

DAMOP13-2013-000026

Abstract for an Invited Paper
for the DAMOP13 Meeting of
the American Physical Society

High-resolution spectroscopy of cold HD⁺ molecular ions

STEPHAN SCHILLER, Heinrich-Heine-Universität Düsseldorf

HD⁺ is a fundamental quantum system: it is a three-body bound quantum system that can be accurately described ab initio by Quantum Electrodynamics, using as input certain fundamental constants. A comparison between experimental HD⁺ transition frequencies and the ab initio results therefore provides a test of the validity of theoretical treatments, and/or a determination of these fundamental constants. At present, the experimental inaccuracies of the transition frequency measurements is still higher than the theoretical or fundamental constants inaccuracies, resulting in an on-going experimental challenge. Many applications of cold molecular ions have been proposed. They would benefit strongly from availability of advanced manipulation techniques, already standard in atomic physics. These are not straightforward for molecules, and for charged molecules have not yet been demonstrated [1]. In this respect, trapped cold HD⁺ is also a useful model system. We performed THz and laser spectroscopy as well as quantum state manipulation of this molecular ion species. The ions are trapped in an ion trap and sympathetically cooled by laser-cooled atomic ions (Be⁺) in order to reduce spectroscopic line broadening. A novel frequency-comb-based, continuous-wave 5 μm laser spectrometer was employed and spectroscopy at the Doppler-limit was performed [2]. To our knowledge, the achieved spectral resolution is the highest obtained so far in the optical domain on a molecular ion species. We were thus able to optically resolve the hyperfine structure. We found agreement of the measured absolute transition frequencies and of the hyperfine splittings with ab-initio theory, the experimental inaccuracy being up to approx. $1 \cdot 10^{-9}$ [3]. This work also represents the most precise test yet of the ab-initio theory of any molecule. We demonstrated addressing of individual hyperfine states of ro-vibrational levels by excitation of individual hyperfine transitions, and controlled transfer of population into a selected hyperfine state [3]. We also report on the first observation of the fundamental pure rotational transition in this molecule [4] and on ongoing developments towards more complete manipulation of the hyperfine states.

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[4] J. Shen, A. Borodin, M. Hansen, and S. Schiller, Phys. Rev. A 85, 032519 (2012).