

Laser Excitation of the 1S – 2S Transition in Singly-Ionized Helium

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Communications Physics.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

Authors demonstrate the two-photon (of not equal wavelength) excitation of the ground state of the hydrogen-like helium ion. This is a significant achievement, but not necessarily very interesting for a wide audience. Moreover, I do not see a clear path how it translates into precision spectroscopy of He^+ . Maybe, it is due to lack of my experience in the uv laser spectroscopy. Independently on this, this paper is well written. Nevertheless, I would have few specifics comments, which may enhance the writings:

1: I would suggest to mention in the first paragraph other important hydrogenic systems, such as positronium and muonium

2: In the third paragraph, I would not use the term „energy gap” for the energy difference between discrete levels

3: The logic of the sentence: „This makes excitation by two photons of the same wavelength... very challenging in He^+ because two 61 nm photons are required.” is not very obvious

4: I would mention in the Introduction about the Gamma Factory Project at CERN, which aims for photon excitations of heavy ions. Here the idea is to employ optical photons with large gamma factors of incoming ions. This approach can also be used for He^+ .

In conclusions, I do not have a definite recommendations at this stage.

Reviewer #2

(Remarks to the Author)

Report on Manuscript COMMSPHYS-24-0750-T, "Laser Excitation of the 1S – 2S Transition in Singly-Ionized Helium" by Gründemann et al.

The paper reports on the first ever laser excitation of the 1S-2S 2-photon transition in hydrogen-like He ions. This is a milestone in physics and should therefore be published in Nature or Nature Communications. The manuscript is very well written.

Quantum Electro Dynamics (QED) is the theory that explains most of the world we experience (with the exception of gravity and radioactive decay), from atoms to chemistry to lasers and more. It's the best-tested theory, agreeing with experiment on the 13th decimal digit in measurements such as the anomalous magnetic moment of the electron or transitions in atomic hydrogen H.

In H, the 2-body theory has been refined over the last several decades, and comparison between experiment and theory tests QED or reveals fundamental physical constants with great accuracy.

The present paper is concerned with He, the next atom in the periodic table. For years, precision measurements have been performed on neutral He, and many discrepancies between measurements have been revealed. From theory, however, the 3-body system of neutral He atoms is much more difficult to calculate, and higher-order QED contributions have only recently been calculated. Still, significant discrepancies persist, for example also between the 3He - 4He isotope shift comparison of ordinary He atoms and muonic He ions.

The present manuscript reports on the first successful measurement of the missing link between these 2 worlds: It is the first laser excitation of the 1S - 2S transition in the 2-body system He^+ . This transition can in principle be measured with outstanding precision, given that the 2S state is metastable and the resonance is therefore very narrow. Unfortunately, the laser excitation requires a very difficult laser wavelength in the XUV, which has not been available until very recently. Most importantly, however, the He^+ ion can be calculated using the very well established 2-body theory from hydrogen.

In Gründemann et al, the 1S - 2S transition is excited not by a symmetric 2-photon transition with two 60nm photons, but with a 30nm photon plus an infrared photon. This is the key to the first ever laser excitation of He^+ .

Great care has been taken to ensure that the signal (He^{2+} ions) is really from the 2-photon transition 1S - 2S ->ionization, and not from the 1-photon 1S - 2P ->ionization process. Both measurements and detailed simulations

While the present experimental uncertainty is still huge, the expectation is that this will soon improve dramatically and open the way to the highly important comparison of He, He^+ , and He (and Li^+ , Be^{2+} , C^{4+} etc. all of which have been measured with outstanding precision in recent years).

This will allow improved tests of QED and the Standard model (any discrepancies between e.g. nuclei with different numbers of protons or neutrons could point towards new forces outside the Standard Model), and it will most definitely help guide the theorists to further develop QED of 3-body systems.

