Peer Review File

Manuscript Title: Observation of the radiative decay of the ^{229}Th nuclear clock isomer

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referees' comments:

Referee #1 (Remarks to the Author):

229Th is a unique nucleus which has an excited isomeric state with a very small energy separation from the ground state. The isomer energy was measured recently using conversion electron spectroscopy and a micro-calorimeter with the uncertainty of about 0.2 eV. Until now, the radiative decay of the isomeric state was not directly observed. Although physicists have been working towards breakthrough transition energy measurements, the recently measured energy values make direct laser excitation of the isomer quite challenging due to a large uncertainty. Therefore, a significant decreasing of the isomer energy uncertainty is the priority task for a development of nuclear optical clocks.
The manuscript by S. Kraemer et al reports first direct observation of radiative decay photons of the Th-229 isomer. The isomer energy is measured with the uncertainty of 0.024 eV which is seven times lower than recent numbers published by Nature and PRL in 2019-2020. The observation of the radiative decay in large-bandgap crystals and the half-life measurements of 229mTh demonstrate the possibility to create solid-state nuclear clock based on CaF2 and MgF2 crystals. The result is very important because of some discussions that the nuclear transition lifetime is significantly quenched in a solid-state environment.
This excellent experimental result is important to researchers in atomic, nuclear, and particle physics and opens a pathway for further direct laser excitation of the isomeric state in thorium. Obviously, this is a great step toward a development of nuclear clocks and subsequent tests of fundamental physics. I expect this work to generate broad interest in the physics community.
The manuscript is well written, covering relevant results in a clear way. Statistic and systematic effects of the measurements are clearly described. However, in my opinion, the authors too often cite review articles instead of original research papers.
Therefore, I recommend publication of this article by Nature after the authors consider the comments below. In principle, most comments are optional, but I assume a few minor changes will improve the perceptions of the article by a broad audience.

Specific comments:
1. In the introduction [Lines 039-041] of the article the authors mention the fundamental physics tests citing the references [3] and [4]. I assume, it is correct to mention the original papers, for example, of V. Flambaum and coauthors and but not only the review articles. Just as an example I give a reference to the following article: V.V. Flambaum, Enhanced effect of temporal variation of the fine structure constant and strong interaction in 229Th. Phys. Rev. Lett. 97, 092502 (2006).
2. [Line 097] I propose to mention a few articles where the ideas were established (not only reviews 2 and 11).

3. [Line 104] Can the surface quality influence crystal defects? Was any surface treatment provided prior to implantation?
4. I would consider important to explain more about the behavior of crystal defects and, if possible, to add a few words about a time evolution of the 183 nm. Personally, I do not see any reasons why the peak at 148 nm does not belong to the isomer decay. Anyway, I assume for a wide audience of readers it will be important to emphasize why the 148 nm line is not attributed to crystal defects appearing in both MgF2 and CaF2 substrates due to differences in the decay chains of mass number A=229 ions in comparison with A=230.
5. In the Methods section the calibration and the systematic uncertainty analysis is well described. Nevertheless, it looks like in Fig.1 (with the large 3 mm slit) the left wing of the A=229 183 nm line is significantly shifted to the shorter wavelength (blue line) in comparison with the A=230 wing. Also, looking at the blue lines, the center of the 183 nm line for A=229 crystal defect in CaF2 seems to be shifted by approximately 1 nm to the shorter wavelength relatively to the A=230 peak. Is it just a visual effect due to log scale, or is it an indication of a systematic shift of the spectrometer?
6. The lattice analysis is well described in the manuscript for CaF2 crystals. The charge compensated configurations which are predicted to suppress conversion-electron decay are mentioned. But the lifetime measurements of the thorium isomer are performed in MgF2 [Line 231]. The result is accompanied by the statement that it should be considered as a lower limit because of the potential presence of non-radiative decay channels. I am not a solid-state physicist and from my point of view it would be quite useful to add some words how to transfer the properties of thorium embedded in CaF2 to MgF2 lattice as a conclusion of the Characterization of the Lattice Location chapter.
7. [Line 185] The systematic uncertainty in the wavelength of 0.41 nm is called "conservative". What does it mean? Does it correspond to 1-sigma or is it an expanded uncertainty?
8. Is the half-life of the isomer taken into account for the fit in Fig.3?
9. The left panel in Fig.4 shows the results for the slit sizes of 0.25, 0.5, and 1 mm. For the final energy value shown on the right panel only the data points with the slit sizes ≤ 0.5 mm are used. Is it somehow important to show the results obtained with 1 mm slit on the right panel? If the authors provided more measurement with large slits sizes, it will be interesting for readers to see all data points in a separated figure.
10. The right panel in Fig.4 doesn’t contain the result obtained in 2019 by Yamaguchi et al. Perhaps, the authors didn’t include the result because of a large uncertainty of the measurements. Anyway, it would be correct to give the value at least in the figure caption.

Referee #2 (Remarks to the Author):

Referee report
Nature manuscript 2022-08-13403A

Observation of the radiative decay of the 229Th nuclear clock isomer
S. Kraemer et al

The manuscript by Kraemer and colleagues, submitted to Nature, presents a milestone achievement

in what can arguably be considered one of the most interesting isomers throughout the nuclear landscape. The low-lying isomeric state in 229Th, originally inferred through gamma-ray spectroscopy measurements in the mid-1970's, gained renewed interest in the early 2000s when the concept of exploiting the isomer as a nuclear clock was first proposed (Ref. 1 of the submitted article). It would take another decade to achieve the first direct evidence of the existence of the isomer through internal conversion studies (Ref. 6). That work provided a solid platform for a variety of subsequent experiments to better characterize the isomer (e.g., via laser spectroscopy, Ref. 7) as well as refinements in the energy using different techniques, published in Nature and other high-impact journals (Refs. 9, 10). The current work uses a very different approach to produce the isomer and provides several outstanding results that will most certainly impact the search for direct laser excitation of the nuclear state in the future.

The key results presented are as follows: for the first time, the radiative decay has been detected. This detection has been achieved following implantation of 229mTh into large-bandgap solid-state crystals, transparent to VUV photons. This result thus critically validates one of two approaches currently being pursued towards the development of a nuclear clock. The extracted energy of the isomeric state agrees with two earlier measurements (Refs. 9, 10), however improves the uncertainty by a factor of 7. Lastly, the half-life of the isomeric state, embedded in the host crystal, has been determined. Given the recent history of 229mTh (reviewed in several of the references), it is clear this isomer has attracted the attention of different communities. The nuclear clock itself has been proposed for tests of the variation of fundamental constants. Solid-state methods are applied as well as ion trapping, synchrotron facilities have been used for state population, X-ray microcalorimetry to determine the energy and so on. All these techniques and potential applications illustrate the cross-disciplinary interest in the isomer. Given these factors and the combination of the key results presented here, it is my view that this work merits publication in Nature. In the following, I will assess aspects of the work in more detail and present some final suggestions for the authors to consider.

