**Letter of Reply to the Editors and Referees.**

**We would like to thank the Editors and Referees for their careful reading of the manuscript, the many helpful comments and their positive decisions regarding the publication process.**

**In the following each point is shortly addressed directly in the comment letter:**

**Editorial comments:**  
Changes to be made by the Author(s):  
1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.

We have revised and proofread the manuscript.  
2. Please revise lines 44-50 to avoid previously published text; see attached iThenticate report for details.

The lines have been completely revised in order to avoid previously published text.  
3. Please obtain explicit copyright permission to reuse any figures from a previous publication. Explicit permission can be expressed in the form of a letter from the editor or a link to the editorial policy that allows re-prints. Please upload this information as a .doc or .docx file to your Editorial Manager account. The Figure must be cited appropriately in the Figure Legend, i.e. “This figure has been modified from [citation].”

All copyright permission has been obtained and is uploaded. Figures are cited accordingly.  
4. Please rephrase the Summary to clearly describe the protocol and its applications in complete sentences between 10-50 words: “Here, we present a protocol to …”

The summary has been rephrased.  
5. Please remove the headers in the Introduction.

The headers have been removed.  
6. Please define all abbreviations before use.

Abbreviations have been defined where required.  
7. Please include a space between all numbers and their corresponding units: 15 mL, 37 °C, 60 s; etc.

The spaces have been introduced.  
8. Please adjust the numbering of the Protocol to follow the JoVE Instructions for Authors. For example, 1 should be followed by 1.1 and then 1.1.1 and 1.1.2 if necessary. Please refrain from using bullets, dashes, or indentations.

The numbering of the Protocol has been completely revised and adapted to the Jove requirements.  
9. Please reference figures showing experimental set-up in the Protocol.

As all numbers in the protocol refer to Fig. 2 of the manuscript, we have referenced to this figure once in the beginning of the protocol.

10. Please revise the protocol to contain only action items that direct the reader to do something. The actions should be described in the imperative tense in complete sentences wherever possible. Avoid usage of phrases such as “could be,” “should be,” and “would be” throughout the Protocol. Any text that cannot be written in the imperative tense may be added as a “Note.” Please revise A.4.4, A.5.6, A.5.7, A.6.1, B.5.4, B.6.5, B.6.6, B.6.7, B.7.1, B.7.5, etc., accordingly.

The mentioned protocol steps have been accordingly revised.  
11. Please add more details to your protocol steps. Please ensure you answer the “how” question, i.e., how is the step performed? Alternatively, add references to published material specifying how to perform the protocol action.

The following references have been added to provide further background information: [30,31,32,33,34,36].  
12. A.5.7: Please describe how to perform a mass scan with the QMS user Interface.

The required information has been added.  
13. A.6.6, B.6.5, B.6.7: Please provide more details here.

More details have been provided.  
14. Please include single-line spaces between all paragraphs, headings, steps, etc., and align all text to the left margin. After that, please highlight 2.75 pages or less of the Protocol (including headings and spacing) that identifies the essential steps of the protocol for the video, i.e., the steps that should be visualized to tell the most cohesive story of the Protocol.

The text has been accordingly edited. About 2.75 pages have been marked in yellow.  
15. Please include volume and issue numbers for all references.

All required information is provided for all references.  
16. Although the majority of this does involve a custom-built system, please include any other materials, equipment, and/or programs necessary to carry out this protocol in the Table of Materials.

A complete table of materials has been added.  
  
**Reviewers' comments:**  
  
  
  
Reviewer #1:  
  
Manuscript Summary:  
The manuscript describes in detail a setup and the operation procedure to realize a beam of Thorium ions, which a 2% fraction in the low-energy isomeric state. This state is of highest interest for a continuously growing community in atomic and nuclear physics as well as metrology.  
  
Major Concerns:  
None, the manuscript is clear and VERY detailed. The setup and protocol described here has lead to a multitude of breakthrough results and is currently the defining system in the field.  
Even if it is unlikely that someone will reproduce this setup to follow the described procedures, the individual technical and physical solutions found here are of great importance.  
  