The manuscript is very well written, but I would like to raise one point:

Maybe the fact that this measurement took place using the isotope 3He (for very good reasons!) could be stated earlier, possibly already in the abstract? I was reading the manuscript with the (probably natural) assumption that 4He was used and I only learned about 3He on the 2nd page. While this is not a big issue, it left me a bit puzzled, and I had to re-read the beginning of the paper again, because:

The most notable difference between 3He and 4He is of course the presence of a hyperfine splitting in 3He . This is however not mentioned anywhere in the text, not even in the supplement. Has the HFS been taken care of in the simulations? Are there any important differences in the excitation process between 4He^+ (no HFS) and 3He^+ (with HFS)? I would like to read about this in the paper. If space constraints exist then please write a brief statement in the paper, with more detailed explanations in the supplement.

And 2 very minor suggestions:

"unequal-wavelength excitation" sounds a bit strange to me (not a native speaker, though). How about "asymmetric 2-photon excitation"?

line 102: " He^{2+} signal does not coincide with background.." could be made clearer by stating the reason "is separated by the charge/mass ratio"

Reviewer #3

(Remarks to the Author)

In this manuscript the authors report the first successful laser excitation of the 1s - 2s transition in the hydrogenic helium ion. As a culmination of the long term technical developments in the authors' group, this result is a milestone towards precision

spectroscopy of the He⁺, which again is a major opportunity for new tests of the standard model. Additionally, the manuscript reports on a model simulation of the excitation process based on a solution of the time-dependent Schrödinger equation, which, to a certain degree, seems consistent with the experimental observations.

The manuscript is very clearly written, easy to follow and an interesting read.

The authors use a somewhat complementary method to the 2x61nm scheme with two unequal, almost resonant photons.

However, especially the very high intensity of the fundamental NIR beam causes significant state dressing and correspondingly dramatic Stark shifts. While a precision determination of the 1s-2s frequency clearly hasn't been a priority in this work, the unperturbed wavelength has actually been extrapolated based on measurements with different intensities.

Here, personally I would have anticipated a discussion of the (probably complex) error budget. The manuscript lacks any information on which uncertainties are included in the presented number (it seems to implicitly rely on the observed (in exp and sim) linear intensity relation of the Stark shift). Couldn't there be e.g. non-linear effects connected to the ionisation position (not included in the TDSE?) of the He atoms coupled to the saturation observed in the excitation and second ionisation processes? A short discussion, possibly outside of the main text, could be very helpful especially to judge the upcoming challenges for the RCS, where varying Stark shifts between the two Ramsey pulses or between cycles, connected to residual motion of the trapped ion, might become a challenge.

Despite this, the presented work and the overall quality of this manuscript makes this work interesting to the atomic physics community and beyond and in my opinion renders it an imperative to publish this manuscript, once the authors have reacted to my aforementioned remarks.

Version 1:

Reviewer comments:

Reviewer #3

(Remarks to the Author)

The authors have appropriately answered my questions and comments and correspondingly adjusted the manuscript.

Looking at the comments of the other referees, I do agree with R1 that an early mention (not explanation (asymmetric (strong) two photon excitation->Ramsey)) of a future precision measurement concept could be helpful to support the reader in understanding the impact of this work. Otherwise, I support publication of the manuscript.

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Amsterdam, September 12, 2024

Dear Editor and Reviewers,

First, we like thank the reviewers for their positive recommendations and the comments that helped us improve the manuscript.

The point-by-point response to the reviewer comments starts on the next page, but here we want to address an issue we mentioned to the Editor earlier: after we sent in the manuscript, we became aware of a publication about 1S-2S excitation in He^+ with a free-electron laser (FEL): “Five-photon sequential double ionization of He in intense extreme-ultraviolet free-electron laser fields”, Y. Hikosaka et al., Phys. Rev. A 90, 053403 (2014). We had not found this paper before because the title does not immediately indicate 1S-2S excitation of He^+ . However, the authors used a FEL to induce multi-photon ionisation of He, and saw a signal peak of He^{2+} when varying the FEL wavelength near 60 nm. It is a sign of 1S-2S excitation in He^+ with two photons at 60 nm as an intermediate step.

This demonstration with a free-electron laser has no relevance for precision measurements due to the incoherent nature of FELs, so our demonstration is the first that does have the potential for precision measurement. But in our manuscript we claim to be the first to excite the transition optically by any means, and that appears to be not correct. Therefore we changed this part in the abstract:

“We report to the best of our knowledge the first laser excitation of the 1S – 2S transition in singly-ionized helium (He^+), a hydrogen-like ion with much higher sensitivity to QED than hydrogen itself. The transition requires two-photon excitation in the challenging extreme ultraviolet range. We achieve this ...”

