Precision Spectroscopy in the Deep-UV and Shorter Wavelengths for Tests of Quantum-Electrodynamics and the Proton Radius Puzzle

M. Favier[1], L.S. Dreissen[1], C.F. Roth[1], E.L. Gründeman[1], J.J. Krauth[1], E.J. Salumbides[1], W. Ubachs[1], R.K. Altmann[1], and K.S.E. Eikema[1]
[1]LaserLaB, Department of Physics and Astronomy, Vrije Universiteit Amsterdam, De Boelelaan 1081, 1081 HV Amsterdam, The Netherlands
k.s.e.eikema@vu.nl

***Abstract*—In this paper, we describe our experimental efforts towards tests of fundamental physics by performing high-resolution spectroscopy of simple atomic and molecular systems at deep-UV and shorter wavelengths. Using a new spectroscopy technique that combines high laser power with high accuracy, we demonstrate accurate spectroscopy in the deep UV, and key steps towards spectroscopy in the extreme ultraviolet (XUV) with kHz or better precision. These experiments are aimed at performing highly accurate tests of quantum electrodynamics, and the relevance for the 'proton radius puzzle' will also be emphasized.**

***Index Terms*—Spectroscopy, ultrafast optics, precision measurements, frequency measurements, atomic measurements, molecular measurements.**

I. Introduction: Spectroscopy, Quantum Electrodynamics and the Proton Radius

Quantum electrodynamics (QED) is arguably one of the most successful and best tested theories in physics [1]. The historic testing-ground for such high-precision experiment/theory comparisons is laser spectroscopy of atomic hydrogen [2]. However, energy levels in atomic hydrogen have been experimentally determined with much higher accuracy than what QED theory can provide, because it is hampered by the uncertainty in the experimental determination of one of its key parameters: the proton charge radius. Therefore hydrogen spectroscopy is used to obtain a proton radius, under the assumption that the QED effects are known with sufficient accuracy.

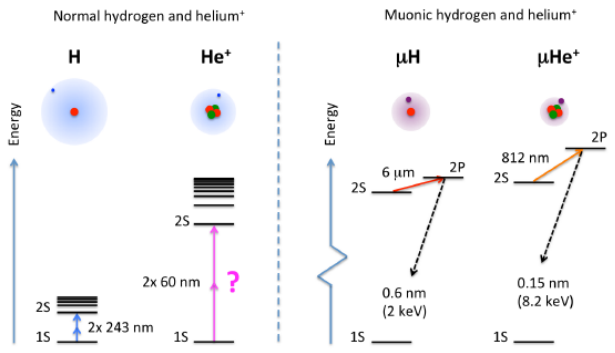

Fig. 1. Principle of testing QED and nuclear size by high-resolution spectroscopy at different wavelengths in hydrogen (2-photon at 243 nm), helium$^+$ (2-photon at 60 nm), and their muonic counterparts with single photon transitions at 6 μm and 812 nm respectively (simplified level schemes).

However, when spectroscopic measurements were performed on muonic hydrogen, a 4% smaller proton radius was found, 5.6 σ away, also compared to the most recent CODATA 2014 value [4]. This problem is now known as the proton radius problem. A more recent value for the proton radius was obtained from 2S-4P spectroscopy [3] in electronic hydrogen and it confirmed the muonic value. This confusing state of matters requires further experimental tests, and comparisons of QED and the Rydberg constant derived from both electronic and muonic species spectroscopy (see Fig. 1).

We pursue two different routes in order to contribute to solving the puzzle. One is precision spectroscopy of molecular hydrogen on the two-photon X-EF band to extract a proton radius and to test molecular quantum theory. The targeted accuracy is 10 kHz. This level of accuracy, together with the H_2 dissociation energy and similar improvements in the theory would provide a value of the proton radius with a precision of 1%. The second experiment is based on 1S-2S spectroscopy in electronic helium ions, to be compared with spectroscopy on the 2S-2P transition in muonic helium ions performed by the CREMA collaboration. This comparison will either provide new information on the proton radius puzzle by a comparison of the helium nuclear radius [4], or it could lead to an improved test of bound-state QED. In both experiments deep-UV and shorter wavelengths are needed.