Minor Concerns: None

We thank Reviewer 1 for the report.  
  
  
  
Reviewer #2:  
  
Manuscript Summary:  
The authors describe the methods to obtain isotopically pure ion beam of 229Th, which is important for the search of candidate nuclei of nuclear clock. The methods are used to and can be extended to get long-lived isotopically pure ion beams. The steps of the experiment are described in detail and clearly, and can be followed by the readers. The authors highlighted the critical steps in the provided protocol. The results are useful to the researchers who are concerning the nuclear clocks and isomers. However, the abstract is more like for scientific paper, it would be better if the authors rewrite it and provide more key experimental points. In the materials, the following improvements can be made:

We thank Reviewer 2 for the report. The abstract has been revised as proposed above. Each point will be discussed individually in the following.  
  
1. the figures 1 and 2 are almost duplicate, it could be better to give one figure describing the experimental setups, pointing out the detection part for decay detection and lifetime measurement with inset, the other figure with all the diagrams/sketches. Maybe this is more appropriate for the JoVE.

We have followed this proposal and condensed figures 1 and 2 into a single figure (now Fig. 2). Further, a figure showing the experimental setup was added (now Fig. 1).  
2. in figure 3, the upper three panels are not necessary, because the whole spectrum is presented in the upper panel of figure 4, which is a much nicer presentation. it would be better to rescale the spectrum from 60 to 280. Only the lowest panel, the enlarged plots are useful. This means the authors have to reconsider how to arrange the figures.

We agree that some information given in previous Fig. 3 was redundant. We now reference to Fig. 4 (now Fig. 3) instead und have revised the text accordingly.  
3. in line 436 and 441, Figure 6 (top panel)... and the middle panel of fig 6 ... where top and middle panels are not existing in the figure.

The corrections were implemented.  
  
The method deserves for publication after the modifications suggested above.  
  
  
  
  
  
Reviewer #3:  
  
Manuscript Summary:  
The submitted article presents a detailed overview and protocol that must be followed in order to successfully extract 229Th recoil ions and subsequent daughter isotopes from an ultra-pure buffer gas cell. As illuminated in the abstract and introduction, 229Th is a current hot topic, hosting the lowest energy isomeric state of any nucleus, indirectly deduced to be a few eV above the nuclear ground state. This unique state offers a rich playground for precision spectroscopy and applications, branching fields of nuclear physics, atomic physics as well as time and frequency standards. It is this latter application to which the authors pay particular attention as the relative linewidth of the ground-state transition is such that the isomer potentially qualifies as a nuclear frequency standard of unprecedented precision which, if realized, could be essential for a vast field of applications and fundamental tests. Until recently however the isomeric state itself had only been inferred from indirect measurements and much controversy arose over the years as to whether the radioative decay of the state had been seen. It is now known that this was not the case as a number of background signals can mimic the decay. The Munich team, with a number of outstanding recent results, conclusively proved the existence of the isomeric state through a detection of the internal conversion electrons emitted from the decay of the state in a neutral (atomic) form. In this current manuscript the procedure to repeat such measurements is presented in a clear and step-by-step manner such that other groups, if they had access to the sources and a similar setup would be able to identify the isomeric decay.  
  
The isomer is populated via a 2% branch from the alpha decay of 233U. In order to spatially separate the resulting population from other contaminants including daughters in the radioactive decay chain of 233U, the authors choose to stop the recoil products in an ultra-pure buffer-gas stopping cell, guide them using an electrode system to the exit nozzle, inject into a radiofrequency quadrupole and mass separate using a quadrupole mass spectrometer (QMS). Depending on whether direct detection of the isomeric state is the goal, or whether a lifetime measurement of the state is required, the ions may be gently impinged onto the surface of a multichannel plate detector coupled to a phosphor screen and CCD detector. A number of verification measurements have been supported to prove that registered electron signals indeed originate from the decay of the nuclear isomer.  
  