Traditionally, 229mTh has been populated via the alpha decay of 233U. Kraemer and colleagues present a different approach that has several advantages, namely via the beta decay of the parent isotope, 229Ac, which itself is populated through a chain of beta-decaying isotopes produced at the ISOLDE radioactive ion beam facility. The feeding probability into the isomer can be estimated from previous studies (Ref. 21) to be at least a factor of 7 higher than the 2% branch following the alpha decay of 233U. The radioactive beams are implanted into VUV transparent crystals (MgF2 and CaF2). The bandgaps of these crystals are larger than the isomeric state energy, preventing internal conversion and thus facilitating observation of the radiative decay mechanism. As the beta decay process produces an almost recoil-free production, this critically minimizes potential lattice damage that would reduce the bandgap, a probable reason for the non-observation of the decay in previous attempts using 233U in solid-state hosts. Detection of the radiative decay involves a VUV spectroscopy setup, with implantation of beams into different crystals mounted on a sample holder wheel that can be rotated to the detection position in front of the spectrometer. Photons are collected and focused onto a suitably sensitive PMT detector. The wavelength is scanned by rotating a grating in the spectrometer. A separate experiment performing emission channeling measurements using 231Th as a proxy to 229Th to investigate the thorium incorporation on the lattice of CaF2 has been made. This aspect of the work is outside my expertise and thus I am unable

to comment in detail on those results (Fig. 5), however the data and discussion regarding the interpretation of the observed VUV photons does indicate that the large bandgap has been preserved and thus the conversion-electron decay mechanism has been suppressed.

Validation of the detection of the radiative photons is arguably the most important aspect in this approach. Figures 1 through 3 present selected spectra from different crystals as well as crystal thicknesses. The wavelength range of immediate interest - given the existing knowledge of the energy of the isomer - is highlighted (~150 nm), and a scan range up to about 190 nm is given. Clear features are seen, with a photon peak in the region of interest in all crystals. To definitively assign the peak at 148.7 nm to the radiative decay of the isomer, the authors change the mass of the implanted radioactive beam to A=230. Given similar beam intensities and expected radioluminescence background (from the beta decay), there is a notable absence of a peak (Fig. 1) using the A=230 beam. Table 1 of the Methods gives corresponding nuclear decay information that has been used to clearly identify the isotopes implanted (I would have liked to have seen a figure in the Methods of a gamma-ray spectra to strengthen this aspect). The authors discuss the different components that give rise to the wavelength spectra (detector dark count rate, radiative decay, continuous Cherenkov background) and the resulting fitting procedure applied in the three figures. By thoroughly understanding the contribution of the beta-decays in the decay chain to the Cherenkov radiation and corresponding half-lives, the time evolution of the 148.7-nm peak is studied (Fig. 2), from which a half-life of the isomer in the crystal host can be determined. The quality of the data and presentation in the different figures indicates the validity of this approach, as well as the rigorous analysis to interpret the features observed. In my view, observation of the radiative photons is thus convincingly made.

The data analysis procedure is appropriate, with a more extensive discussion of the systematic uncertainties arising from the VUV spectrometer presented, namely the reproducibility of the grating position, the position of calibration light sources and the distribution of light emission in the crystal due to the uncertainty of the profile of the implanted radioactive beam. A total (conservative) systematic uncertainty for spectrometer slit settings <0.5 mm is given (0.41 nm), dominating the uncertainty on the energy of the isomeric state (0.0023 eV), a factor of 7 improvement over previous measurements. This is clearly seen in the summary of the results compared to recent literature in Fig. 4.

In summary, the novel production method and data obtained in this work are convincing. The abstract, figures and supporting tables as well as the conclusions are fully appropriate. The use of referencing to earlier work is also applied correctly. I do have some suggested improvements and brief questions I would like the authors to consider in the following, nevertheless, I will repeat my earlier statement from above, that it is my view that the key results presented here represent a milestone in a long history of work towards the development of a nuclear clock using 229mTh, and that this work merits publication in Nature.

Suggested improvements:
1. I was a little surprised to see no form of schematic overview of the critical parts of the setup. This might have been due to limited space however I would encourage the authors to consider adding such a figure. Should that take up too much space, I wonder whether figure 5 could be added to the

methods section? Such a figure could for example include the VUV spectrometer (crystal position, calibration light source etc); the implantation foils and wheel; location of the detectors used to measure the relative intensities and composition of the implanted beam.
2. Less importantly to a schematic figure, would be an example gamma-ray spectra e.g., for A=229 and A=230 that could support the extended data table 1. It is my understanding that this validates the composition of the implanted beam and thus directly supports the discussion of changing the beam from A=229 to A=230 to compare the resulting VUV spectra. I think this may provide a useful visual link to the reference A=230 implantation. Again, this could be added to the Methods if space allows.
3. The reduction in the uncertainty of the energy of the isomeric state by a factor of 7 compared to previous work is one of the key results. In the abstract the authors allude to the consequences for the direct laser excitation searches. Would the authors consider giving some context to this statement? For example, the gain in search time should a VUV laser of say 1 MHz linewidth be used would be considerably reduced compared to the previous uncertainty.
4. Can the authors comment on whether they understand the origin of the peak at 183 nm and why it is only seen in CaF2? Is this likely to be associated with an impurity affecting only one type of crystal?
5. Can you clarify whether the beam intensity was consistent (within expected variations) between the two thick crystals of MgF2 and CaF2? I noticed that the data in Fig. 1 indicate a higher count rate for CaF2 in the A=229 case, whereas in the A=230 case the rate was higher in the MgF2 crystal. Please comment.
6. In connection with Fig. 4 left panel, it was a little difficult to see what effect, if any, the slit size has on the uncertainty of the 148.7-nm peak centroid. I understand that a reduced slit size should lead to better resolution yet with a compromise in the overall detection efficiency of photons. In the Methods section I noted that 2.5 nm and 5.5 nm are observed for a 0.5 mm and 3 mm entrance slit, respectively. Fig. 4 does not include data above a slit size of 1 mm, so I presume the increase resolution sensitivity as one reduces the slit size below 1 mm is not as clear.

Referee #3 (Remarks to the Author):

The manuscript by Sandro Kraemer and co-authors reports on the first observation of the radiative decay of the 8 eV 229-Th isomer. Due to the potential application of a nuclear clock, this unique isomer has received increased attention in the past decade. The radiative excitation and decay of the isomer is a crucial step towards the nuclear clock. As such, the efforts of Kraemer and co-workers are highly welcome and the results deserve attention by the community. The experiment that they report on is interesting, very complicated and provides an alternative approach to routes investigated so far. In particular, the production of the isomer does not occur via alpha decay of 233-U, but via beta decay of 229-Ac. This appears to be a key ingredient for the successful observation of the radiative decay of the isomer, which had remained so far elusive.

The manuscript reports on the observation of the VUV photons from the isomer decay and pins down the photon energy. In addition, a radiative lifetime of the isomer is reported. Of the two, I find the second piece of information more critical, because the correct extraction of the radiative lifetime from the observed counts relies on many assumptions and values which might not be completely

accurate. I find the obtained isomer energy value more reliable than the lifetime value.

The manuscript is well written and provides a large amount of pertinent information on the performed experiment. However, there are a number of questions that still require some additional discussion, along the following lines:

1) ion beam production and implantation

If I understand correctly, the implanted beam is a mixed beam of A=229. The 30 keV beam energy is per amu or per nucleus? Are any other nuclear processes possible in the implantation process, such as Coulex, or inelastic scattering? How does the 20% uncertainty of the production rate reflect in the evaluation of the transition energy, and in particular of the radiative lifetime of the isomer? And why is it that the production rates for the 50 nm CaF2 crystal were 3 times lower than the ones reported in the table?

2) the population of the isomeric state

It is not clear to me how much of the beta decay chain end up in the isomer state of 229-Th. The authors write that the total feeding probability of the isomer is increased by a factor 7 to 47 compared to the 233-U alpha decay. This is quite some span. In Ref. [21] I could find that the branching ratio I am interested in is more than 14%, giving a signal to background ratio better by a factor of 7 compared to 233-U. Where does the factor 47 come from?

