Precision spectroscopy on the Lyman-α transitions of H and He

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1 Introduction

The development of well-controlled, wavelength-tuneable lasers has had a tremendous impact on atomic and molecular physics. New methods to investigate the properties of atoms and molecules became available, with major applications in the field of metrology. Also exciting new possibilities to manipulate the motion of atoms were introduced. Hänsch and co-workers over the years played, and continue to play a major role in this field, developing new techniques for high-resolution laser spectroscopy and applying them e.g. to ultra-precise studies of the hydrogen atom, as well as initiating innovative research in the field of cold atoms and Bose-Einstein condensation. Already in 1971 Hänsch *et al.* introduced the now well-known technique of laser-saturation spectroscopy in an experiment on the I_2 molecule [1] and the Na-atom [2], later improved and refined with the introduction of various polarization schemes [3, 4]. This method was applied by Hänsch *et al.* to study the Balmer-α line of the H-atom in high resolution [5]. The technique to eliminate first-order Doppler effects in a two-photon excitation process, for the first time experimentally applied to investigate the 3S-5S transition in Na by Biraben *et al.* [6], was subsequently used by Hänsch *et al.* [7] to study the 1S-2S transition in H with a pulsed laser system. This immediately resulted in improved data on Lamb shifts and Rydberg constant and pointed the direction for future work, involving sophisticated CW laser light sources at 243 nm and the use of cooled atoms to reduce second-order Doppler-shifts. Using a highly stable CW laser source at 243 nm in combination with a cooled beam of hydrogen atoms the frequency of the 1S-2S transition has been determined with an accuracy of 1.8 parts in 10^{14} [8]. When this transition frequency is used together with frequencies for other transitions (e.g. 2S-8D measured by Biraben *et al.* [9]) values for Lamb shifts and Rydberg constant with unprecedented accuracies can be established.

In 1975 Hänsch and Schawlow already pointed out that laser radiation pressure could be applied to cool atoms [10], suggesting future applications on cooling atomic hydrogen. This requires a source of, preferably CW, vacuum ultraviolet Lyman-α laser radiation at 121.5 nm, a source that was recently built and applied by Eikema *et al.* in the Hänsch group [11, 12]. This source and its first operation in a study of the Lyman-α 1S-2P transition will be discussed in paragraph 2 of this contribution. In paragraph 3 a similar measurement on the 1 ^{1}S - 2 ^{1}P Lyman-α transition in neutral helium, at 58.4 nm, will be presented [13, 14, 15]. In paragraph 4 new possibilities to measure the 1 ^{1}S - 2 ^{1}S transition in helium in a two-photon experiment with high accuracy will be discussed. The proposed approach is based on recent, exciting developments in the Hänsch group to measure large optical frequency differences with a train of femtosecond laser pulses, recognizing that the regularly spaced comb of frequency modes of a mode-locked femtosecond laser can be used as a precise ruler [16]. With this optical ruler technique not only the most accurate value for the frequency of the 1S-2S transition in atomic hydrogen has been determined [8], but it also made an unprecedented accuracy of 1.1 parts in 10^{14} possible for an optical transition in the mercury ion [17].

2 The 1S - 2P Lyman-α transition in hydrogen

The 1S-2P transition in hydrogen at 121.56 nm is rather special. Apart from being the strongest optical dipole transition in hydrogen, it is also the one required to perform closed-cycle laser cooling from the 1S ground state. Application of laser cooling can reduce the second-order Doppler effect and enhance the number of atoms at low velocities in precision spectroscopy on a beam of hydrogen. It is therefore remarkable that even after almost 40 years of laser spectroscopy, the 1S-2P transition had not been observed up to now with its natural linewidth of 100 MHz (1.6 ns lifetime of the 2P state). There is however a more pressing reason to finally harness the 1S-2P transition: the antihydrogen atom. Creation of antihydrogen seems imminent given the tremendous progress in production and handling of elementary antiparticles. If optical transitions in hydrogen and antihydrogen could be compared to high precision, exciting new tests of CPT and possibly gravitation can be performed. One of the challenges of antihydrogen is its rapid annihilation on contact with normal matter. Non-destructive optical techniques such as laser cooling and optical detection using the 1S-2P transition (assumed to be the same in antihydrogen) will be vital.