II. Ramsey-Comb Spectroscopy: Precision and Power Combined

As mentioned above, some of the most interesting spectroscopic targets for tests of QED and the proton radius puzzle lie in the deep UV or even XUV. The synthesis of coherent (laser) radiation at such extreme wavelengths with enough power to excite narrow transitions is a great experimental challenge. To that end, we developed a method that combines high power with high precision, called Ramsey-comb (RC) spectroscopy [5]. It is based on excitation with different pairs of amplified and subsequently up-converted frequency comb laser pulses.

The "macro-delay" between the two selected pulses can be coarsely tuned by integer multiples of the repetition time of the comb, up to several μs, and finely tuned on an attosecond scale around a particular macro-delay to scan a Ramsey fringe. The

978-1-5386-0974-3/18/$31.00

frequency of the transition is obtained by fitting the relative phase of a set of these Ramsey fringes (see Fig. 2), making the technique intrinsically immune to any phase shift that is independent of pulse delay. The frequency comb provides an absolute time and frequency calibration, while a parametric amplifier provides the amplification of selected pairs of comb pulses to the mJ level for efficient subsequent up-conversion, enabling kHz-level vacuum-ultraviolet (VUV) spectroscopy.

III. High-Resolution Ramsey-Comb Spectroscopy of Molecular Hydrogen

Using the Ramsey-Comb spectroscopy method, we measured the two-photon EF-X transition (v=0, J=1) in H_2 [7] at 202 nm in a beam of hydrogen molecules generated from a collimated pulsed supersonic expansion. The 202 nm light is

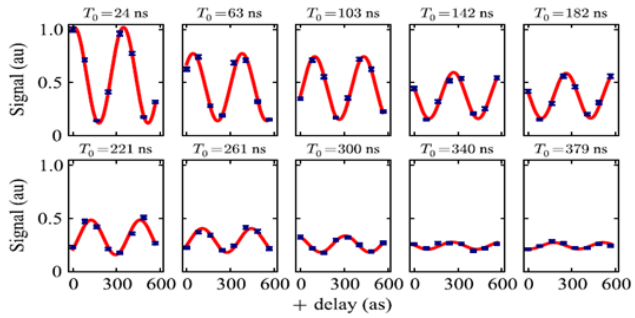

Fig. 2. Measured Ramsey-comb fringes while interrogating the EF-X transition in H_2 at 202 nm. The graphs show the excited state population as a function of delay between the two Ramsey-comb pulses, for 10 different macro-delays (see section II). The contrast decay arises mainly because of the finite lifetime of the excited state.

applied in a counter-propagating-pulse geometry which allows the suppression of the 1st order Doppler effect. The final accuracy is in the 10^{-11} range in fractional units, nearly 2 orders of magnitude better than previous measurements, and further improvements are envisioned to reach the targeted 10 kHz level accuracy [7].

IV. Towards Ramsey-Comb Spectroscopy of the 1S-2S Transition in He^+

Building on the very promising results from the H_2 experiment, we want to perform the first-ever measurement of the 1S-2S transition in singly-ionized helium. We will combine

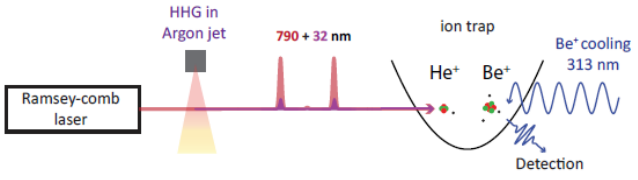

Fig. 3. Scheme of the He^+ experiment. He^+ and Be^+ are co-trapped in a high-secular-frequency ion trap and the cooling of the He^+ ion as well as the detection of its internal state is done sympathetically via the Be^+ ion.