3) the CaF2 crystal

The 183 nm peak is observed only in the thicker CaF2 crystal. Do the authors have any idea why it does not appear in the thinner sample?

Why is it that for the characterization of the lattice location, only a single occupied site is considered? This is what I get from the main manuscript text. I would assume that in the implantation process the final location of the Th ion is random, and several sites are possible. This is at least what is assumed to happen upon crystal doping, and implantation would seem to me more random a process than crystal growth. The fits in Fig. 5 look good (except for the comparison between b and f), but this is not to say that using several sites would have not produced a fit of similar quality. How does the assumption of the single site affect the extracted energy and lifetime values?
The discussion in the text is for somewhat misleading, because later on in the Methods it is written that “each measured pattern is fitted with a linear combination of simulated sites”.

Finally, the lattice site is discussed for CaF2, but the lifetime measurement was using the MgF2 crystal.
I would like to know what is the reason for investigating the time evolution of the 148 nm photon peak intensity only in the MgF2 crystal.

4) extraction of the half-life

This is the part that I am most concerned about. From the methods I understand that the half-life of the isomer is obtained in a fit which includes also a scaling factor. In this scaling factor, we recover the fraction of the embedded 229-Th isomers which decay radiatively. This figure is itself difficult to pin down – the authors themselves admit that several parameters are needed for which only estimates are available. In particular, we have a factor of 7 between the reported 1 to 7%, which in turn relies on the total beta feeding probability. The scaling factor contains however once more the total feeding probability, which then affects twice the extraction. At the moment, I cannot judge from the manuscript how conservative the extracted value is and how well the error bar reflects our knowledge or lack of knowledge on the mechanisms of population and decay of the isomer.

Author Rebuttals to Initial Comments:

Referee 1, Comment 1

"*In the introduction [Lines 039-041] of the article the authors mention the fundamental physics tests citing the references [3] and [4]. I assume, it is correct to mention the original papers, for example, of V. Flambaum and coauthors and but not only the review articles. Just as an example I give a reference to the following article: V.V. Flambaum, Enhanced effect of temporal variation of the fine structure constant and strong interaction in ^{229}Th. Phys. Rev. Lett. 97, 092502 (2006).*"

The first paragraph has been changed due to space constraints and reads now as:

The radionuclide thorium-229 features an isomer with an exceptionally low excitation energy that enables direct laser manipulation of nuclear states. It constitutes a leading candidate for use in next-generation optical clocks [1–3]. This nuclear clock will be a unique tool for precise tests of fundamental physics [4–9]. While indirect experimental evidence for the existence of such an extraordinary nuclear state is significantly older [10], the proof of existence has been delivered only recently by observing the isomer's electron conversion decay [11]. The isomer's excitation energy, nuclear spin and electromagnetic moments, the electron conversion lifetime and a refined energy of the isomer have been measured [12–16]. In spite of recent progress, the isomer's radiative decay, a key ingredient for the development of a nuclear clock, remained unobserved. Here, we report the detection of the radiative decay of this low-energy isomer in thorium-229 (^{229m}Th). By performing vacuum-ultraviolet spectroscopy of ^{229m}Th incorporated into large-bandgap CaF_2 and MgF_2 crystals at the ISOLDE facility at CERN, photons of 8.338(24) eV are measured, in agreement with recent measurements [14–16] and decreases the uncertainty by a factor of seven. The half-life of ^{229m}Th embedded in MgF_2 is determined to be 670(102) s. The observation of the radiative decay in a large-bandgap crystal has important consequences for the design of a future nuclear clock and the improved uncertainty of the energy eases the search for direct laser excitation of the atomic nucleus.

Doing this, we have changed the references in order to include more original research papers. The second sentence in the introductory paragraph introduces the nuclear clock and cites now:

[1] Peik, E. & Tamm, C. Nuclear laser spectroscopy of the 3.5 eV transition in Th-229. EPL 61 (2), 181 (2003)
[2] Campbell, C. J. et al. Single-ion nuclear clock for metrology at the ^{19}th decimal place. Phys. Rev. Lett. 108 (12), 120802 (2012)
[3] Beeks, K. et al. The thorium-229 low-energy isomer and the nuclear clock. Nat. Rev. Phys. 3 (4), 238–248 (2021)

The concept of the nuclear clock was originally proposed in [1] and, to our knowledge, the first thorough evaluation of the achievable accuracy, showcasing the full potential of the clock, can be found in [2]. We however believe that the nuclear clock is a fast evolving research topic and recent review papers help the novice reader to get a quick overview over the field and have therefore decided to keep the review papers in the references.

The third sentence discusses potential applications of the nuclear clock as a quantum sensor. The following references have been included:

[4] Flambaum, V. V. Enhanced Effect of Temporal Variation of the Fine Structure Constant and the Strong Interaction in ^{229}Th. Physical Review Letters 97 (9), 092502 (2006)
[5] Peik, E. et al. Nuclear clocks for testing fundamental physics. Quantum Sci. Technol.6 (3), 034002 (2021)
[6] Uzan, J.-P. Varying Constants, Gravitation and Cosmology. Living Reviews in Relativity 14 (1), 2 (2011)
[7] Derevianko, A. & Pospelov, M. Hunting for topological dark matter with atomic clocks. Nature Physics 10 (12), 933–936 (2014)
[8] Arvanitaki, A., Huang, J. & Van Tilburg, K. Searching for dilaton dark matter with atomic clocks. Physical Review D 91 (1), 015015 (2015)
[9] Thirolf, P. G., Seiferle, B. & von der Wense, L. Improving our knowledge on the ^{229m}Thorium isomer: Toward a test bench for time variations of fundamental constants. Ann. Phys. 531 (5), 1800381 (2019)

Reference [4] can indeed be considered as one of the original papers on the idea of a measurement of variations of the fine structure constant with the clock, complemented with [6]. Reference [5] and [7] describe the idea of the use of clocks to detect ultralight dark matter. These two aspects make the clock a well-suited tool for fundamental physics tests and distinguishes the concept from other very stable atomic optical clocks. Reference [8] describes the use of atomic clocks to search for topological dark matter. Reference [9] is the review paper already referred to in the manuscript that we keep for above stated reasons.

Referee 1, Comment 2

[Line 097] I propose to mention a few articles where the ideas were established (not only reviews 2 and 11).

Both ideas, the trap and the solid state approach have been established in the original paper of on the nuclear clock, reference [1]. However, significant experimental progress has been made since then. The review article references have been extended by the following articles to reflect in a non-exhaustive way the developments on the two methods:

[5] Peik, E. et al. Nuclear clocks for testing fundamental physics. Quantum Sci. Technol. 6 (3), 034002 (2021)
[22] Campbell, C. J., Radnaev, A. G. & Kuzmich, A. Wigner Crystals of for Optical Excitation of the Nuclear ^{229}Th Isomer. Physical Review Letters 106 (22), 223001 (2011)
[23] Rellergert, W. G. et al. Constraining the Evolution of the Fundamental Constants with a Solid-State Optical Frequency Reference Based on the ^{229}Th Nucleus. Physical Review Letters 104 (20), 200802 (2010) .
[24] Kazakov, G. A. et al. Performance of a ^{229}Thorium solid-state nuclear clock. New Journal of Physics 14 (8), 083019 (2012)

References [22] and [5] discuss the trap-based approach followed at the University of Michigan and a setup under development at the University of Munich within the Thoriumclock consortium. The solid-state approach is followed at the University of California Los Angeles (reference [23]) and within the Thoriumclock consortium (references [5] and reference [24]).

