

Frequency ratio of the $^{229\text{m}}\text{Th}$ isomeric transition and the ^{87}Sr atomic clock

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Version 0:

Reviewer comments:

Referee #1

(Remarks to the Author)

The manuscript by Zhang et al. presents the initial demonstration of nuclear excitation using a frequency comb. This work represents a historical achievement that will open a truly novel avenue for discovery in fundamental physics and a plethora of applications in various fields of science.

The excitation of nuclei by means of incoherent or partially coherent X-ray sources has a long tradition. Similarly, X-ray transitions observed through the Moessbauer effect have previously been employed in both fundamental (e.g., relativity and gravitation) and applied research studies. The present work also represents a significant contribution to the advancement of scientific knowledge on both aspects.

In contrast to the aforementioned earlier works, the authors employ a frequency comb that is capable of accessing the vacuum-ultraviolet (VUV) range for the excitation of the only known nuclear transition in this spectral domain: the isomeric lowest excited state of ^{229}Th at 8.3 eV. The decay of this state in systems that are protected from internal conversion is very slow, with a duration that depends on the surrounding medium, reaching several hundred seconds. Consequently, the transition is inherently narrow and well-suited for use as a frequency reference in the VUV range, a photon energy region that is significantly broader than the optical one and of paramount importance in atomic and molecular physics. As no other frequency reference has yet been demonstrated, this constitutes a sufficient reason for publication in Nature in itself.

Moreover, the 8.3-eV transition in ^{229}Th is of particular interest for investigating a possible time variation of fundamental constants, given that its sensitivity to a variation of the fine-structure constant α is more than one order of magnitude higher than that of other atomic systems that could be studied with frequency metrology. The delicate balance between Coulomb repulsion and nuclear binding forces gives rise to this exceptionally low isomeric energy. Any fluctuation or variation of this in the electron-photon sector or at the level of forces between baryons would result in a potentially measurable change in frequency.

The ^{229}Th nuclear clock, which has been demonstrated here, represents the most rapid approach to the discovery of an effect manifesting as a shift in frequency, which may arise from a variation in time or space of the fine-structure constant. Various models and theories have postulated such changes, but their discovery has remained beyond the reach of even the most accurate methods. What are the most accurate sensors currently available for such a search? Although gravitational wave detectors possess a higher differential sensitivity to strain than available optical clocks to an α variation, the former lack an absolute reference in contrast to the latter. Consequently, optical clocks are well placed to explore this important realm of physics beyond the standard model, and the nuclear clock, due to its extreme sensitivity, occupies the pole position in this race.

In light of these considerations, a number of research groups have devoted considerable resources to the development of this technology. The present manuscript represents a significant advance in the field, integrating a range of techniques that have overcome the limitations of previous approaches. It builds upon the expertise of the Vienna group in manufacturing a CaF_2 crystal with a high density doping of ^{229}Th , the preparation of the probe and the detection system for the slow photonic decay. This technique has also been crucial for the success of the very recent work at PTB, which use a pulsed laser for the excitation. The Colorado group has developed a VUV frequency comb with sufficient power and stability to excite the isomeric transition with one mode of the 7th harmonic on an infrared frequency comb. They have also developed methods to lock it to a Sr lattice clock for frequency metrology, to determine its absolute frequency in the VUV, including the generation of a VUV frequency comb, and the overall control of the experiment. Contributions to this work have been made by Garching and Ann Arbor collaborators. The pooling of their expertise has yielded a truly remarkable result, and the title "Dawn of a nuclear clock..." is not an exaggeration.

