Spectroscopy of the molecular ion HD⁺ in the Lamb-Dicke regime: towards determination of fundamental constants at the 10⁻¹⁰ level

Soroosh Alighanbari¹, Florin L. Constantin^{1,2}, Gouri S. Giri¹, Vladimir Korobov³, <u>Stephan Schiller¹</u>

1. Institut für Experimentalphysik, Heinrich-Heine-Universität Düsseldorf, Düsseldorf, Germany

2. Laboratoire PhLAM CNRS UMR 8523, University Lille 1, Villeneuve d'Ascq, France

3. Bogoliubov Laboratory of Theoretical Physics, Joint Institute for Nuclear Research, Dubna, Russia

Precision measurements with cold atoms and molecules allow testing the predictions of quantum electrodynamics, determining fundamental constants, probing their possible time variation, and searching for new fundamental interactions. The effective control of atoms' and molecules' external and internal degrees of freedom paves the way to increased accuracy. Molecular hydrogen ions (MHIs) are three-body quantum systems for which comparison of *ab initio* theory and experiment can provide an independent determination of the Rydberg constant, of the mass ratios of electron to proton and electron to deuteron, and ultimately also of the proton's and deuteron's charge radius. This program is enabled by recent strong advance in *ab initio* theory [1, 2], which has reached $\approx 1 \times 10^{-11}$ inaccuracy.

On the experimental side, the spectroscopy of MHI has so far been limited by Doppler broadening even at the low temperatures (10 mK) achieved using sympathetic cooling, leading to line resolution not better than 5×10^{-7} [3, 4]. Recently, we have for the first time achieved Doppler-free spectroscopy of the fundamental rotational transition of HD⁺ at 1.3 THz, enabled by the transverse confinement of the MHI clusters in the Lamb-Dicke regime [2]. Line resolution of 1×10^{-9} was achieved.

In this contribution we present an up to 300-fold further decrease in linewidth, to the 3×10^{-12} level (Fig. 1, arrow). This line resolution is now better than the theoretical inaccuracy of the *ab initio* prediction. This excellent resolution allows us to probe systematic shifts very sensitively. For example, we determined an upper limit of 1×10^{-11} for the light shift induced by 266 nm radiation on the rotational transition, and also resolved the ultra-small pure rotational Zeeman shift (0.55 kHz/G). To date, we measured 6 hyperfine transitions, including a number of Zeeman components (Fig. 1). Our resolution of the hyperfine structure improves on the best previous measurements of any MHI by a factor of 10 [5].

With the data obtained we are able to test the recently improved theory of the spin structure of the molecular ion [6] at the 0.1 kHz uncertainty level. We are also currently analysing the data towards determining the fundamental constant $R_{00}m_e(m^++m^+)$ with a goal uncertainty on the low-10⁻¹⁰ level. Our value can then be

compared with the combined results from atomic hydrogen spectroscopy and mass spectrometry in Penning traps [7, 8]. The results of the analysis will be presented at the conference.

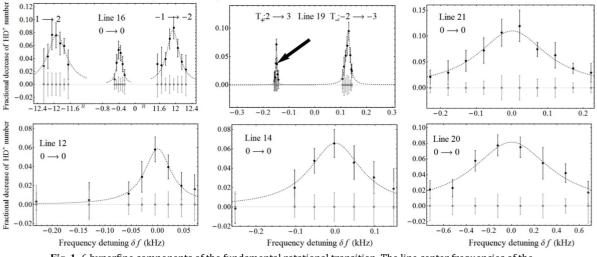


Fig. 1. 6 hyperfine components of the fundamental rotational transition. The line center frequencies of the components differ by values of the order 10 MHz. The arrow indicates an ultranarrow Zeeman component (4 Hz FWHM).

References

 V. Korobov, et al., Fundamental Transitions and Ionization Energies of the Hydrogen Molecular Ions with Few ppt Uncertainty, Phys. Rev. Lett. 118, 233001 (2017).

[2] S. Alighanbari, et al., Rotational spectroscopy of cold and trapped molecular ions in the Lamb-Dicke regime, Nat. Phys. 14, 555 (2018).

[3] U. Bressel, et al., Manipulation of Individual Hyperfine States in HD⁺ and Frequency Metrology, Phys. Rev. Lett. **108**, 183003 (2012).

[4] J. Biesheuvel, et al., Probing QED and fundamental constants through laser spectroscopy of HD⁺, Nat. Commun. 7, 10385 (2016).

[5] K.B. Jefferts, *Hyperfine structure in the molecular ion* H_2^+ , Phys. Rev. Lett. 23, 1476 (1969).

[6] V. Korobov, et al., Theoretical Hyperfine Structure of the Molecular Hydrogen Ion at 1 ppm Level, Phys. Rev. Lett. 116, 053003 (2016).

[7] F. Heiße et al., High Precision Measurement of the Proton's Atomic Mass, Phys. Rev. Lett. 119, 033001 (2017).

[8] S. L. Zafonte and R. S. Van Dyck Jr, Ultra-precise single-ion atomic mass measurements on D and He-3, Metrologia 52, 280 (2015).