Referee 1, Comment 3

[Line 104] Can the surface quality influence crystal defects? Was any surface treatment provided prior to implantation?

The channeling measurements provide information on the crystal quality. In CaF_2, the emission channeling results presented in the manuscript, revealing a large substitutional fraction and a strong channeling anisotropy (Figure 5), are themselves evidence that the near-surface (implanted) region is highly crystalline. In addition, in preparation for the reported experiments, we performed Rutherford backscattering and channeling analysis of the CaF_2 bulk crystals and thin films, which confirmed the high crystal quality. In the meantime, we have also performed preliminary emission channeling measurements on MgF_2 crystals: similarly to CaF_2, the observed strong channeling anisotropy clearly indicates that the near-surface (implanted) region is highly crystalline. Although one can indeed expect that the surface quality and thus the amount of crystal defects may vary across different crystals, we do not see clear indications of such differences in our present channeling data.

All bulk crystals used are VUV grade single crystals, polished by the manufacturer. Prior to the VUV spectroscopy measurements, the crystal surface was wiped with isopropanol. Before the emission channeling measurements, the crystals were wiped with acetone, as a preemptive measure, to remove debris that could cause electron scattering. The 30 keV implantation is shallow, hence close to the surface indeed, but a corresponding 15 nm implanted depth guarantees an implantation beyond a hydrocarbon surface layer and into the CaF_2 or MgF_2 crystals. For clarification, we added information on the surface treatment performed prior to implantation in the Methods section – Large band-gap crystals. The new paragraph reads as:

The 5 mm MgF_2 and 5 mm, 0.7 mm and 0.5 mm CaF_2 are commercially available VUV grade crystals, polished by the manufacturer. Prior to the VUV spectroscopy measurements, the crystal surface was wiped with isopropanol, because our tests revealed that this enhanced their transmission, likely due to removal of surface contaminants introduced during polishing and handling. Before the emission channeling measurements, the crystals were wiped with acetone, as a preemptive measure, to remove debris that could cause electron scattering. The growth of the 50 nm CaF_2 thin film is described in the following paragraph. This thin film crystal is used as grown, without further surface treatment. Emission of VUV photons from ^{229m}Th is observed for all crystals.

In addition, information about the type of surface polishing performed by the manufacturers was added as an extra column to Extended Data Table 2.

Referee 1, Comment 4

I would consider important to explain more about the behavior of crystal defects and, if possible, to add a few words about a time evolution of the 183 nm. Personally, I do not see any reasons why the peak at 148 nm does not belong to the isomer decay. Anyway, I assume for a wide audience of readers it will be important to emphasize why the 148 nm line is not attributed to crystal defects appearing in both MgF_2 and CaF_2 substrates due to differences in the decay chains of mass number A=229 ions in comparison with A=230.

The instrument used in this work consists of a grating monochromator and a position insensitive detector and records only one wavelength at a time (see Extended Data figure 3). Unfortunately, during the on-line experimental campaign, monitoring specifically the time dependence of the 183 nm line was not possible because of time constraints (limited beam time). We can therefore not comment on the time behaviour of its intensity.

The similar masses and identical implantation energies allow us to assume that the A=229 and A=230 ion beams interact similarly with the host crystals during implantation and therefore create the same crystal defects along the path of implantation. The energies of the emitted electrons after beta decay are different but both decay chains produce electrons of high enough energy to displace atoms and can therefore be considered to be in similar energy regimes. It is thus reasonable to assume that both decay chains produce the same type of crystal defects and these lines should show up in the spectra of both masses.

We clarify the text by adding the following sentence at line 145:

Therefore, radiation-induced excitation of electronic modes associated with crystal defects [41] should be observed after implantation of either A=229 and 230 beams into the same crystal while the signal from the ^{229}Th radiative decay will only appear in the A=229 case.

We additionally clarify the text by changing line 108 from:

The VUV photon peak can be caused by the radiation-induced excitation of electronic modes associated with crystal defects [31]. The latter should...

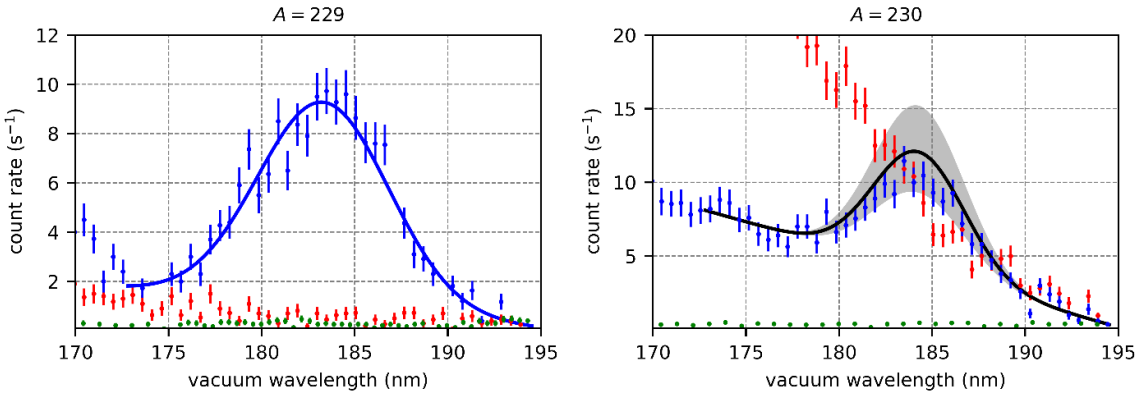
to

A VUV photon peak can be caused either by the ^{229m}Th radiative decay or by the radiation-induced excitation of electronic modes associated with crystal defects [41]. The latter should […].

Referee 1, Comment 5

In the Methods section the calibration and the systematic uncertainty analysis is well described. Nevertheless, it looks like in Fig.1 (with the large 3 mm slit) the left wing of the A=229 183 nm line is significantly shifted to the shorter wavelength (blue line) in comparison with the A=230 wing. Also, looking at the blue lines, the center of the 183 nm line for A=229 crystal defect in CaF_2 seems to be shifted by approximately 1 nm to the shorter wavelength relatively to the A=230 peak. Is it just a visual effect due to log scale, or is it an indication of a systematic shift of the spectrometer

The peak is found at the same position in both spectra. Plotting the same data on a linear scale and zooming into the 170 nm to 195 nm region shows the similarity between the peaks:



The conservative systematic uncertainty estimate for an entrance slit of 3 mm would result in approximately 2 nm. From the figure it becomes obvious that within this uncertainty, the two peaks agree in position.

Referee 1, Comment 6

The lattice analysis is well described in the manuscript for CaF_2 crystals. The charge compensated configurations which are predicted to suppress conversion-electron decay are mentioned. But the lifetime measurements of the thorium isomer are performed in MgF_2 [Line 231]. The result is accompanied by the statement that it should be considered as a lower limit because of the potential presence of non-radiative decay channels. I am not a solid-state physicist and from my point of view it would be quite useful to add some words how to transfer the properties of thorium embedded in CaF_2 to MgF_2 lattice as a conclusion of the Characterization of the Lattice Location chapter.

The reported isomer lifetime does not consider any combination of potential charge configuration mechanisms and is an experimental value deduced from the fit. This lifetime measurement procedure provides a lower limit of the pure radiative decay lifetime because the presence of charge compensation mechanisms and their effect on the electron conversion decay channel of the thorium isomer are unknown. It should be noted that any complementary decay channel next to the radiative decay would lower the measured half life value, as such the reported half life is a lower limit.

