# Tests of Fundamental Physics using Ramsey-Comb Spectroscopy on the Hydrogen Molecule

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High-precision spectroscopy on simple systems such as hydrogen has reached an unprecedented level of accuracy in recent years [1]. Experimentally determined energy levels are used to test bound-state quantum electrodynamics (QED). However, theoretical values for the energy levels were limited by the uncertainty of experimentally determined parameters such as the proton charge radius (r_p). In 2010, the CREMA collaboration performed a spectroscopic measurement on muonic hydrogen. From this, r_p was extracted with a ten times higher accuracy but also showed a 5σ discrepancy to the CODATA-2010 value [2]. This so-called proton radius puzzle remains unsolved. A possible solution to this problem can be obtained from measurements in other systems.

Molecular hydrogen (H_2) is an interesting candidate as its energy structure can be calculated with high precision. We therefore investigate transition frequencies in H_2 to test molecular QED calculations and potentially extract r_p. In particular, a measurement of the dissociation energy D_0 with an accuracy of 10 kHz can be used to probe r_p to 1% . This value can be obtained by combining the ionization potential of H_2 ($E_i(H_2)$), the ionization energy of H and the dissociation energy of H_2^+. The determination of $E_i(H_2)$ consists of a first measurement from the electronic ground state X to an intermediate electronically excited state (GK or EF state) which is performed at the LaserLaB in Amsterdam while the group of Prof. Merkt at the ETH Zurich measures from that intermediate state up to $E_i(H_2)$ through a Rydberg state. Recent results using the GK state yield an accuracy of 750 kHz on D_0 in ortho-H_2 [3]. We measured the $EF^1\Sigma_g^+-X^1\Sigma_g^+(0,0)$ Q_1 transition, corresponding to the excitation from the electronic ground state to the EF state in ortho-H_2, with an accuracy of 73 kHz [4]. This measurement paves the way for a better determination of ortho-D_0 [5]. Furthermore, we aim to measure the Q_0 transition to be able to determine para-D_0. The fundamental ground-tone of H_2 can also be determined experimentally with a predicted level of accuracy around 10 kHz by combining the Q_0 transition with the $EF^1\Sigma_g^+-X^1\Sigma_g^+(0,1)$ transition, where we will excite starting from the first excited vibrational level (Fig.1a).
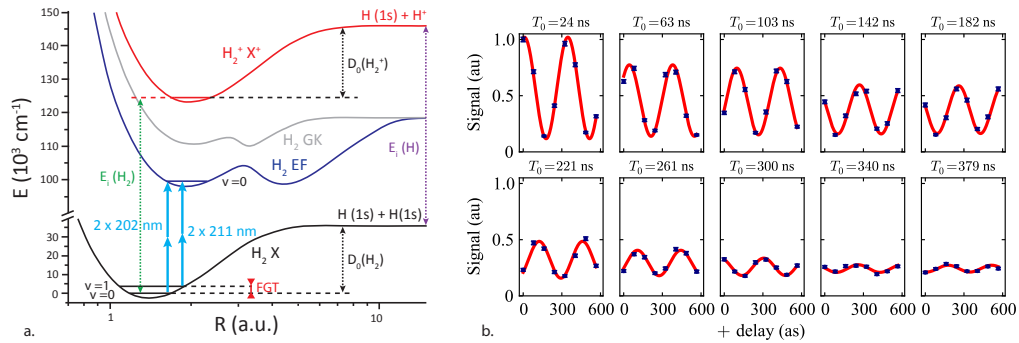
Fig. 1 a. Two measurements must be combined to obtain the fundamental ground tone (FGT). b. The relative phase evolution of this series of Ramsey-fringes is used to extract the transition frequency of the Q1 transition to 73 kHz.

The two-photon transition Q_1 at 202 nm in H_2 has been measured using Ramsey-comb spectroscopy (RCS). For this, different pulse pairs from a frequency-comb (FC) laser at 808 nm are amplified to the mJ-level using an optical parametric chirped pulse amplifier. They are then up-converted to 202 nm and overlapped with the molecular beam in a counter-propagating configuration to reduce the first-order Doppler effect. The excited molecules are then ionized and detected.We obtain a Ramsey-fringe by changing the repetition rate f_{rep} of our FC laser. A series of these fringes is recorded at different interpulse delays corresponding to multiples of $1/f_{rep}(= 8$ ns$)$. We extract the transition frequency by analysing the relative phase evolution of the series of Ramsey-fringes (Fig.1b). Constant systematic shifts like the AC-Stark shift are canceled using this method. We are currently improving our molecular beam and laser system to reach an accuracy of 10 kHz for the previously mentioned transitions.

References

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