To get the most out of 1S-2P laser cooling (Doppler limit 2.4 mK) and spectroscopy, a narrow band continuous source of Lyman-α is required. Although several *pulsed* (broad-band >100 MHz) sources of Lyman-α were demonstrated over the last two decades, not a single coherent *continuous* source was realized. One of the difficulties is that Lyman-α is at the edge of the vacuum ultraviolet (VUV) spectral region. Not only is this radiation readily absorbed in air, optics has to be made from single crystal MgF_2 or LiF material, which typically absorbs 50% of Lyman-α in just a few mm. To produce VUV, high power pulsed lasers have been used to drive third-order nonlinear interactions. Lyman-α can be generated by third harmonic conversion of 365 nm in Krypton, or by frequency mixing using up to three different wavelengths in e.g. mercury. Simply replacing multi-megawatt pulsed lasers by continuous lasers with only a few Watt optical power, would reduce the VUV yield to below a photon per second. For detection and laser cooling of antihydrogen at least 10 nW ($\approx 10^{10}$ ph/s) is required. Strong focusing of the laser beams in the nonlinear medium does improve VUV yield for continuous lasers, but it is not sufficient.

The solution is to choose all wavelengths involved in the mixing process close to, or in coincidence with atomic resonances to enhance the nonlinear susceptibility. Mercury has the appropriate level structure to implement this approach. As can be seen in Fig. 1, the combination of a frequency doubled single-mode argon laser (257 nm), and a frequency-doubled Ti:Sapphire laser (399 nm) can be made exactly resonant with the $7s\ ^1S_0$ state. A dye laser at 545 nm is then sufficient to reach the Lyman-α photon energy. The combination of exact and near-resonances, and the 100% duty-cycle of continuous lasers basically compensates the much lower peak power compared to pulsed lasers. In this way up to 20 nW Lyman-α radiation has been generated in mercury vapor. At a slightly longer wavelength of 122.1 nm, up to 200 nW was produced [12]. Both the short wavelength and the yield are unprecedented for continuous four-wave-mixing in the VUV. As all the fundamental lasers have a bandwidth below a few MHz, the generated VUV radiation has a bandwidth well below 10 MHz. The result is a tuneable

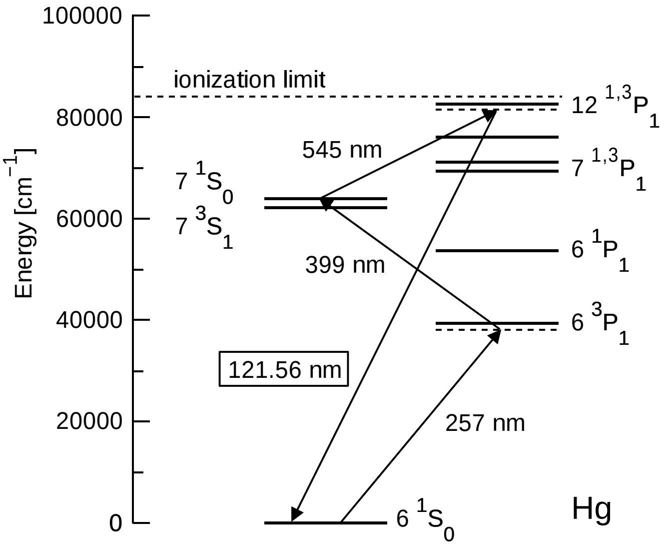

Figure 1: *Four-wave-mixing scheme in mercury for producing continuous coherent Lyman-α.*

source with an average Lyman-α power comparable to the pulsed source previously used for cooling and spectroscopy of magnetically trapped hydrogen [18], but with an almost 2 orders of magnitude narrower bandwidth. Several improvements, such as changing to a pure solid-state laser system or using isotopically pure mercury, could improve the Lyman-α yield even further by at least 1 or 2 orders of magnitude.

To demonstrate the capabilities of this source, and its prospects for antihydrogen, 1S-2P spectroscopy in a beam of normal atomic hydrogen was performed. The natural resonance width is 100 MHz, but in hydrogen already at a transverse temperature of 1K a Doppler broadening of 1 GHz occurs. Therefore a liquid-nitrogen-cooled hydrogen source was used in combination with a narrow slit-skimmer to obtain a beam with a temperature of $\approx$80 K and a divergence of 1:400. This beam intersected the carefully collimated Lyman-α light perpendicularly. Due to the optics in the Lyman-α beam for spatial filtering and collimation about 1-2 nW reached the hydrogen interaction zone. Excitation of the 1S-2P was then monitored by photon counting the resulting fluorescence with a solar-blind photomultiplier. In Fig. 2 the excitation scheme and the experimental result are shown. Most measurements were performed on the $1s\ ^2S_{1/2} - 2p\ ^2P_{3/2}$ transition that can