The manuscript presents a well-written and detailed account of the motivation, background, and description of the experimental methods, with additional references provided in the paper and methods section. This provides the reader with a comprehensive understanding of the experimental process. The authors establish a direct frequency link to the SR clock through the use of a frequency comb, demonstrating the generation and stabilization of high harmonics with an hour-long stable output. This illustrates the advancement of this new field in a clear and accessible manner. The experimental procedure, line centre and comb mode determination are explained in sufficient detail. It is evident that the authors have devoted considerable attention to the background suppression required for the measurement, which must have been a serious challenge. The observation of five transitions that can be assigned to the hyperfine splitting resulting from the nuclear quadrupole moment, in agreement with a substantial improvement in accuracy compared to earlier work on the same matter, provides compelling evidence to support the claims made in the paper. A precise lifetime measurement and its analysis in the context of the refractive index of the host crystal contribute another valuable piece of physics insight from this work. Graphics and figures convey all necessary information, and the discussion addresses the implications of this result. It is hoped that a further stabilisation of the primary frequency comb will soon allow for more accurate studies of the effects of crystal fields on the absolute frequency and of possible broadening mechanisms arising from them.

Although the ultimate accuracy of a nuclear clock may necessitate the implementation of the nuclear clock with single ions in precision traps, the potentially superior statistics that solid-state ^{229}Th probes embedded in crystals could provide could also be employed to stabilise a local oscillator in conjunction with advanced lattice clocks. This could also prove advantageous for other, more technical clock applications.

Summarizing, the fundamental operation of a nuclear clock based on the measurement of the VUV isomeric transition in ^{229}Th has been demonstrated through a frequency comparison with a reference optical clock in the infrared. This expands frequency metrology to nuclear transitions and provides a powerful method for investigating the time variation of the fine-structure constant with respect to the strong interaction force. The methods are original and valid, the data and their statistical quality convincing, and the presentation is of a high quality. The manuscript is well written, figures and graphics are very clear, and the list of references covers all prior work, including the context. The abstract, introduction and conclusions are perfectly adequate for a Nature publication. The paper should be published as soon as possible, and in interest of timeliness in its current form, as it represents a significant achievement with immediate implications for the broader scientific community and will likely prompt a surge of research by other groups.

Referee #2

(Remarks to the Author)

A.Summary of the key results

The manuscript reports on the laser excitation of the isomer transition of Thorium-229 nuclei, centered at 148.3 nm, via the radiation produced with a VUV comb.

After the first reported direct laser excitation of this low-energy nuclear transition at PTB [ref. 26], this timely article extends previous works by precisely determining the nuclear transition frequency against the frequency of a Strontium-87 optical clock. Furthermore, it reports on the direct observation and characterization of its nuclear quadrupole structure.

B.Originality and significance: if not novel, please include reference

This work represents the first absolute frequency determination of the isomer nuclear transition and opens the field of nuclear clocks. The measurement has been done through a direct comparison of the nuclear transition against an optical standard. This outstanding work opens the way to several fundamental physics studies. Specific experimental techniques are novel and are independent from previous results.

Indeed this result represents a high precision, timely, and independent confirmation of the first observation done only recently at PTB [ref 26].

C.Data & methodology: validity of approach, quality of data, quality of presentation

The article is well-written and clear. The quality of the presentation is very high in general, not only with informative designs of the experimental details and techniques but also, with detailed high-quality datasets, showing for the first time transition linewidth of only 300 kHz on a nuclear transition.

The data presented well support the results described in the manuscript, namely the nuclear-optical frequency ratio, the measure of the lifetime, and the determination of the nuclear quadrupole constants.

D.Appropriate use of statistics and treatment of uncertainties

Throughout the paper, an appropriate use of statistics and uncertainty treatment has followed.

One single minor remark.

While it is clear that on the frequency ratio, the uncertainty reported is only statistical, in the determination of the quadrupole constants and electric field gradient no comment is reported about the type of uncertainty. In this particular determination of constants, are expected systematic effects (maybe coming from the potential systematic shift of the nuclear transitions)?

Please clarify if the uncertainties also in this case are only statistical.

E.Conclusions: robustness, validity, reliability

No particular comment on the conclusions of the paper. The research is valid and robust. Furthermore, the results shown here represent a solid confirmation of previous observations.

F.Suggested improvements: experiments, data for possible revision

The authors might clarify how a single tooth of the comb interacts with thorium transitions, this might be useful, especially for researchers outside AMO physics.

