nuclear physics the study of weak interactions and especially of beta decay has played a vital role. Those studies helped, among others, to establish the V-A structure of weak interactions. Nowadays, this interest in exploring beta decays and related issues is rather reduced but still there are interesting topics to investigate like the endpoint measurements of tritium and $^{185}$Re electron spectra to determine the neutrino mass [1] or to search for S,T,V contributions to the weak interaction [2]. In addition to these “beyond the standard-model” searches also interesting nuclear physics questions are still open which can be addressed by studying highly forbidden beta decays. The majority of beta emitters is characterized as allowed or single forbidden, however there are a few isotopes in nature which are even at least 4-fold forbidden [3]. Their decays are extremely rare with half-lives well beyond $10^{13}$ years. Compared to those decays, which are at least 5-fold forbidden transitions (like $^{48}$Ca and $^{90}$Zr), even a double beta decay is more likely to happen.

In this paper the focus is on the 4-fold forbidden $\beta$-decay of $^{50}$V. There are only three nuclei in nature which permit a feasible study of 4-fold forbidden beta decay, namely $^{50}$V, $^{113}$Cd and $^{115}$In, all of them are non-unique ($\Delta I^\Delta = 4^+$). Half-lives of these transitions are long ($\geq 10^{14}$ years) and would produce very low count rates in typical experiments. Such measurements can only be performed in well shielded facilities with a low radioactive background. Recent measurements of half-lives of $^{113}$Cd are reported in [4] and of $^{115}$In in [7,8], the latter including also a transition into the first excited state. These activities triggered for the first time theoretical attempts to calculate the energy spectra of such beta decays [3,10].

The isotope $^{50}$V is quite unique in the sense that in contrast to $^{113}$Cd and $^{115}$In the ground state transition is even higher-forbidden, leaving only 4-fold forbidden non-unique decay modes into the first excited states of $^{50}$Cr and $^{50}$Ti, both characterized as $6^+ \rightarrow 2^+$ transitions. The ground state transitions to both isotopes are even 6-fold forbidden non-unique decays. The decay scheme is shown in Fig. 1. The Q-value for the beta decay into $^{50}$Cr is $(1037.9 \pm 0.3)$ keV and for electron capture (EC) into $^{50}$Ti it is $(2205.1 \pm 1)$ keV, respectively [11]. There is only one excited state in each daughter nucleus which can be populated. The corresponding gamma lines to search for are $1553.77$ keV for EC into the first excited state of $^{50}$Ti and $783.29$ keV for the beta decay into the first excited state of $^{50}$Cr, respectively. The photon emission probability of both E2 transitions is 1 (with a negligible uncertainty).

Various attempts have been made to observe the decay of $^{50}$V into $^{50}$Ti [12,21]. However, the deduced half-life changed by several orders of magnitude over the decades, while uncertainties claimed were typically well beyond 20%. For the beta decay into the excited state of $^{50}$Cr only lower limits were published except one vague indication of an observation [21]. The measurements within the last 45 years are compiled in Tab. 1. All these measurements were performed more than at least two decades ago.

| Mass  | Time  | $\tau_{EC}^{1/2}$ | $\tau_{\beta}^{1/2}$ | Ref. |
|-------|-------|-------------------|----------------------|-----|
| g     | d     | $10^{17}$ a        | $10^{17}$ a          |     |
| 4000  | 48.88 | > 8.8             | > 7.0               | [18]|
| 4250  | 35.5  | 1.5$^{+0.3}_{-0.7}$| > 4.3               | [19]|
| 100.6 | 8.054 | 1.2$^{+0.4}_{-0.3}$| > 1.2               | [20]|
| 337.5 | 46.21 | 2.05 ± 0.49       | 8.2$^{+13.3}_{-3.1}$| [21]|
| 255.8 | 97.8  | 2.29 ± 0.25       | > 15                | this work |

TABLE I: Measurements of $^{50}$V decays in the last 45 years.
FIG. 1: Decay scheme of $^{50}$V. Two excited states can be populated, one via electron capture to $^{50}$Ti under emission of a 1553.8 keV gamma and the beta decay into the first excited state of $^{50}$Cr resulting in a 783.3 keV gamma.

II. EXPERIMENTAL PROCEDURE

The measurement was performed using an ultra-low background Ge-detector (ULB detector) at the underground laboratory for dosimetry and spectrometry of the PTB (UDO) close to Braunschweig (Germany). UDO is located inside the Asse salt mine providing shielding with respect to the secondary cosmic radiation of about 1200 m of water equivalent. The p-type extended range ULB detector has an efficiency of 89\% and an energy resolution of 2.1 keV at 1.33 MeV. The mass of the Ge single crystal is 1.9 kg. It is surrounded by a shielding of low activity made of inner 10 cm of electrolytic copper and two outer layers of lead with a $^{210}$Pb content of 2 Bq/kg and 6 Bq/kg, respectively. For a detailed description of the apparatus see \[22\].