a 790 nm photon with its 25th-harmonic to perform two-photon RC spectroscopy with unequal photon wavelengths to enhance the transition probability (with respect to a 2×60 nm equal photons scheme, thanks to the higher power of the fundamental 790 nm beam). He^+ will be trapped in a linear Paul trap and sympathetically cooled with Be^+ since no available transition allows for direct cooling of He^+ ions (See Fig. 3 for a schematic depiction of the He^+ experiment). The targeted ultimate accuracy for this measurement is below 1 kHz, providing one of the most stringent tests of bound-state QED, and giving valuable insight into the proton radius puzzle (see section I). However, it remains to be demonstrated how the high-harmonic generation process will affect the accuracy of the experiment. Therefore, an intermediate experiment is in progress where we combine the RC technique with high-harmonic generation by exciting xenon with a pair of up-converted pulses at ≃113 nm.

V. Conclusion

In conclusion, high-precision spectroscopy in the deep UV and XUV regions of the spectrum, while still technically very challenging, will enable new tests of bound-state QED and the proton radius puzzle. We have demonstrated high-resolution spectroscopy by exciting H_2 at 202 nm using the Ramsey-comb excitation method, and measured the transition frequency with 10^{-11} accuracy. This measurement paves the way for improved fundamental tests with molecular hydrogen. We have also presented progress towards extending these capabilities further into the XUV, with the prospect of exciting for the first time the 1S-2S transition in singly-ionized helium.

Acknowledgment

The work in this paper was funded by the ERC Advanced Grant QED-PROTONSIZE, by the Foundation for Fundamental Research on Matter (FOM) through its Program 125 and Projectruimte 12PR3098, and by The Netherlands Organisation for Scientific Research (NWO) through its program MYSTP (165).

References

[1] S.S. Schweber, "QED and the men who made it: Dyson, Feynman, Schwinger and Tomonaga," *Princeton University Press*, 1994.

[2] A. Matveev, C. G. Parthey, K. Predehl, J. Alnis, A. Beyer, R. Holzwarth, T. Udem, T. Wilken, N. Kolachevsky, M. Abgrall, D. Rovera, C. Salomon, P. Laurent, G. Grosche, O. Terra, T. Legero, H. Schnatz, S. Weyers, B. Altschul, and T. W. Hänsch, "Precision measurement of the hydrogen 1S2S frequency via a 920-km fiber link," *Phys. Rev. Lett.* 110, 230801, 2013.

[3] A. Beyer, L. Maisenbacher, A. Matveev, R. Pohl, K. Khabarova, A. Grinin, T. Lamour, D. C. Yost, T. W. Hänsch, N. Kolachevsky, T. Udem,"The Rydberg constant and proton size from atomic hydrogen," *Science*, 358, 6359, 79-85, 2017.

[4] R. Pohl et al. (The Crema Collaboration),"The size of the proton," *Nature*, 466, 213-216, 2010; A. Antognini et al. (The Crema Collaboration),"Proton Structure from the Measurement of 2S-2P Transition Frequencies of Muonic Hydrogen," *Science*, 339, 6118, 417-420, 2013.

[5] J. Morgenweg, I. Barmes and K. S. E. Eikema "Ramsey-comb spectroscopy with intense ultrashort laser pulses," *Nature Physics* 10, 30-33, 2014.

[6] R. K. Altmann, S. Galtier, L. S. Dreissen and K. S. E. Eikema "High-precision Ramsey-comb spectroscopy at deep ultraviolet wavelengths," *Phys. Rev. Lett.* 117, 173201, 2016.

[7] R. K. Altmann, L. S. Dreissen, E. J. Salumbides, W. Ubachs, and K. S. E. Eikema "Deep-ultraviolet frequency metrology of H_2 for tests of molecular quantum theory," *Accepted for publication in Phys. Rev. Lett.* 2018.