Advancing Precision Ramsey-Comb Spectroscopy through High-Harmonic Generation

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Summary

The energy structure of simple atomic systems, in particular the hydrogen atom, has been studied for more than a century. The theoretical description of the energy levels of a hydrogen atom was first given by Niels Bohr, who stated that the electron in the atom can only occupy discrete energy levels, which later led to the formulation of quantum mechanics. The experimental investigations of transitions between these energy levels is called spectroscopy. Such transitions can be enabled by light, if the energy of the photons (related to the wavelength or frequency) of the light matches exactly with the energy between the two states. Therefore the field of spectroscopy has improved tremendously with the invention of the laser, as it has a high spectral purity. With increasing spectroscopic accuracy, substructure in the energy levels of the atom were revealed. These additional features led to the development of more extensive theoretical models, for example by including relativistic effects, and eventually to the formulation of the theory of quantum electrodynamics (QED). This theory describes the interaction of light and matter and the influence of the creation and annihilation of virtual particles on the energy structure of the atom. The ongoing comparison between more accurate measurements and improved calculations has tested this theory with extremely high precision and it has passed every time. The invention of the frequency-comb laser (referenced to an atomic clock) has been instrumental in this process because it provides an accurate absolute frequency calibration for the spectroscopy laser. As a result, the $1S - 2S$ transition frequency in atomic hydrogen has been determined with a dazzling 15−digit precision. The theoretical prediction of the transition frequency also relies on several fundamental quantities, such as the proton charge radius or the Rydberg constant, which can only be extracted from measurements. In some cases the accuracy of theoretical energy level predictions is limited by the accuracy of those fundamental quantities, and this hampers further tests of

fundamental theory. In hydrogen it is the case with the proton charge radius and therefore the accuracy of the value was improved with spectroscopy on a more exotic atom: *muonic* hydrogen. The extracted value from this measurement was ten times more accurate but also 4% off from the generally accepted value, corresponding to a 5.6σ deviation. This conundrum is known as the proton radius puzzle, which is being investigated by performing more accurate measurements in atomic hydrogen, but also in other species as this might provide new insights.
The work presented in this thesis is focused on precision spectroscopy of singly-ionized helium and *molecular* hydrogen. As the first target is hydrogen-like its energy level structure can be calculated with a similar high level of precision. Because QED terms scale with higher-orders of the atom number (Z) they are bigger in He^+. Moreover, a similar comparison as in hydrogen can be made with measurements which have been performed in muonic He^+. The second target, the hydrogen molecule, is a benchmark system for tests of molecular quantum theory, as its ground state energy (relative to the dissociation or ionization energy) has been calculated with an impressive level of accuracy. The additional degrees of freedom, such as rotation an vibration, enrich the energy level structure and therefore enable extra possibilities to investigate potential extensions of the standard model, including hypothetical fifth forces on the angstrom length scale. The experimental challenge arises from the short excitation wavelength that these species require. Excitation to the first electronically excited state in H_2 requires 202 nm, which is deep in the ultraviolet part of the spectrum. The $1S - 2S$ transition in He^+ requires an even shorter wavelength of 60 nm or lower. This is in the extreme ultraviolet spectral range, which is absorbed by most materials including air. At these wavelengths, no (known) laser materials exist and therefore generation of this light for precision spectroscopy relies (mostly) on nonlinear processes for up-conversion of the frequency. For these processes, a high peak-intensity is required and therefore pulsed lasers are generally used for this purpose. The broad bandwidth of a pulsed laser typically limits the spectroscopic precision. However, the repetitive nature of a frequency-comb laser enables a much higher level of accuracy. We employ the Ramsey-comb spectroscopy technique, based on two amplified frequency-comb pulses, to combine the required peak-intensity with high spectroscopic precision. It was

developed in our lab and has shown to perform well in the near-infrared and deep-ultraviolet wavelengths using low-harmonic generation in nonlinear crystals. The technique can readily be used for high-precision spectroscopy of the $X - EF$ transition in H_2 by fourth-harmonic generation of 808 nm. One of the main goals of this work was to also extend this method with high-harmonic generation for $1S - 2S$ spectroscopy in He^+.