The detector was calibrated by recording the spectra of cans filled with solutions of well known activity provided by PTB. The activity of single nuclides within these solutions is known within an uncertainty between 1\% and 2\%. The solutions covered an energy range of strong emission lines from 21.0 keV to 1836 keV, using 11 radionuclides producing 18 major emission lines. To obtain the efficiency the sample-detector geometry was modeled by applying the Monte Carlo code GESPECOR (some geometrical parameters of the detector known from the technical drawing were altered slightly to achieve a better agreement between measurement and Monte Carlo simulation). Summing corrections were obtained by using the same code. The code served to calculate the efficiency of the detector concerning the $^{50}$V sample as well. An efficiency transfer was calculated from a 100 ml can containing a known radionuclide concentration to the same 100 ml can filled with vanadium powder (with a natural isotopic composition). This procedure led to an uncertainty of the efficiency of less than 3\% in the covered energy region. More information on this detector and its performance can be found in \[22\].

The uncertainties published in this article were calculated by adding all known uncertainty contributions quadratically to obtain the value of the total uncertainty. This procedure is in agreement with the Guide to the expression of uncertainty in measurement, GUM \[23\]. The uncertainties in this article are expressed as standard uncertainties (coverage factor $k = 1$).

III. EXPERIMENTAL DETAILS

The background of the applied detector in the region of the 1553.8 keV line, integrated from 1553 keV to 1555 keV, is lower than 0.025 counts per day. The pure background in the region of the 783.3 keV line is also rather low: Integrated from 782 keV to 784 keV, it is about 0.25 counts per day. But thorium and uranium impurities of the sample lead to a much higher background in the region of the 783.3 keV line (see below), while there are no natural lines near the 1553.8 keV line. As the measurement is completely dominated by contaminations within the sample, the intrinsic background of the set-up can be neglected.

A sample of vanadium powder with a total mass of 255.82 g, corresponding to a volume of 100 ml, was filled in a cylindrical film can (made of plastic) and placed on top of the above mentioned detector. The total measuring time was 97.8 days resulting in a total detector mass times measuring time product of 185.8 kg d. The measurement confirmed that the sample was not highly purified because various emission lines from the natural decay chains of uranium and thorium are visible, caused by activities in the mBq range.

Vanadium is a base metal, hence the surface will oxidize when it has contact with air. In addition, it can absorb some water. The can in which the vanadium powder was kept was not completely hermetically sealed, so that it had contact with the surrounding air. The oxygen content of the sample of (6.13±0.12) % by mass was measured by the German Federal Institute for Materials Research and Testing (BAM) with a high precision. The water content of (1.445 ± 0.010) % by mass was determined by PTB by drying the sample at low temperatures (< 100° C). The mass of the oxygen and water was subtracted from the mass of the total sample before calculating the activity concentration and half-lives of vanadium.

IV. RESULTS

A. The 1553.8 keV line from electron capture

The energy range around the expected line from EC is shown in Fig. 2. The region shows two prominent lines, one at the expected energy and one at 1588 keV. The latter is emitted by $^{228}$Ac, a progeny of $^{232}$Th (nuclear data information taken from \[24\]). A fit to the first line results in 123 net counts. By taking into account the
FIG. 2: Energy spectrum in the range from 1500 keV to 1590 keV. The $^{50}$V EC decay line is marked in grey.

FIG. 3: Energy spectrum from 750 keV to 820 keV. The region, where the beta decay line of $^{50}$V is expected, is marked in grey.

detection efficiency of 2.17 % and the natural abundance of $^{50}$V of 0.25 % [24], this can be converted into a partial half-life of (2.29 ± 0.25) · 10$^{17}$ years. The fact that the 1554.1 keV line from $^{234m}$Pa (from the $^{238}$U series) could interfere with the line of interest has to be taken into account. This $^{234m}$Pa line is in equilibrium with a gamma line at 1001.1 keV, which should include 139 times more counts because of a higher emission probability and a higher efficiency of the detector at that energy. As the line at 1001.1 keV is not visible at all in the measured spectrum, the $^{234m}$Pa line contribution at 1554.1 keV is neglected.