The characterization of the CaF_2 crystal indicates that the measured position of the thorium atoms agrees with the ground state configuration calculated with DFT and it should not favor the electron conversion channel. The lifetime measurement was, however, performed in the MgF_2 crystal because during the on-line campaign it became clear that the VUV photon rate was larger in MgF_2 compared to CaF_2. Unfortunately, due to lack of beam time we were not able to determine the lifetime in the CaF_2 crystal.

For MgF_2 we have no experimental results on the lattice position or present charge compensation mechanisms. Literature suggests that thorium occupies a magnesium substitutional position, and the lowest energy charge compensation mechanism is a magnesium vacancy, but disagrees on the effect this configuration has on the band gap and possible electron conversion. Whereas we would also expect to find substitutional thorium in a MgF_2 crystal it is not possible to comment on the impact on the electron conversion channel or lifetime without further theoretical and experimental work.

Referee 1, Comment 7

[Line 185] The systematic uncertainty in the wavelength of 0.41 nm is called "conservative". What does it mean? Does it correspond to 1-sigma or is it an expanded uncertainty?

Both, the exact positioning of the calibration source and the distribution of the light emitted in the crystal with respect to the optical axis of the system can systematically shift the observed peak in the wavelength spectrum. Under “conservative” we understand a mismatch between the optical axis of the calibration light source and the optical axis of the entrance of the spectrometer of up to 1 degree and misalignment of the light source implanted into the crystal

as far off as the slit height and width allow. Values quote a one-sigma confidence interval.

The following sentence in the Methods section has been changed from

The total systematic uncertainty for slit settings ≤0.5 mm, including the systematic effect of the wavelength calibration light source, the implantation beam profile and the grating drive reproducibility amounts to a conservative estimate to <0.41 nm.

to

The total systematic uncertainty for slit settings ≤0.5 mm (one sigma confidence level), including the systematic effect of the wavelength calibration light source, the implantation beam profile and the grating drive reproducibility amounts in a conservative estimate to <0.41 nm.

Referee 1, Comment 8

Is the half-life of the isomer taken into account for the fit in Fig.3?

The time behaviour of the signal and background is taken into account in the fit and follows the ^{229}Ac half life behaviour as the collection of the spectrum was started 32 minutes after the end of the implantation when the influence of the ^{229m}Th half life vanishes (see Fig. 2).

In order to clarify, the following sentence has been added to the caption of figure 3:

The scan was started 32 min after the end of the implantation.

Referee 1, Comment 9

The left panel in Fig.4 shows the results for the slit sizes of 0.25, 0.5, and 1 mm. For the final energy value shown on the right panel only the data points with the slit sizes ≤ 0.5 mm are used. Is it somehow important to show the results obtained with 1 mm slit on the right panel? If the authors provided more measurement with large slits sizes, it will be interesting for readers to see all data points in a separated figure.

All the available data with slit sizes ≤1 mm are shown in the left panel of figure 4. The agreement between the different crystals taking into account only the statistical uncertainties shows that no significant shift of the final wavelength value is expected from including the 1 mm entrance slit data (two data points in CaF_2 50 nm). The overall systematic uncertainty would however be significantly increased.

The data with a broader slit of 3 mm in figure 1 left panel results in peak centroids at 148.81(11)stat. nm (blue), 148.52(14)stat. nm (red) and 148.18(13)stat. nm (green). The disagreement with the values in figure 4 left panel becomes clearly apparent for the green dataset, which is expected because of the strong asymmetry of the observed peak, hinting towards a systematic shift due to the beam implantation profile. A conservative estimate of the systematic uncertainty for a 3 mm entrance slit setting would result in an error of +/-2.1 nm to be attributed to each of the values, bringing the values in agreement with the reported result. We believe that showing these values in figure 4 does not provide additional evidence considering the large systematic uncertainty, as it would deviate the attention away from the good agreement at the small linewidth measurements.

Referee 1, Comment 10

The right panel in Fig.4 doesn't contain the result obtained in 2019 by Yamaguchi et al. Perhaps, the authors didn't include the result because of a large uncertainty of the measurements. Anyway, it would be correct to give the value at least in the figure caption.

The value of Yamaguchi et al. Is reported with an uncertainty of 0.92 eV, a significantly larger uncertainty than the measurements of Seiferle et al. and Sikorsky et al. With 0.17 eV each. We refer to this value in the introductory

paragraph, but plotting the value in the right panel of figure 4 would increase the required range of the ordinate axis such that the error bar of the result of the present work becomes barely visible. We therefore chose to only take the most precise values available in literature for comparison. The value of Yamaguchi et al. has been added to the comparative discussion in the main text in order to highlight with this important result. The discussion was changed from

Our value is in agreement within the one σ confidence interval from conversion electron spectroscopy reported in [12] and two σ confidence interval of the recent microcalorimetric measurement [13], and decreases the uncertainty of the isomer's excitation energy by more than a factor of seven.

to

Our value is in agreement within the one σ confidence interval from conversion electron spectroscopy reported in [14] and one σ and two σ confidence intervals of the recent microcalorimetric measurements in [15] and [16] respectively. It decreases the uncertainty of the isomer's excitation energy by more than a factor of seven compared to each of the two most precise studies available in literature [14, 16].

The caption of figure 4 was adapted to include the following sentence to underline the choice of presented literature comparisons:

The right panel compares the energy value deduced from the scans with slits ≤0.5 mm in all three crystals, including the statistical and systematic uncertainties, to results of the two most precise recent studies [14, 16]

Referee 2, Comment 1

I was a little surprised to see no form of schematic overview of the critical parts of the setup. This might have been due to limited space however I would encourage the authors to consider adding such a figure. Should that take up too much space, I wonder whether figure 5 could be added to the methods section? Such a figure could for example include the VUV spectrometer (crystal position, calibration light source etc); the implantation foils and wheel; location of the detectors used to measure the relative intensities and composition of the implanted beam.

A figure containing a technical drawing of the instrument used with the different components has been added as Extended Data figure 3 and is referenced in the first sentence of the Vacuum-Ultraviolet Spectroscopy subsection of the Methods section.

Referee 2, Comment 2

The reduction in the uncertainty of the energy of the isomeric state by a factor of 7 compared to previous work is one of the key results. In the abstract the authors allude to the consequences for the direct laser excitation searches. Would the authors consider giving some context to this statement? For example, the gain in search time should a VUV laser of say 1 MHz linewidth be used would be considerably reduced compared to the previous uncertainty.

The reduction in the uncertainty of the energy would result in a gain of a factor of seven in measurement time, however a more quantitative statement depends on the methodology used in these laser probing studies (see e.g. von der Wense et al., Eur. Phys. J. D (2020) 74: 146). Therefore, we would refrain from commenting on this in the paper.

Referee 2, Comment 3

Can the authors comment on whether they understand the origin of the peak at 183 nm and why it is only seen in CaF_2? Is this likely to be associated with an impurity affecting only one type of crystal?