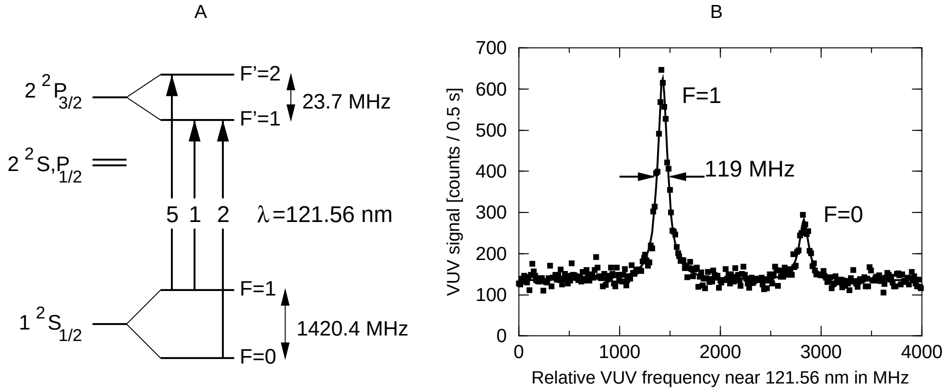

Figure 2: *The* $1s\ ^2S_{1/2} - 2p\ ^2P_{3/2}$ *excitation scheme, and recorded resonance in atomic hydrogen using continuous coherent Lyman-*α*. The numbers in the arrows indicate the relative strengths of the transitions.*

be used for laser cooling. A resonance width of 119(2) MHz was measured, based on 24 recordings. This is the first 1S-2P recording with a near natural linewidth (the excess 19 MHz is ascribed to residual Doppler broadening). The 23.7 MHz hyperfine structure of the $2p\ ^2P_{3/2}$ is less than the natural width of 100 MHz and causes little extra broadening (2 MHz) as mainly the F=1 to F'=2 is excited. It does, however, change the double peak separation between excitation from the F=0 and F=1 by effectively 20 MHz. The resulting theoretical value of 1400.5 MHz is in good agreement with the experimental value of 1396(6) MHz. From the results presented it is clear that the continuous coherent Lyman-α source can play an important role in the future for detection, spectroscopy, and laser cooling of antihydrogen.

3 The 1 ^{1}S - 2 ^{1}P Lyman-α transition in helium

For many years atomic hydrogen was the only atomic system in which QED theory could be tested and this work was primarily pushed by Hänsch and coworkers in Garching. Progress in theoretical calculations also made multi-electron atoms and ions of interest for QED studies. Energy level calculations without QED and higher-order relativistic effects in e.g. helium are nowadays so accurate that experimental transition frequencies can be used as a test for Lamb-shift calculations, just like in atomic hydrogen. Helium

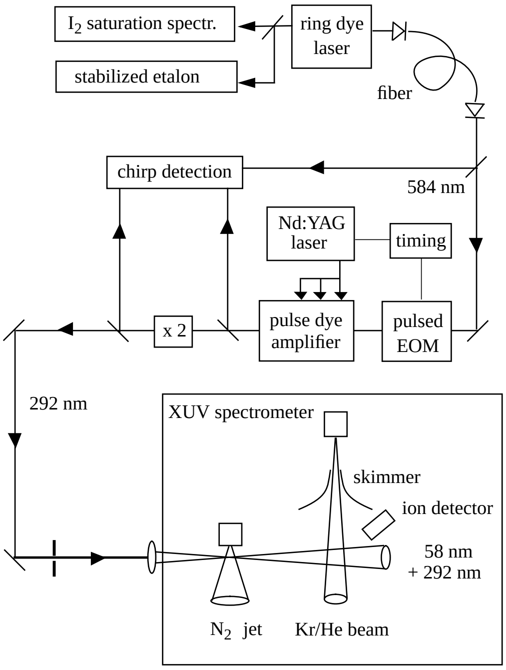

Figure 3: *Overview of the experimental setup to measure the 1* ^1S *Lamb shift in helium.*

is particularly interesting because of two-electron contributions to the Lamb shift. These contributions are best tested in ^1S states where they contribute up to 10% to the total Lamb shift. Of course the Lamb shift is largest in the 1 ^1S ground state. However, the ground state is difficult to access with laser radiation due to the large energy difference with excited states. In 1993 our group in Amsterdam showed in a preliminary experiment on the $1\ ^1S - 2\ ^1P$ transition at 58.4 nm that this energy difference can be bridged using high-power pulsed laser systems and harmonic upconversion [13]. This encouraged us to start a dedicated experiment using a CW dye laser at 584 nm and pulsed amplification techniques. A schematic of the setup is shown in Fig. 3. A pulse-dye-amplifier (PDA), pumped by a Nd:YAG laser, amplifies light at 584 nm from a ring dye laser in 6.5 ns pulses. This output is frequency doubled in a nonlinear crystal and subsequently focused in a pulsed jet of N_2 for fifth harmonic generation. In this process 10^5 - 10^6 photons per pulse at 58.4 nm are generated. To reduce Doppler effects the