As mentioned before, Ramsey-comb spectroscopy relies on a Ramsey-type excitation with two amplified (and up-converted) frequency-comb pulses. In Ramsey spectroscopy, excitation with two consecutive pulses leads to interference between two created superpositions of the ground and the excited state. For two phase-locked pulses, the final excited state population is determined by the delay and the phase difference between the pulses. Scanning either one of these parameters leads to an oscillation of the excited state population, known as a Ramsey fringe, from which the transition frequency can be determined. The pulses emitted by a frequency-comb laser are ideal for Ramsey spectroscopy, as they have a fixed (and known) inter-pulse delay and a well-defined phase relation. However, a significant (and typically unknown) phase shift is induced by amplifying a pair of these pulses for efficient up-conversion, which leads to a shift of the extracted transition frequency. In order to circumvent this problem, a series of Ramsey fringes is recorded at intervals that are an integer multiple of the repetition time of the laser, while only the differential phase of the fringes is used to determine the transition frequency. This leads to a significant suppression of any constant phase shift (and also the ac-Stark shift). The extension of this technique with high-harmonic generation is not trivial, as the up-conversion process itself can lead to a phase shift which is delay-dependent. A more extensive description of this technique and other physical concepts are given in Chapter 2. It starts from some basic aspects of pulses and pulse trains, and is followed by a description of the key properties of the frequency-comb laser and nonlinear processes in both the perturbative and in the strong-field regime. After this, the method of Ramsey-comb spectroscopy is explained and the expected consequences from combining this technique with high-harmonic generation is discussed.

The experimental implementation of the Ramsey-comb technique is described in the first part of Chapter 3. The setup is based on a Ti:sapphire

frequency-comb laser with an emission spectrum centered around 800 nm. The modes of this laser are is referenced to a commercial cesium-atomic clock. Two pulses emitted by the frequency-comb laser are selectively amplified in an optical parametric amplifier (also based on non-linear frequency conversion) to achieve peak-intensities of $\sim 10^{14}\,\mathrm{W/\,cm^2}$, which is sufficient for high-harmonic generation. The amplifier is pumped by two pulses from a home-built pump laser, of which a specific pair of pump pulses, and therefore also a pair of amplified frequency-comb pulses, can be selected. The amplified frequency-comb pulse pair is up-converted and used for Ramsey-comb spectroscopy.

For the experiments described in Chapter 4, the fourth harmonic was generated in non-linear crystals to excite the two-photon $X - EF$ transition in H_2. A molecular beam was created from a liquid-nitrogen cooled valve and the molecules were excited in a counter-propagating geometry to reduce Doppler effects. The obtained transition frequency of 2 971 234 992 965(73) kHz presents a hundred-fold improvement of the accuracy over the previous determination and has contributed to the most accurate determination of the dissociation energy of H_2.

The experiments described in Chapters 5 and 6 were performed in preparation of $1S - 2S$ spectroscopy in He^+. For this purpose a new 3 m-long vacuum setup was designed and built-up. The main considerations for the design are given in the second part of Chapter 3. The setup consists of seven vacuum chambers, in which high-harmonics are created and refocused in a spectroscopy chamber using a toroidal mirror pair. In the future, an ion trap will be placed in the center of this chamber, where a single He^+ ion will be trapped alongside a single Be^+ ion for sympathetic cooling purposes. The setup was tested by performing Ramsey-comb spectroscopy on the $5p^6 \rightarrow 5p^5 8s\ {}^2[3/2]_1$ transition in xenon at 110 nm (the seventh harmonic of 770 nm). Here, the xenon atoms were excited in an atomic beam at 90° angle to reduce the first-order Doppler shift. The main results of this experiment are given in Chapter 5. They show that the phase shifts from up-conversion with high-harmonic generation are significant on short timescales (up to 1 rad at 8 ns), but disappear almost completely when increasing the pulse delay. Therefore the absolute transition frequency could be calibrated with sub-MHz accuracy, which is unprecedented for spectroscopy with light from a high-harmonic generation source. The obtained tran-

sition frequency of 2 726 086 012 471(630) kHz has a relative accuracy of 2.3×10^{-10} and improves upon the previous determination by a factor 10^4. A more extensive description of the experiment in xenon and the obtained results is given in Chapter 6.

The results from Chapters 5 and 6 show great promise for future measurements of the $1S - 2S$ transition in He^+, as the interaction time will then be much longer (because of the long upper state lifetime of 1.9 ms and the fact that the ion can be trapped for a long time) leading to a much higher accuracy. Chapter 7 gives a brief overview of the status of the He^+ experiment, and an outlook on future improvements to the system to reach a higher spectroscopic accuracy in H_2. These future experiments will enable ppm-level tests of QED in the case of He^+ and searches for physics beyond the standard model.