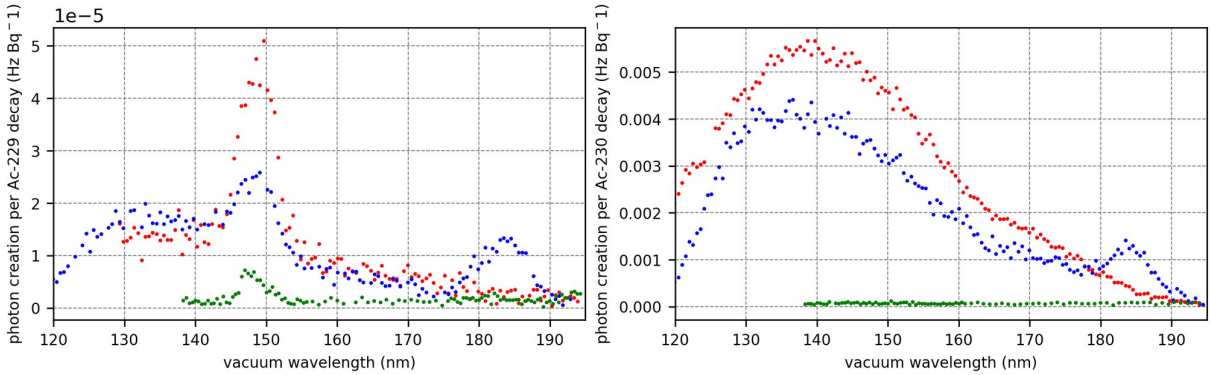
(See also reply to comment 4 of referee 1) We currently do not know which defect is responsible for the 183 nm line. Since it is visible for both implanted masses A=229 and A=230 in CaF_2, which behave similarly during implantation

and produce beta electrons in similar energy regimes, it can be assumed to be caused by a crystal defect. The data do not allow to distinguish between different types of defects or possible defect origins, e.g. whether the defect causing the 183 nm line was created during implantation, during beta-decay or was already present in the crystals at the surface or in the bulk. We can however safely presume that this 183 nm line does not have to be present in the MgF_2 crystals. Indeed, if the same type of defect was created (or present) in both crystals, for example an anion vacancy, these lines would show up at different wavelengths because of the differences in crystal structure, chemical composition and electronic structure. Therefore, the non-presence of the 183 nm line in MgF_2 is an extra argument that the 148 nm line is associated to a nuclear effect related to the implanted beam and thus to the thorium isomer.

Referee 2, Comment 4

Can you clarify whether the beam intensity was consistent (within expected variations) between the two thick crystals of MgF_2 and CaF_2? I noticed that the data in Fig. 1 indicate a higher count rate for CaF_2 in the A=229 case, whereas in the A=230 case the rate was higher in the MgF_2 crystal. Please comment.

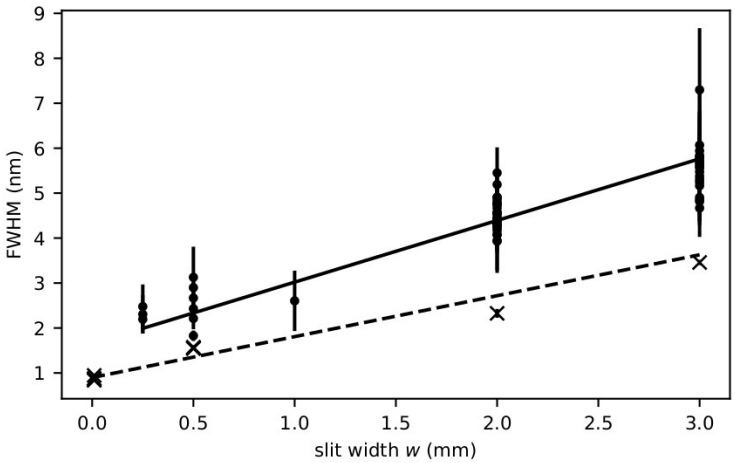
To answer this question, we created two spectra (see below) based on the data shown in Fig. 1. These spectra are normalized to the instantaneous activity calculated at the moment the data point was collected and based on the implanted source strength. It should be noted that the activity has a large systematic uncertainty and that the position and shape of the implanted beam influence the photon collection and detection efficiency. These data (red MgF_2 5 mm, blue CaF_2 5 mm, green CaF_2 50 nm) seem to indicate that the rate of VUV photons from ^{229m}Th from is larger for the MgF_2 crystal, however, as explained in the method section, the implantation profile of the radioactive beam was not fully under control and we refrain from drawing firm conclusions from these observations.



Referee 2, Comment 5

In connection with Fig. 4 left panel, it was a little difficult to see what effect, if any, the slit size has on the uncertainty of the 148.7-nm peak centroid. I understand that a reduced slit size should lead to better resolution yet with a compromise in the overall detection efficiency of photons. In the Methods section I noted that 2.5 nm and 5.5 nm are observed for a 0.5 mm and 3 mm entrance slit, respectively. Fig. 4 does not include data above a slit size of 1 mm, so I presume the increase resolution sensitivity as one reduces the slit size below 1 mm is not as clear.

The linewidth (FWHM) as a function of the entrance slit width looks as follows:



The linewidth of a isotropically emitting light source on a crystal at 3 mm from the entrance slit is represented by the dots (measurements) and the solid line (fit of the expected linear relation). The crosses and the dashed line are for a more collimated calibration light source and can be ignored in this context.

The uncertainty induced by the non-negligible linewidth is small compared to the influence of potential systematic shifts. The following table shows the contributions to the systematic uncertainty as a function of slit size:

	1 mm slit	0.5 mm slit	0.25 mm slit
Alignment of the calibration source	0.15 nm		
Position of implanted beam	0.67 nm	0.33 nm	0.17 nm
Grating drive	0.2 nm		

In order to limit the systematic uncertainty only data taken with slit sizes below 1 mm were considered in the final result.

Referee 3, comment 1

If I understand correctly, the implanted beam is a mixed beam of A=229. The 30 keV beam energy is per amu or per nucleus? Are any other nuclear processes possible in the implantation process, such as Coulex, or inelastic scattering? How does the 20% uncertainty of the production rate reflect in the evaluation of the transition energy, and in particular of the radiative lifetime of the isomer? And why is it that the production rates for the 50 nm CaF_2 crystal were 3 times lower than the ones reported in the table?

The 30 keV beam energy is the total kinetic energy, not the energy per amu. At these implantation energies, nuclear reactions and Coulomb excitation can be fully neglected. The 20% uncertainty of the production rate has no influence on the determination of the transition energy nor on the life time.

For energy determination, the fit includes the ^{229}Ac lifetime behaviour as the spectra are taken long after the end of the implantation (see also comment 8 of referee 1). For the lifetime determination, only the relative production rates between Fr and Ra are important and this uncertainty is included, while the product of the absolute activity and the unknown VUV detector efficiency is included as a free fit parameter (see also reply to comment 3.6).

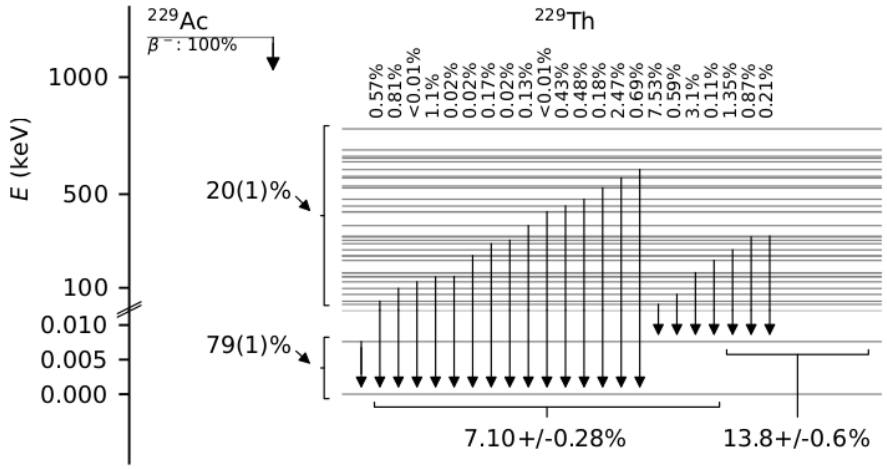
The experiment with the 50nm CaF_2 crystal was performed in a different beam time, using a different target-ion source radioactive beam system and isotope separator. The beam production was lower, which is a typical beam time to beam time variation. Moreover, the radioactive beam profile depends on the target-ion source perfomance and might differ from beam time period to beam time period. It could not be determined in this campaign.