To:

“We report laser excitation of the 1S – 2S transition in singly-ionized helium He^+ , a hydrogen-like ion with much higher sensitivity to QED than hydrogen itself. The transition requires two-photon excitation in the challenging extreme ultraviolet wavelength range, which we achieve with a tabletop coherent laser system suitable for precision spectroscopy. The transition is excited ...”

And in the main text, lines 55-59, we changed “As a consequence We overcome...” to:

“As a consequence, laser excitation of the 1S – 2S transition has only been reported with a high-power but incoherent free-electron laser source [Hikosaka2014] that is not suitable for precision spectroscopy. In this article we report, to the best of our knowledge, the first laser excitation of the 1S-2S transition in He^+ with a coherent frequency-comb laser-based source. We overcome ...”.

and we added the Hikosaka et al. paper to the references.

On the next page is a list of our response/actions (in blue) based on the specific comments/questions raised by the reviewers.

We hope that with the applied changes, the manuscript can be accepted for publication in Communications Physics.

Best regards,
On behalf of all the authors,
Kjeld Eikema

List of our response/actions (in blue) based on the specific comments/questions raised by the reviewers.

Reviewer #1

1.1: “Moreover, I do not see a clear path how it translates into precision spectroscopy of He^+ . Maybe, it is due to lack of my experience in the uv laser spectroscopy”

We understand it is not immediately obvious, but a brief explanation is given in the ‘Conclusion and Outlook’ section of the paper where we also refer to our previous work. In the Supplementary Material section V we gave more details, and discuss how we will deal with the ac Stark shift and Doppler shift in future precision Ramsey-Comb spectroscopy. There is no room for all the details, therefore we are writing now an extensive feasibility paper.

1.2: “I would suggest to mention in the first paragraph other important hydrogenic systems, such as positronium and muonium”

We now added both systems to the introduction by inserting at line 18: “positronium [Sheldon2023], muonium [Kanda2021]”

1.3: “In the third paragraph, I would not use the term „energy gap” for the energy difference between discrete levels”

We now changed it to “energy difference” at line 44 of the current manuscript.

1.4: “The logic of the sentence: „This makes excitation by two photons of the same wavelength... very challenging in He^+ because two 61 nm photons are required.” is not very obvious”

Indeed, this is a bit cryptic due to our effort to shorten the text. We now changed this sentence (line 54) to:

“... very challenging in He^+ because a high photon flux is required for simultaneous absorption of two 61 nm photons.”

1.5: “I would mention in the Introduction about the Gamma Factory Project at CERN, which aims for photon excitations of heavy ions. Here the idea is to employ optical photons with large gamma factors of incoming ions. This approach can also be used for He^{+} .”

We think that the Gamma Factory (GF) project is very interesting, but it seems aimed towards rather different spectroscopy at much higher energy (keV) and with orders of magnitude (at least 6-7) lower energy resolution than we aim for with He^+ . We think that the GF does not really fit in the current manuscript and would complicate the flow of the text.

Reviewer #2

2.1: “Maybe the fact that this measurement took place using the isotope ^3He (for very good reasons!) could be stated earlier, possibly already in the abstract?”

We agree; although it is not essential for our demonstration, it is indeed nicer to mention the use of $^3\text{He}^+$ in the abstract and earlier in the main text. Therefore, in the abstract we changed “ He^{+} ” to “ $^3\text{He}^{+}$ ”, and we moved the sentence “Note that we use ^3He ...” to line 85 and changed it to:

“We use ^3He instead of the more abundant ^4He isotope to avoid false signal from background molecular hydrogen (see Methods). The hyperfine structure of ^3He has not been taken into account because it is much smaller than the resolution of the current experiment and therefore has no noticeable influence.”

2.2: “The most notable difference between ^3He and ^4He is of course the presence of a hyperfine splitting in ^3He . This is however not mentioned anywhere in the text, not even in the supplement. Has the HFS been taken care of in the simulations?”