Following an appropriate abstract and introduction, with an extensive reference list which suitably covers the current status in the field as well as recognizing the earlier (now historical) efforts, the protocol is presented. Mounting of the source, initial evacuation of the vacuum chamber and baking out of impurities in the system, preparation of the buffer gas and supply of ultra-pure helium, application of suitable electric guiding fields (and later the needed modifications required for ion bunching and lifetime measurements), tuning of the ion extraction and the mass separation and final detection of the state are detailed. In order to follow the steps two very detailed figures accompany this aspect of the article, with all components numbered accordingly. A set of representative results (already published prior to this manuscript) are given which present the reader with what one might expect to see, from a mass spectrum of ions extracted from the gas cell, to the signal obtained using the CCD camera which indicates direct detection of the isomeric state. This is useful as the results also indicate what one might expect to detect should a different isotope of uranium be used as a source - again, verifying the existence of the isomer populated in the decay of 233U. Finally, the critical steps are summarized, troubleshooting options are presented, and the method presented here is placed in context with other worldwide efforts which aim at the study of the isomeric state using vastly different methodologies. It should be noted that currently these other methods have either not worked or are currently the subject of significant discussion in the scientific community.  
  
In the following I have some minor comments. The article as it is currently presented is suitable for publication and is thorough enough to be a useful resource for other researchers aiming to learn about this technique.

We thank the reviewer for this comprehensive report. In the following the comments will be considered individually.  
  
1. Abstract, line 28-29, "enabling the investigation of…."

The abstract has been completely rephrased.  
2. Line 48, "For the past 40 years…."

The first part of the introduction has been rephrased.  
3. Line 107, it might be better to use the word "viewed" rather than "inspected" when referring to the CCD camera.

This has been corrected.  
4. Line 108, "…the underlying procedure to generate…"

This has been corrected.  
5. Line 108, "foundation for studying"

This has been corrected.  
6. In the protocol of part A 1.1, the authors state that the source thickness should not exceed 16 nm, however surely this could be exceeded? The only drawback is that for a given source strength, the overall recoil efficiency would be reduced due to the limited range of 229Th recoils in the uranium. But this would presumably be a matter of handling stronger sources with redundant source material.

This is of course correct and the sentence has been accordingly rephrased.  
7. In protocol 2 and 3 the evacuation of the chamber and bake out procedure is presented, followed by the preparation of the gas supply. In this procedure I do not see any explicit mention of monitoring the pressure in the subsequent vacuum chambers. This could be noted, especially after the filling of 30 mbar of helium into the buffer gas cell, so that one might understand the baseline pressures to be expected in the QMS chamber.

The information has been added.  
8. Line 191, protocol 4.4. After this first line I suggest to add (27) as this is where the extraction RFQ is mentioned for the first time.

The information has been added.  
9. Line 249, protocol 5.6. "…passing through the QMS…"

This has been corrected.  
10. Protocol 6.4. Here an accelerating potential of typically +6000 V is applied to the phosphor screen, however in protocol 5.3 only +5000 V is applied. Does this additional voltage make much difference?

For electron attraction the voltage difference between the second MCP plate and the phosphor screen is set to +4000 V. The second MCP plate is changed from +900 V (Step 5.2) to +1900 V (Step 6.4). The phosphor screen is accordingly adapted. In case that this adaptation is not performed, the kinetic electron energy at impact will be reduced by 1/4. This may slightly reduce the brightness of the spot produced on the phosphor screen during electron impact, but would most likely not affect the concept.  
11. Line 290, protocol 1.10. Would it also be an option to vent the system with dry nitrogen from a bottle for example, or is it preferable to use the boiled off nitrogen which seems to be suggested here?