All known sources of uncertainty bigger than 0.001 %, which have an influence on the calculated life-time, are listed in Table II. The total uncertainty of the half-life of the EC decay is clearly dominated by the uncertainty of the determination of the number of detected counts (i.e. fitting of the 1553.8 keV line and its background).

| Contribution                                      | Uncertainty in % |
|--------------------------------------------------|------------------|
| $^{50}$V abundance in sample (total)              | 2.5              |
| including:                                       |                  |
| Natural $^{50}$V isotopic abundance               | 1.6              |
| $V$ concentration due to manufacturer             | 0.1              |
| Water content of sample                           | 0.7              |
| Oxygen content of sample                          | 2.0              |
| Weight of sample                                  | 0.04             |
| Activity determination                            | 10               |
| including:                                       |                  |
| Number of detected counts                         | 10               |
| Detector efficiency                               | 3                |
| Total uncertainty of half-life                    | 11               |

TABLE II: Contributions to standard uncertainty assigned to the half-life of the EC decay of $^{50}$V. Contributions smaller than 0.001 % are not listed (especially the atomic weight of vanadium as well as the measuring time are known with a much higher precision).

B. The 783.3 keV line from beta decay

Many more lines are visible in the range of the potential $\gamma$-line from the beta decay into the first excited state of $^{50}$Cr (Fig. 3). At around 783 keV there are six known lines from the U and Th chains: 782.1 keV ($^{228}$Ac), 783.4 keV ($^{234m}$Pa), 785.4 keV ($^{212}$Bi), 786.0 keV ($^{214}$Pb), 786.1 keV ($^{214}$Bi) and 786.3 keV ($^{234m}$Pa) [25]. The contributions of the $^{234m}$Pa lines can be neglected for similar reasons that were explained in the section before. But in direct proximity to the energy of interest, 783.3 keV, the contributions of the remaining lines form peak-like structures. A line at 783.3 keV is not visible. On the contrary, a dip is found at that energy. An upper detection limit of 27 events (using a significance level of $\alpha = 5$ %) for a potential line can be calculated according to the ISO 11929 standard [26], a well defined standard procedure for sample measurements. With a detection efficiency of 3.35 % this converts into a lower limit on the partial half-life of the beta decay 1.5 · 10$^{18}$ years. This is larger than the claimed observation in [21]. If the partial half-life published in [21] had been correct, a total of 39 events should have been observed in our measurement, which is excluded by two standard deviations, if the large upper error of 160 % stated in [21] is not taken into account. The lower limit of the half-life of the beta decay published in this article shows that the value of 8.2 · 10$^{17}$ years entering current nuclear tables (like [25]) is too small, as the new value of the lower limit is nearly twice as high.

The uncertainty budget of our evaluation of the 783.3 keV line is very similar to that of our evaluation of the 1553.8 keV line, but here an uncertainty resulting
from the determination of the number of detected counts is not applicable. As the evaluation of the region of the 783.3 keV line only results in a lower limit, this value is not combined with an uncertainty.

V. SUMMARY AND CONCLUSIONS

More than two decades since the last attempt, the 4-fold forbidden decays of $^{50}$V were investigated again with an ultra-low background Ge-detector system located deep underground. For the first time ever oxygen and water content of the vanadium sample were determined to get a more accurate value for the real vanadium mass. The electron capture decay branch, which populates the first excited state of $^{50}$Ti, is clearly visible with a half-life of

$$T_{1/2}^{(50\text{V} \rightarrow 50\text{Ti}+1553.8 \text{keV})} = (2.29\pm0.25)\cdot10^{17} \text{a} \quad (1)$$

This is roughly a factor of two more precise compared to the best claim\textsuperscript{[21]}. The beta decay branch, which leads to the first excited state of $^{50}$Cr, could not be observed and a lower half-life limit of

$$T_{1/2}^{(50\text{V} \rightarrow 50\text{Cr}+783.3 \text{keV})} > 1.5\cdot10^{18} \text{a} \quad (2)$$

is concluded (with a significance level of 5 %). This is barely in agreement with the only positive claim made of this decay in the past. From both results of this article a combined total half-life of $^{50}$V with a lower limit of $2.0\cdot10^{17}$ years can be derived. That means that 7.1 % of the decays happen by beta decay at maximum. As a consequence, the branching ratio of the two decay modes of $^{50}$V is not correctly implemented in\textsuperscript{[22]}.

The current search was limited by an intrinsic contamination of the vanadium sample and of the detector. The actual background spectrum of the detector itself is about a factor of 4 lower concerning the $^{238}$U decay chains and a factor of 2 better concerning the $^{232}$Th decay chain. Thus, after purifying the vanadium, a follow-up measurement allows a more sensitive search for the beta decay mode of $^{50}$V.

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