Hyperfine structure has not been taken into account in the simulations or the experiment because it is so much smaller than the bandwidth of the XUV/NIR pulses involved, the resolution of the experiment, and the induced ac-Stark shifts. Therefore, it has no noticeable influence on the excitation process in our single-pulse experiment. This is now explicitly stated in the text (see our answer at point 2.1).

2.3: “Are there any important differences in the excitation process between $^4\text{He}^+$ (no HFS) and $^3\text{He}^+$ (with HFS)? I would like to read about this in the paper. If space constraints exist then please write a brief statement in the paper, with more detailed explanations in the supplement.”

As explained at remark 2.2, for the current 1-pulse excitation experiment there is no relevant difference between $^4\text{He}^+$ and $^3\text{He}^+$, but for precision spectroscopy with Ramsey-Comb Spectroscopy (RCS) in the future there is. In the case of $^4\text{He}^+$ just two Ramsey fringes are required to determine the transition frequency. For $^3\text{He}^+$ more Ramsey fringes are needed because the hyperfine structure leads to multiple transitions, giving rise to an additional modulation of the Ramsey signal. Fortunately, the hyperfine structure of the 1S and 2S are both well-known from RF-measurements done in the past by other groups, so that it can easily be taken into account. Even without this knowledge, RCS can resolve and determine the HFS from multiple Ramsey scans, as we showed with our first demonstration of RCS on rubidium (Nat. Phys. 10, 30 (2014) and Phys. Rev. A 89, 052510 (2014)). We feel that these details go beyond the scope of the current paper, but we will discuss them in the upcoming feasibility study we are currently writing about RCS of a single trapped $^3,4\text{He}^+$ ion.

2.4:

““unequal-wavelength excitation” sounds a bit strange to me (not a native speaker, though). How about “asymmetric 2-photon excitation”?”

We changed it now in line 64 to the suggested “asymmetric 2-photon excitation”

2.5:

line 102: “ He^{2+} signal does not coincide with background..” could be made clearer by stating the reason “is separated by the charge/mass ratio”

We moved this sentence and reformulated it, see our answer at point 2.1.

Reviewer #3

3.1: ” While a precision determination of the 1s-2s frequency clearly hasn't been a priority in this work, the unperturbed wavelength has actually been extrapolated based on measurements with different intensities. Here, personally I would have anticipated a discussion of the (probably complex) error budget. The manuscript lacks any information on which uncertainties are included in the presented number (it seems to implicitly rely on the observed (in exp and sim) linear intensity relation of the Stark shift). Couldn't there be e.g. non-linear effects connected to the ionisation position (not included in the TDSE?) of the He atoms coupled to the saturation observed in the excitation and second ionisation processes?”

Indeed, the 1S-2S frequency extrapolation as a function NIR intensity was only done for identification of the transition (as stated in the manuscript) and was not meant as a precision measurement, and therefore not presented as such. However, we understand the curiosity about the determination

uncertainty and the question about possible nonlinearities in the extrapolation. We assumed strict linear extrapolation and combined the fitting error with the calibration uncertainty of the simple fiber-spectrometer we used (calibrated with a reference laser diode as explained in the first section in Methods). No nonlinearities are expected from the ionization position because the detected ions originate from a small region of space of ~ 6.25 micron FWHM (which is so small due to the small size of the 17th harmonic creating the ions, and the 25th harmonic exciting the ions). The minimum NIR beam diameter is about 43 micron FWHM and therefore much larger, so that all detected ions experience at least 98% of the peak intensity. This means that the used NIR intensity for extrapolation is representative for all ions.

Saturation effects are not expected because of the maximum excitation probability of 10^{-3} . The simulation (including 9-photon ionization) nicely extrapolates linearly to a value of 789.88(2) nm, consistent with the expected theoretical value, indicating that on a single-atom level there is also no significant nonlinear effect.

We extended section III in the Supplementary Material to include the above explanation and how the total error of the extrapolated transition value was obtained. This part (indicated in blue) starts with: “The zero intensity extrapolation ...”, and ends with “..., as is described in Sec. V.”