Indeed this would be an alternative and has been added as a note. However, when venting with nitrogen from a bottle, care has to be taken (e.g. by adding an overpressure valve to the vacuum system) not to exceed atmospheric pressure and enter an overpressure regime. Thus the vacuum system would become (unnecessarily) slightly more complex.  
12. Line 339, "The trigger module allows an adjustment of…"

This has been corrected.  
13. Protocol 7.5. Can you verify whether the trigger module which is labeled (30) here should be (24)?

Indeed that was a mistake in labeling.  
14. Representative results. In the first paragraph it is stated that the 229Th3+ and 233U3+ are extracted with about equal intensities. This is also the same for the doubly-charged state according to Fig. 3, however not the case for the singly-charged state. It is claimed to be noteworthy, however no explanation of why this is noteworthy is given. I suggest to expand a little here on why this may be important.

An explanation has been added.  
15. Line 481. "Besides" should be worded, "In addition to…."

This has been corrected.  
16. Line 500, "…for a given source activity"

This has been corrected.  
17. Line 515-516 is a little confusing. Perhaps, "A successful application of voltage is monitored via the current… This current will increase in the case of sparks. If sparks have occurred…"

The sentence has been rephrased in accordance with the proposal.  
18. Line 576-577, "Thus the present paradigm that this would require….can be circumvented".

This has been corrected.  
19. Please note that both figures 1 and 2 are currently very poor quality which needs addressed. It was very hard to read the numbering and text written in these figures - which are the basis of much of the discussion in the protocol list.

We provide the figure now as a vector graphic and hope that this will solve the problem of readability.  
20. I noticed that the valve immediately after the backing pump was not labeled. Should this also be (6)? Is it worth an additional mention or is this valve always open?

This valve is a safety valve which remains open during normal operation, for this reason we think that no labelling is required at this point.

Reviewer #4:  
  
Manuscript Summary:  
The authors propose to visualize the preparation of a pure 229Th ion beam for studies of 229mTh. The isomer of this isotope has an energy of 7.8 (5) eV. At neutral thorium it decays via internal conversion with a half-life of 7 µs. In charged states this channel is energetically not allowed and a radiative decay is the only possible decay channel which has a lifetime, according to calculations, of up to 10.000 s resulting in a very narrow line width. As pointed out in the Introduction of the manuscript there are many potential and rather important applications relying on such an isomer.  
No doubt, the presented material is valid. It has been presented prior to this paper in highly rated refereed journals.

We thank Reviewer 4 for the report. In the following, the points will be discussed individually.  
  
Major Concerns:  
The authors focus in the yellow highlighted part on the preparation procedure of the isomeric beam and the detection of the isomeric decay. It describes in detail the rather involved figure 1 which indicates the essential parts of the protocol to be filmed. I strongly support the production of such a film although I have no idea how this can be done on the basis of such an involved diagram. However, this is not my task as has been pointed out in the criteria for reviewing the article. Anyway, I have the feeling that the film must show the experimental setup and the signal generation, e.g., on the basis of figure 2 and 3 of the Nature Vol. 533 article. Such figures are missing in the manuscript. I do not know whether this is of importance for the film production, or not.

We partly follow the proposal and have added a figure showing a drawing of the experimental setup (new Fig. 1). We consider the physical details of the detection process to be of minor importance in this rather technical journal format. This, however, can be subject to further discussion also with the editors.  
  
Minor Concerns:  
Line 62: Add explanations of the acronyms GPS, GLONASS or GALILEO in words.

The explanation of the acronyms has been added as far as possible. As it turned out “Galileo” is not an acronym but rather the name of the program.   
Line 414: The quoted charge state for thorium 3+ should be 2+.

This has been corrected.  
Line 419: The voltage -900 V is inconsistent with regard to the nature article and the figure caption.

This has been corrected.  
Line 436: Figure 6 (top panel) is not in accord with the shown figure where it is a left panel.

This has been corrected.  
Line 441: The middle panel of Figure 6 does not exist as well in the shown figure.

This has been corrected.  
The table JoVE\_Materials is empty.

A complete Material list has been added.