Referee 3, Comment 2

It is not clear to me how much of the beta decay chain end up in the isomer state of 229-Th. The authors write that the total feeding probability of the isomer is increased by a factor 7 to 47 compared to the 233-U alpha decay. This is quite some span. In Ref. [21] I could find that the branching ratio I am interested in is more than 14%, giving a signal to background ratio better by a factor of 7 compared to 233-U. Where does the factor 47 come from?

Two components feeding the isomer state can be distinguished: The indirect feeding takes place via the feeding of higher energetic nuclear levels in the β-decay and a consecutive γ-cascade ending up in the isomer state. These value can be estimated from γ-spectroscopy data and are found in literature. The available data hint towards a feeding probability of 13.8(6)%, cited in Verlinde et al. The direct feeding in the β-decay, on the other hand, was, due to the small excitation energy of the isomer, not measurable in previous studies and only a value of 79(1)% for feeding both, the ground and the isomeric state was given. As we don't know this contribution and an estimate is difficult to obtain from the theory of β-decay, we are left with a branching ratio between 13.8% and 13.8% +79%=93%.

Comparing this value to the 2% branching in the decay of uranium-233 (where the assumption is made that the isomeric state direct feeding is negligible according to α-decay theory), a branching ratio higher by a factor between 7 and approx. 47 is obtained.



Referee 3, Comment 3

The 183 nm peak is observed only in the thicker CaF_2 crystal. Do the authors have any idea why it does not appear in the thinner sample?

The electrons from the radioactive decay lose much less energy in the thin crystals compared to the thick ones (as can be seen from the Cherenkov background). As such the intensity of the 183 nm line, if it stems from a bulk defect, will be strongly reduced. In addition, the type of defects and their concentration in thin films compared to thick crystals can be vastly different.

Referee 3, Comment 4

Why is it that for the characterization of the lattice location, only a single occupied site is considered? This is what I get from the main manuscript text. I would assume that in the implantation process the final location of the Th ion is random, and several sites are possible. This is at least what is assumed to happen upon crystal doping, and implantation would seem to me more random a process than crystal growth. The fits in Fig. 5 look good (except for the comparison between b and f), but this is not to say that using several sites would have not produced a fit of similar quality. How does the assumption of the single site affect the

extracted energy and lifetime values?
The discussion in the text is for somewhat misleading, because later on in the Methods it is written that "each measured pattern is fitted with a linear combination of simulated sites

We recognize that our description of the analysis procedure was perhaps insufficiently clear. We extended this part of the text in order to improve that:

In order to identify and quantify which lattice sites are occupied, measured and simulated emission patterns are numerically fitted [43]. For that purpose, simulated patterns are calculated for various lattice sites using the Manybeam code [34], including Ca and F substitutional sites, as well as various interstitial sites. Each measured pattern is then fitted with one as well as linear combinations of two or three of all the simulated sites, from where the respective occupation fractions are obtained. In the present case, if only one occupied site is considered, the best fit is obtained for thorium in a substitutional calcium site (Fig. 5). An occupation fraction of 77(4) % is obtained from the fit. If a linear combination of additional high-symmetry sites for 231Th is considered, the quality of the fit does not improve, indicating that eventual fractions of such sites are below the detection limit (typically of the order of 5 %).

In other words, we do consider various possible combinations of sites, as the reviewer correctly suggests as appropriate. In previous studies (see e.g. M.R Silva et al., A versatile apparatus for on-line emission channeling experiments. Rev. Sci. Instrum. 84 (7), 073506, 2013), combinations of multiple sites are indeed identified. However, in the present case (and, e.g. U. Wahl et al., Recent Emission Channeling Studies in Wide Band Gap Semiconductors, Hyperfine Interactions, 159, 363-372, 2005) only one site is found to be occupied, with eventual fractions in other sites being below the detection limit of our method (typically of the order of 5%).

Referee 3, Comment 5

Finally, the lattice site is discussed for CaF_2, but the lifetime measurement was using the MgF_2 crystal. I would like to know what is the reason for investigating the time evolution of the 148 nm photon peak intensity only in the MgF_2 crystal.

See reply to referee 1, comment 6.

Referee 3, Comment 6

This is the part that I am most concerned about. From the methods I understand that the half-life of the isomer is obtained in a fit which includes also a scaling factor. In this scaling factor, we recover the fraction of the embedded 229-Th isomers which decay radiatively. This figure is itself difficult to pin down – the authors themselves admit that several parameters are needed for which only estimates are available. In particular, we have a factor of 7 between the reported 1 to 7%, which in turn relies on the total beta feeding probability. The scaling factor contains however once more the total feeding probability, which then affects twice the extraction. At the moment, I cannot judge from the manuscript how conservative the extracted value is and how well the error bar reflects our knowledge or lack of knowledge on the mechanisms of population and decay of the isomer.

The time behavior of the implanted decay chain elements can be calculated from Bateman equations using the known implantation rates ***r*** obtained from γ-spectroscopy. The half-lives with exception of the half-life of the isomeric state are known from literature.

$$A_{\mathrm{Fr}}(t) = \frac{dN_{\mathrm{Fr}}}{dt} = r_{\mathrm{Imp}}^{\mathrm{Fr}}(t) - \frac{\ln(2)}{T_{\mathrm{T_1/2}}^{\mathrm{Fr}}} N_{\mathrm{Fr}}(t),$$

$$A_{\mathrm{Ra}}(t) = \frac{dN_{\mathrm{Ra}}}{dt} = r_{\mathrm{Imp}}^{\mathrm{Ra}}(t) + \frac{\ln(2)}{T_{\mathrm{T_1/2}}^{\mathrm{Fr}}} N_{\mathrm{Fr}}(t) - \frac{\ln(2)}{T_{\mathrm{T_1/2}}^{\mathrm{Ra}}} N_{\mathrm{Ra}}(t),$$

$$A_{\mathrm{Ac}}(t) = \frac{dN_{\mathrm{Ac}}}{dt} = r_{\mathrm{Imp}}^{\mathrm{Ac}}(t) + \frac{\ln(2)}{T_{\mathrm{T_1/2}}^{\mathrm{Ra}}} N_{\mathrm{Ra}}(t) - \frac{\ln(2)}{T_{\mathrm{T_1/2}}^{\mathrm{Ac}}} N_{\mathrm{Ac}}(t),$$

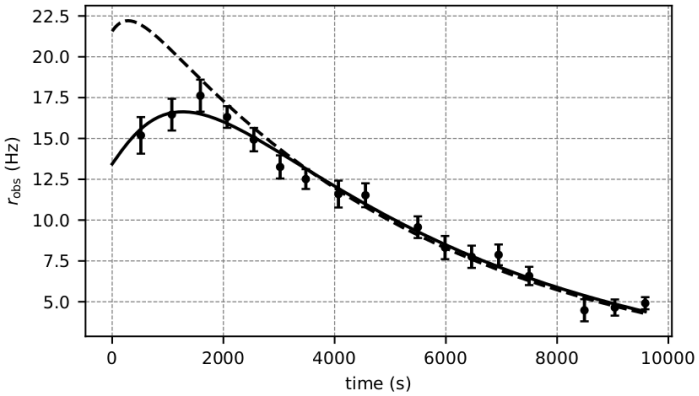
$$A_{\mathrm{iso}}(t) = \frac{dN_{\mathrm{iso}}}{dt} = + BR_{\mathrm{iso}} \frac{\ln(2)}{T_{\mathrm{T_1/2}}^{\mathrm{Ac}}} N_{\mathrm{iso}}(t) - \frac{\ln(2)}{T_{\mathrm{T_1/2}}^{\mathrm{iso}}} N_{\mathrm{iso}}(t),$$

The observed count rate is fitted with the following equation:

$$r_{\mathrm{obs}}(t) = \hat{p}_{\mathrm{photon}} A_{\mathrm{iso}}(t)$$

Here, the shape of the radioactive in-grow and the later exponential decay following the actinium behavior are contained in A_{iso} given by the system of equations above. The exact shape of the activity curve depends on the implantation time and the ratio of the ^{229}Fr to ^{229}Ra production rate (note that the ^{229}Ac production rate was determined to be negligible). The scaling parameter p_{photon} describes the product of all instrumental efficiencies, the branching into the isomer state and the scale of the beam production rate. This parameter scales the overall fitted curve but does not change its shape.