We also changed the sentence at line 144 in the main manuscript to refer to the Supplementary Material.

When we reviewed the extrapolation and calibration again because of the question by the reviewer, we discovered a minor error in the extrapolation of the experimental results, leading to a small shift of 0.02 nm in the NIR fundamental wavelength required to observe the 1S-2S transition (at zero NIR intensity). Therefore, we corrected the experimental value at line 147 from 789.95(27) nm to 789.97(27) nm. This does not change the conclusions, or anything else, in any way. The simulation result was not affected and has remained the same.

3.2: “A short discussion, possibly outside of the main text, could be very helpful especially to judge the upcoming challenges for the RCS, where varying Stark shifts between the two Ramsey pulses or between cycles, connected to residual motion of the trapped ion, might become a challenge.”

Indeed, a variation of the ac Stark shift because of ion motion is something to be careful of. In the Supplementary Material section V (part A) we discuss the effect of a possible variation of the ac Stark shift on RCS due to pulse-to-pulse variations. We also discuss ground state cooling and synchronization of the RCS excitation pulses to the motional period of the ion (part B), so that the ion is always excited in the same position (in the direction of the laser beam). This takes care of the Doppler shift, the recoil shift and ac-Stark variations in the direction of the excitation beam. Sideways motion of the ion (in the direction of the trap axis, or perpendicular to the laser beam and ion trap axis) between two RCS pulses was not discussed, but it will be very small ($\ll$ few micron) compared to the NIR beam size (>43 micron), because in that direction the ion will remain cold (if we separate them adiabatically). In that case the ac Stark shift variation due to sideways motion should be much smaller than the other ac Stark inducing effects (most notably the pulse-to-pulse variation and long-term variation). It does provide another argument why ground state cooling would be good, so we now deleted the remark that ground state cooling is not strictly necessary when synchronization is employed, and we added this remark in Section V (in blue) at the end of part A:

“We can also consider the variation in ac Stark effect by movement of the ion. What matters is again only the difference in ac Stark effect between Ramsey fringes at different time delay. In the direction of the excitation beam the ac Stark effect is already very constant, and variation is further suppressed by synchronisation of the RCS pulses with the motion of the ion, as proposed in the next section to suppress the Doppler and recoil effect. Perpendicular to that axis we expect the ion to remain cold (no recoil from excitation, and any movement of the ions will be done adiabatically so that they remain cold and well localised) with a sideways movement well below a micrometer. This is so much smaller than the NIR beam diameter (FWHM $>43\mu\text{m}$) that no relevant change in ac Stark effect is expected

from motion. Mechanical vibration of the trap or laser beams is also expected to be much smaller than the NIR beam diameter.”

More explanations in the Supplementary Material section is beyond the scope of the current manuscript (it already became quite lengthy). The main purpose of section V in the SM is to show that our excitation method is a viable route towards precision spectroscopy. However, there are many more aspects, and these will be treated in an extensive feasibility study that we are currently preparing.

Response to reviewer comments after a second round of reviewing of the original manuscript
COMMSPHYS-24-0750A

Amsterdam, November 22, 2024

Dear Editor,

There was one final remark by Reviewer 3: “The authors have appropriately answered my questions and comments and correspondingly adjusted the manuscript. Looking at the comments of the other referees, **I do agree with R1 that an early mention (not explanation (asymmetric (strong) two photon excitation->Ramsey)) of a future precision measurement concept could be helpful to support the reader in understanding the impact of this work.** Otherwise, I support publication of the manuscript.”

In our view this was mentioned at the end of the Introduction, but we now changed the wording and order a bit to emphasize it more.

Original sentence:

“Our demonstrated excitation method is fully compatible with Ramsey-comb spectroscopy (RCS) [30–32] based on two amplified pulses from an FC laser, and therefore lays the foundation for future precision studies of hydrogen-like He^+ ”

New sentence:

“Our demonstrated excitation method is an important step towards future precision spectroscopy of hydrogen-like He^+ , which we pursue based on Ramsey-comb spectroscopy (RCS) [30–32] with two amplified pulses from a FC laser.”

Best regards,

On behalf of all the authors,

Kjeld Eikema