Frequency Metrology of the HD + Molecule: Determining Fundamental Constants and Searching for Physics beyond the Standard Model

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Summary : We present high-precision transition frequencies of the hydrogen deuteride molecular ion HD^+, obtained with our novel technique for one-photon spectroscopy in the Lamb-Dicke regime. The comparison with our high-precision *ab initio* calculations of the energy levels, yields agreement at the level 3×10^{-11}. Taking the experimental uncertainty and the uncertainty of the theory prediction into account we could deduce a value of the ratio of electron mass to reduced nuclear mass with accuracy better than direct mass measurements in Penning traps. Alternatively, we determine an upper bound for the strength of a hypothetical spin-independent fifth force between proton and deuteron that is twenty times more stringent compared to previous bounds from HD^+ spectroscopy.

Motivation

Frequency metrology of cold trapped molecules provides exciting opportunities in fundamental physics, including test of *ab initio* calculations (in particular of QED effects), determining fundamental constants, and search for physics beyond the standard model. The HD^+ molecule is a simple bound three-body quantum system that allows a high-precision calculation of rovibrational energy, including relativistic and QED corrections - as of today of up to order α^6 relative to the nonrelativistic energy. Since the theoretical calculation requires as input the values of fundamental constants (proton-electron mass ratio $m_{\mathrm{p}}/m_{\mathrm{e}}$, deuteron-electron mass ratio $m_{\mathrm{d}}/m_{\mathrm{e}}$, proton and deuteron charge radii r_{p} and r_{d}, Rydberg constant R_∞), a comparison of theoretical with experimental transition frequencies provides a means to determine these constants. Also, the level of agreement between theory and experiment sets an upper bound on a hypothetical non-Coulomb proton–deuteron spin-averaged interaction expressed in the form of a Yukawa potential.

Results

We showed that for a string of molecular ions, trapped in a linear ion trap and sympathetically cooled, excitation of transitions with radiation propagating perpendicularly to the trap axis allows spectroscopy in the Lamb-Dicke regime[1]. We have performed spectroscopy of the fundamental rotational[1] (1.3 THz) and the fundamental vibrational[2] (5.1 μm) transitions in the ground electronic level (see figure 1). We measured 6 hyperfine transitions of the fundamental rotational transition and two hyperfine components of the fundamental vibrational transition. Applying a suitable analysis technique, the comparison between experiment and theory results permitted determining a value of $m_{\mathrm{e}}\left(m_{\mathrm{p}}^{-1}+m_{\mathrm{d}}^{-1}\right)$ with uncertainty smaller than CODATA2018 (see figure 2). We could also deduce a value of the proton-electron mass ratio, which is in excellent agreement with the ratio of the directly measured masses.

Finally, our experiment also sets improved upper bounds for the strength of a hypothetical "fifth force" between proton and deuteron (see figure 3).

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[1] S. Alighanbari, et al. "Precise test of quantum electrodynamics and determination of fundamental constants with HD^+ ions", Nature, vol. 581, p. 152-158, 2020.

[2] I.V. Kortunov, et al. "Proton-electron mass ratio by high-resolution optical spectroscopy of ion ensembles in the resolved-carrier regime", Nat. Phys., 2021.

Figure captions :

Fig. 1: Low-lying rovibrational levels of HD^+, and transitions investigated in this work. The vibrational and rotational quantum numbers are denoted by v and N, respectively.

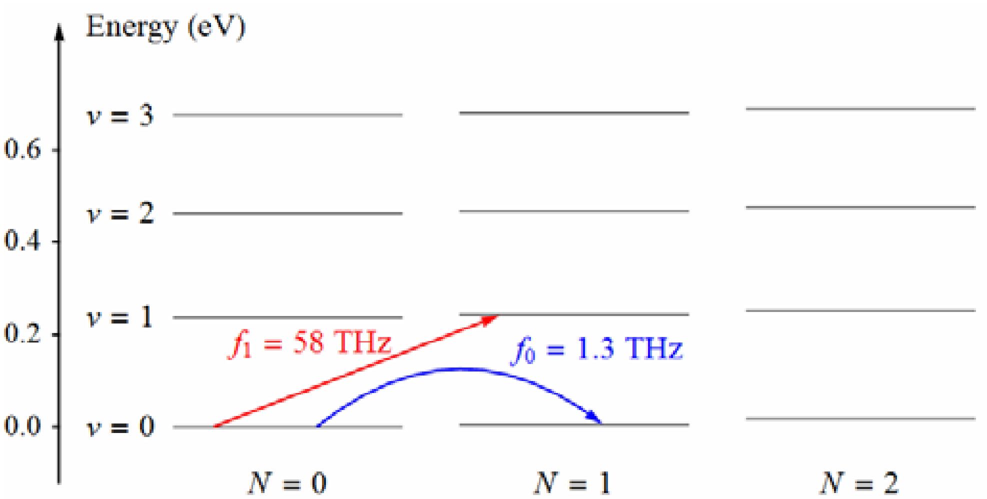

Fig. 2: Electron to reduced-nuclear mass ratio measured in this work and comparison with the recent Penning trap measurement and the CODATA values.

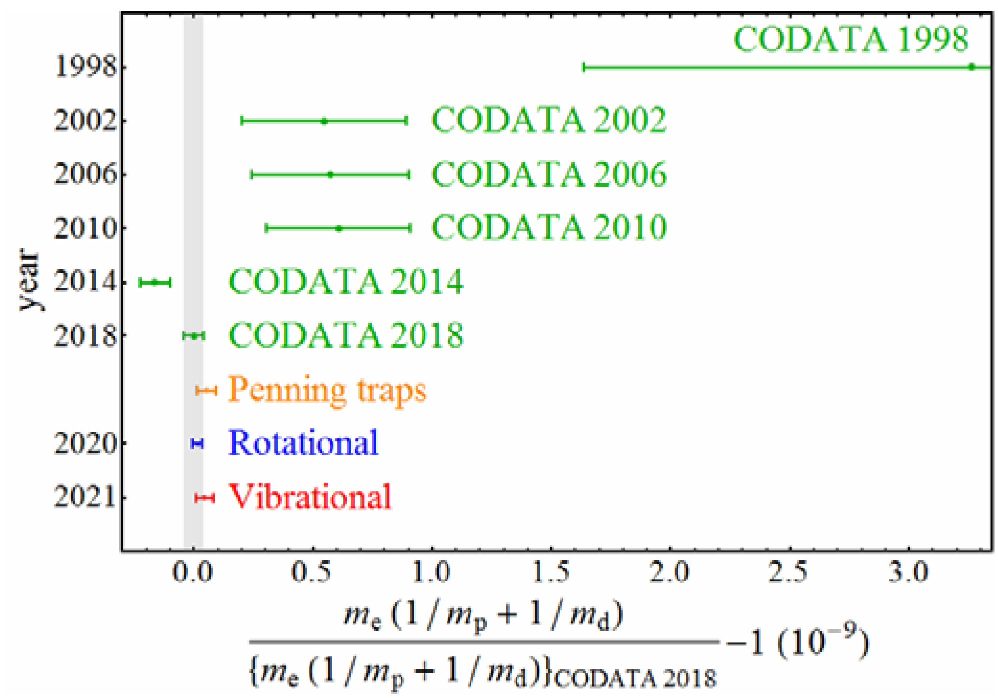

Fig. 3: Experimental constraints on a hypothetical Yukawa-type spin-independent force between proton and deuteron as a function of the assumed range λ of the interaction. The green traces are the results of this work.