To show the sensitivity of the measurement to the half life of the isomer, we show below a fit to the data assuming the VUV photons stem from the beta decay of ^{229}Ac, which is represented by A_{Ac} (see dashed line). For the latter, only the data points from the 6th point onwards were taken into account.



Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

I assume, the revised version of the manuscript looks more understandable to readers. The authors provided additional explanations in the manuscript body and extended the methods section with additional details. Several original papers are also added to the references.

Nevertheless, I would like to make few suggestions which should further increase the value of the revised version.

1. Line [036] It is a quite strong statement to mention the thorium clock as a "leading candidate for use in next-generation optical clocks". There are some other candidates for the next generation of optical clocks (for example, highly charged ions). Therefore, I propose to smooth the sentence as follows: “the thorium nuclear clock is one of the leading candidates…”.
2. Lines [148-152] “The activities of the full decay chain ...are calculated using the half-lives of the first three isotopes know from the literature…”
Are the half-lives known from the literature for crystalline environments?

Referee #2 (Remarks to the Author):

It is my view that this revised version of the manuscript submitted by Kraemer and colleagues coherently addresses the variety of the comments and questions raised following the first round of reviewing. My original summary and overview of different aspects of the manuscript remains valid and I do not repeat that here. In response to my own suggested improvements to the original manuscript, the authors have made several additions, including two additional figures in the Extended Data section. A technical drawing of the VUV spectrometer with the relevant components (target wheel with the large-bandgap crystals, the position of the calibration light source as well as the direction of the radioactive beam) has been included, providing context to the discussion in the Methods section. A figure illustrating the gamma-ray singles spectra labeling the different activities associated with the mass of the implanted beam, both for A=229 and A=230 has been added. This I find to be a useful addition to Extended Data Table 1. The authors also provide a more detailed response to questions raised regarding the potential origin of the peak seen at 183 nm (raised also by referee 1) and use the line of questioning to strengthen the case for having identified the radiative photons from the decay of the isomeric state for the first time. The additional explanations in the rebuttal, with supporting figures, I found to be helpful in clarification of any (minor) queries I had.

I have also read with interest the responses to the questions and comments usefully raised by the other two referees. In my opinion, the authors have responded to the best of their ability, acknowledging that there are interesting aspects that would have required additional beam time to

address (for example a lifetime measurement in the CaF2 crystal). I would hope that such details could be investigated in future experiments now that the groundwork has been laid in this milestone experiment.

I have no further comments or suggestions for improvements of the revised version and am satisfied with the detailed responses provided. I recommend publication in Nature.

Referee #3 (Remarks to the Author):

The revised version of the manuscript has clarified most of my concerns. I am ready to recommend publication of the manuscript, with two small requests:
1) in reply to my comment 3.6, the authors explain how the scaling factor does not chance the overall shape of the fit. Around lines 613, it is explained that this scaling factor contains apart of the VUV detection efficiency also the fraction of embedded Th nuclei and the isomer feeding probability. However, at the beginning of the manuscript, line 155, only the VUV efficiency is mentioned. That part in the main text should be amended to mention also the other two factors. Especially the isomer feeding probability is not so well known quantity.
2) I could not find in the text any comments on the origin of the uncertainties given for the lifetime result.

Author Rebuttals to First Revision:

Referee 1, Comment 1

Line [036] It is a quite strong statement to mention the thorium clock as a "leading candidate for use in next-generation optical clocks". There are some other candidates for the next generation of optical clocks (for example, highly charged ions). Therefore, I propose to smooth the sentence as follows: "the thorium nuclear clock is one of the leading candidates…".

The second sentence has been changed as suggest and reads now as:

It constitutes one of the leading candidates for use in next-generation optical clocks [1–3].

Referee 1, Comment 2

Lines [148-152] "The activities of the full decay chain …are calculated using the half-lives of the first three isotopes know from the literature…"
Are the half-lives known from the literature for crystalline environments?

In view of the involved large energy scale of the β-decay (Q_β>>100keV, see Extended Data Table 1) and considering the experimental precision of lifetime measurements, an effect of the chemical environment is negligible within uncertainties.

The dependence of the half-life on the chemical environment has been the subject of studies, see for instance:

J. R. Goodwin, N. Nica, V. E. Iacob, A. Dibidad, and J. C. Hardy, Measurement of the half-life of 198Au in a nonmetal: High-precision measurement shows no host-material dependence. Phys. Rev. C 82, 044320 (2010)

J C Hardy 1, J R Goodwin, V V Golovko, V E Iacob, Tests of nuclear half-lives as a function of the host medium and temperature: refutation of recent claims. Appl Radiat Isot 2010 68 (7-8)

Referee 3, Comment 1

In reply to my comment 3.6, the authors explain how the scaling factor does not chance the overall shape of the fit. Around lines 613, it is explained that this scaling factor contains apart of the VUV detection efficiency also the fraction of embedded Th nuclei and the isomer feeding probability. However, at the beginning of the manuscript, line 155, only the VUV efficiency is mentioned. That part in the main text should be amended to mention also the other two factors. Especially the isomer feeding probability is not so well known quantity.

The corresponding sentence has been changed from

By scaling the calculated activity to take into account the total VUV spectometer efficiency, a half-life value of 670(102) s for the decay of 229mTh embedded in a MgF2 crystal is obtained.

to

By scaling the calculated activity to take into account the total VUV spectometer efficiency, the isomer feeding probability in the 229Ac β-decay and the fraction of radiative to total decaying isomers, a half-life value of 670(102) s for the decay of 229mTh embedded in a MgF2 crystal is obtained.

Referee 3, Comment 2

I could not find in the text any comments on the origin of the uncertainties given for the lifetime result.

The following pagagraph has been added in the Methods section:

The given uncertainty constitutes a conservative estimate and includes the following contributions: A statistical uncertainty of 79 s is obtained from the Markov-Chain-Monte-Carlo optimization. A variation of the ratio of implantation rates of ^{229}Fr to ^{229}Ra by an order of magnitude compared to the experimentally determined value from γ-data and a conservative uncertainty of 120 s on the period between the end of implantation and the start of the first measurement result in a 5 s and 53 s uncertainty, respectively. Systematic effects resulting from the

need to continuously scan the instrument for wavelength spectra recording and the resulting limited amount of data at the isomer's emission wavelength are estimated using a bootstrapping technique removing one data point at the time. The largest half-life deviation from the result with the full dataset, 35 s, has been added to this conservative uncertainty estimate.