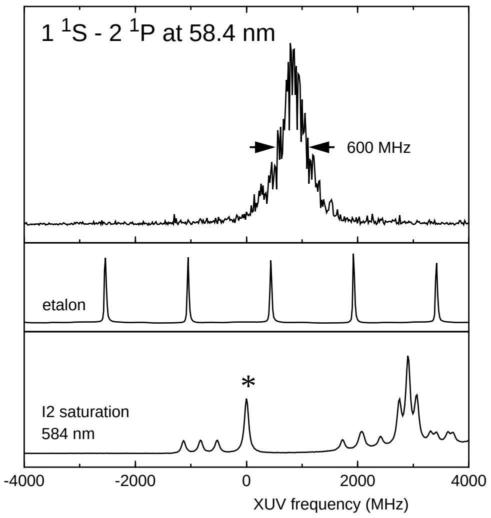

Figure 4: *The* $1\ ^1S - 2\ ^1P$ *resonance transition for anti-chirped PDA pulses.*

$1\ ^1S - 2\ ^1P$ transition is induced in a skimmed and pulsed beam of 10% helium seeded in 90% Krypton in a crossed beam arrangement. A few percent of the atoms excited to $2\ ^1P$ is ionized by the UV light and detected. The primary calibration is performed at 584 nm by saturation spectroscopy on the P88(15-1)-o transition in I_2.

Precision spectroscopy with such a pulsed laser system is hampered by unwanted phase modulation effects in the PDA. Phase modulation results in time-dependent frequency excursions, also called chirp, which lead to a calibration error for the pulsed output relative to the frequency standards based on CW laser saturation spectroscopy in the visible. This chirp is a result of phase modulation due to time-dependent gain in the amplification process. To counteract this chirp a $LiTaO_3$ EOM was developed to modify the phase of the CW seed beam prior to amplification in the PDA. A scan under anti-chirp conditions is shown in Fig. 4. The experimental linewidth is only a factor 2 larger than the natural linewidth of the upper $2\ ^1P$ state, which is 300 MHz. Included in the figure is a saturated absorption spectrum of the relevant I_2 line (calibrated one indicated with an asterisk) as well as an etalon spectrum. The $1\ ^1S - 2\ ^1P$ transition frequency could be determined with an accuracy of 9 parts in 10^9: 5130495083(45) MHz [15]. A Lamb shift for the $1\ ^1S$ ground state of -41224(45) MHz was deduced, in excellent agreement with a theoretical value of 41233 MHz. The accuracy of the theoretical value was recently re-evaluated by Drake [19] leading to an uncertainty in theory of 91 MHz. As in the case of atomic hydrogen, the experimental accuracy is better than the theoretical one.

4 Outlook

The recent use of a mode-locked laser as a frequency ruler is a prime example of the creativity in the Hänsch group. This work is not only important for frequency synthesis and optical interval division. Already in 1978 Hänsch proposed and performed precision spectroscopy using mode-locked pulses from a dye laser to *excite* Na [20]. Although only small intervals could be measured in this experiment (1 GHz), modern modelocked Ti:Sapphire lasers can vastly improve this situation. The principle is that repetitive excitation with phase-controlled pulses results in Ramsey-like resonance features that are much narrower than the bandwidth of the individual optical pulses. This type of spectroscopy does, however, require a better control of the phase reproducibility of the pulses than frequency metrology. In frequency metrology, phase variations can be averaged out by extended counting times as long as no cycles are missed. In the case of direct spectroscopy with pulses, phase noise results in a broadening of the Ramsey resonances. The chirp effect discussed in the previous section can be a significant source of phase distortion. Although this influences the excitation by a single pulse, to first order it merely influences the envelope and not the sharp resonances in multi-pulse spectroscopy. If all sources of phase noise can be sufficiently controlled, then spectroscopy with mode-locked lasers can combine high excitation probability with a high resolution. However, use of short and therefore broad-bandwidth optical pulses limits the Doppler broadening reduction that can be obtained in two-photon spectroscopy. For atoms that are difficult to excite otherwise (e.g. low signals on two-photon transitions with UV or VUV light), but that can be trapped and cooled, this is fortunately not a severe limitation. Antihydrogen ($1S-2S$) and helium ($1s^2\ {}^1S$ - $1s2s\ {}^1S$) are therefore interesting candidates for this type of excitation. Also non-linear optics with ultra short pulses may greatly benefit from the use of phase-controlled pulses. The generation of X-ray pulses with attosecond duration for studies of ultrafast phenomena may be envisioned [21]. Both aspects of phase-controlled optical pulses are currently being investigated in our group.

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