Indeed, several comb components are simultaneously sent to the sample, and simultaneously, other transitions, shifted exactly by a harmonic of the repetition rate, could be directly excited.

I'd ask if the spectrum observed shows any change as a function of the comb repetition rate, or, in other words, if the author might, I'd ask if the comb number determination procedure (data reported in Fig.4) gives the same result for all the six resonances. This would completely exclude any possible role of nearby comb teeth in the excitation process of Thorium. Moreover, although it is clearly stated that the two additional peaks will be the object of future studies, I may ask if it is possible that these additional unexplained peaks could come from other comb components interacting with the Th-doped crystal.

G.References: appropriate credit to previous work?

No comments on references, apart from checking the latest status of pre-print articles, that might have been published meanwhile.

H.Clarity and context: lucidity of abstract/summary, appropriateness of abstract, introduction and conclusions

No further comments on this.

Author Rebuttal letter:

Dear Dr. Levi,

We greatly appreciate your kind effort in getting our manuscript promptly reviewed. We thank the two reviewers for their careful reading of the manuscript, deep understanding of the research subject, thoughtful comments for summarizing and clarifying our work, and extremely positive endorsement for publication in Nature. Below we provide responses to each of the three questions that Reviewer 2 raised. The review comments are typed in black, our response in blue, and additional text in the manuscript in red.

Referees' comments:

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The manuscript presents a well-written and detailed account of the motivation, background, and description of the experimental methods, with additional references provided in the paper and methods section. This provides the reader with a comprehensive understanding of the experimental process. The authors establish a direct frequency link to the SR clock through the use of a frequency comb, demonstrating the generation and stabilization of high harmonics with an hour-long stable output. This illustrates the advancement of this new field in a clear and accessible manner. The experimental procedure, line centre and comb mode determination are explained in sufficient detail. It is evident that the authors have devoted considerable attention to the background suppression required for the measurement, which must have been a serious challenge. The observation of five transitions that can be assigned to the hyperfine splitting resulting from the nuclear quadrupole moment, in agreement with a substantial improvement in accuracy compared to earlier work on the same matter, provides compelling evidence to support the claims made in the paper. A precise lifetime measurement and its analysis in the context of the refractive index of the host crystal contribute another valuable piece of physics insight from this work. Graphics and figures convey all necessary information, and the discussion addresses the implications of this result. It is hoped that a further stabilisation of the primary frequency comb will soon allow for more accurate studies of the effects of crystal fields on the absolute frequency and of possible broadening mechanisms arising from them.

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Summarizing, the fundamental operation of a nuclear clock based on the measurement of the VUV isomeric transition in ²²⁹Th has been demonstrated through a frequency comparison with a reference optical clock in the infrared. This expands frequency metrology to nuclear transitions and provides a powerful method for investigating the time variation of the fine-structure constant with respect to the strong interaction force. The methods are original and valid, the data and their statistical quality convincing, and the presentation is of a high quality. The manuscript is well written, figures and graphics are very clear, and the list of references covers all prior work, including the context. The abstract, introduction and conclusions are perfectly adequate for a Nature publication. The paper should be published as soon as possible, and in interest of timeliness in its current form, as it represents a significant achievement with immediate implications for the broader scientific community and will likely prompt a surge of research by other groups.

Response: We are grateful to the reviewer for writing this detailed and accurate summary of the results and for the extremely supportive comments. We also anticipate that this paper will be of wide interest for a broad scientific community and will prompt a surge of research in the new nuclear clock community.

Referee #2 (Remarks to the Author):

A. Summary of the key results

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The data presented well support the results described in the manuscript, namely the nuclear-optical frequency ratio, the measure of the lifetime, and the determination of the nuclear quadrupole constants.

Response: We also thank the reviewer for the summary and strongly positive comments.

D.Appropriate use of statistics and treatment of uncertainties

Throughout the paper, an appropriate use of statistics and uncertainty treatment has followed. One single minor remark.

