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Improved Determination of Fundamental Constants and Test of Fundamental Physics with Doppler-Free THz Spectroscopy of HD⁺

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Abstract: Improved precision of Doppler-free rotational spectroscopy of trapped and laser-cooled HD⁺ ions allows to confirm accurately high-precision ab-initio molecular ion quantum theory calculations and to determine fundamental constants more precisely than the CODATA2018 values.

The understanding of the quantum nature of atoms and molecules may be tested by comparing accurate theoretical ab-initio predictions with the results of precision spectroscopy experiments. A comparison based on the molecular hydrogen ions, the simplest three-body molecular systems, may be exploited also to determine the values of a set of fundamental constants and to search for any deviation of the nucleus-nucleus interaction from the Coulomb force. Achieving improvements in these results compared to the state-of-the-art is a challenging task on both experimental and theoretical sides.

The precision and resolution in rotational and vibrational spectroscopy of the molecular hydrogen ions, limited in the past by Doppler broadening and unresolved hyperfine structure of the spectral lines [1,2,3], has recently been improved using Doppler-free approaches [4,5]. On the theory side, the precision of the ab-initio predictions of the rovibrational energies has reached 10^{-11} or better [6], a level beyond the accuracy of the electron, proton and deuteron masses provided by the CODATA2018 adjustment [7].

We measured hyperfine components of the fundamental rotational transition of HD⁺ at 1.3 THz on sympathetically and rotationally cooled molecular ions in a linear radiofrequency trap [8]. An electronic THz source locked to a GPS-referenced hydrogen maser, an improved state-selective detection and a stable ion trap apparatus allowed reaching a fractional resolution of 3×10^{-12} (Fig. 1(a)). After correction for the systematic frequency shifts induced by the Zeeman effect and the quadratic Stark effect due to the trapping electric field the experimental frequencies were determined with uncertainties as low as 17 Hz. The theoretical frequency uncertainty is dominated by the spin energy errors, ranging from 0.2 kHz to 1.4 kHz [8].

The prediction of the spin structure theory at order α^2 for a hyperfine splitting is confirmed within 4×10^{-5} fractional theory uncertainty, while the fractional experimental uncertainty is 20 times smaller. Exploiting a composite frequency, defined as a suitable linear combination of hyperfine frequencies that cancel the spin theory uncertainty, confirms the quantum electrodynamics calculation at order α^6 with a total fractional uncertainty of 4.8×10^{-11} limited by the CODATA2018 uncertainties. This agreement allows to improve by 20 times the constraint on a hypothetical spin-independent fifth force between proton and deuteron with range >1 Å. Assuming absence of such a force, our result determines the value of $R_\infty m_e (1/m_p + 1/m_d)$ with an uncertainty

2.4 times improved compared to CODATA2018 (Fig. 1(b)). Combining our result with Penning trap mass measurements in other groups, we deduce values for m_e/m_p and m_p with 2.0×10^{-11} fractional uncertainties, 3 times smaller than and comparable with the CODATA2018 values, respectively.

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Figure caption

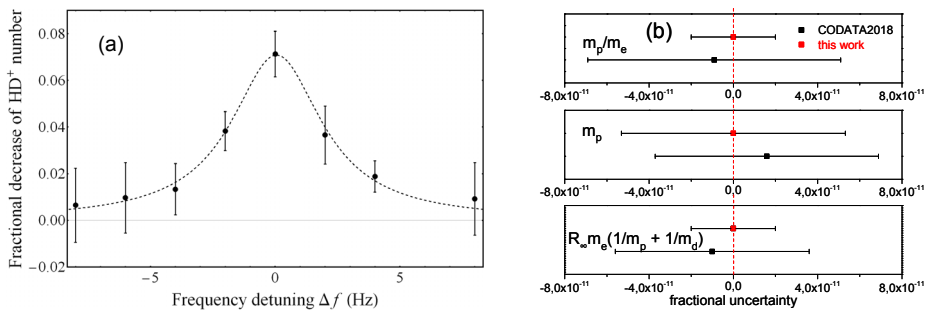


Fig. 1 (a) Rotational spectroscopy of the transition $(v,N,G_1,G_2,F,m_F)=(0,0,1,2,2,2) \rightarrow (0,1,1,2,3,3)$ of HD^+ .
 (b) Determination of fundamental constants from theory-experiment comparison.