While it is clear that on the frequency ratio, the uncertainty reported is only statistical, in the determination of the quadrupole constants and electric field gradient no comment is reported about the type of uncertainty. In this particular determination of constants, are expected systematic effects (maybe coming from the potential systematic shift of the nuclear transitions)? Please clarify if the uncertainties also in this case are only statistical.

Response: We thank the reviewer for asking us to clarify the uncertainty type for the quadrupole constants and the electric field gradient. For the first report of these constants, the uncertainty is also purely statistical, extracted from a Monte-Carlo simulation using the frequency uncertainties of the line-centres. In the next set of experiments, we plan to vary various parameters of the crystal, and upon completion of the future work, we should be able to comment on the systematic uncertainty associated with these measurements.

To clarify this, we added the following sentence to the manuscript:

Page 8, paragraph 2: "The extracted uncertainties are purely statistical."

Page 8, before "Discussion": Replaced "The extracted uncertainties are purely statistical" with "Systematic uncertainty of this frequency ratio will be explored in future studies."

E.Conclusions: robustness, validity, reliability

No particular comment on the conclusions of the paper. The research is valid and robust.

Furthermore, the results shown here represent a solid confirmation of previous observations.

F.Suggested improvements: experiments, data for possible revision

The authors might clarify how a single tooth of the comb interacts with thorium transitions, this might be useful, especially for researchers outside AMO physics.

Indeed, several comb components are simultaneously sent to the sample, and simultaneously, other transitions, shifted exactly by a harmonic of the repetition rate, could be directly excited. I'd ask if the spectrum observed shows any change as a function of the comb repetition rate, or, in other words, if the author might, I'd ask if the comb number determination procedure (data reported in Fig.4) gives the same result for all the six resonances. This would completely exclude any possible role of nearby comb teeth in the excitation process of Thorium.

Response: We thank the reviewer for this request to further clarify the comb spectroscopy technique. The ambiguity for the comb mode number arises from the lack of knowledge of the absolute frequency of a newly discovered resonance. This ambiguity can be fully resolved if we use a sufficient number of different valued repetition frequencies to over-constrain the mode number. For this reason, for each of the 4 strong peaks, we performed multiple scans (reported in Fig. 4) with different repetition rates to extract the absolute mode number in each measurement and thus its absolute transition frequency. As can be seen in Fig. 5, the reported peak frequencies differ from each other by more than 75 MHz (repetition rate). Thus, the comb mode numbers for exciting the different nuclear transitions are different but are all uniquely determined in this measurement procedure.

To be absolutely certain about the mode number determination, we repeated our measurements many times over a few weeks, using a diverse combination of different repetition rates (and thus, different integer mode numbers) and we indeed confirm that they are all consistent with the absolute frequency reported in the manuscript. The transition frequency (~2020.407 THz) and long lifetime (~641 s) are consistent with the recent PTB and UCLA result, confirming they are the nuclear transition instead of crystal artifacts.

To clarify this point, we added the following sentence:

Page 8 after Comb mode determination: "While we illuminate the crystal with all the comb modes simultaneously, each spectroscopic feature corresponds to a nuclear excitation by one single comb mode."

Moreover, although it is clearly stated that the two additional peaks will be the object of future studies, I may ask if it is possible that these additional unexplained peaks could come from other comb components interacting with the Th-doped crystal.

Response: We thank the reviewer for raising this question as well. We have performed careful systematic checks and confirm that the two additional small peaks are not consistent with any of the reported 5 peaks (Fig. 5) up to integer multiples of the comb repetition rates. We believe these two additional peaks belong to different electric field sites in the crystal. Indeed, DFT theory calculations point to the existence of these additional peaks. In our future work we will determine the origin of these peaks by varying a number of crystal parameters and even changing to a different host crystal.

G.References: appropriate credit to previous work?

No comments on references, apart from checking the latest status of pre-print articles, that might have been published meanwhile.

H.Clarity and context: lucidity of abstract/summary, appropriateness of abstract, introduction and conclusions

No further comments on this.

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