DETERMINATION OF ROVIBRATIONAL INTERVALS IN H_2^+ WITH SUB-MHZ ACCURACY

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H_2^+ is the simplest of all molecules and as such an important system for the development of molecular quantum mechanics. The rovibrational energy-level structure of this one-electron system can be calculated extremely precisely by quantum-chemical methods[a]. By comparison with the results of precise spectroscopic measurements of rovibrational intervals, fundamental constants or particle properties, such as the proton-to-electron mass ratio or the proton size, can be determined[b]. Because the rotational and vibrational transitions of H_2^+ are electric-dipole forbidden, the experimental data on its energy-level structure are limited.

We present the determination of spin-rovibrational intervals in H_2^+ from high-resolution measurements of the Rydberg spectrum of H_2 and Rydberg-series extrapolation using multichannel quantum defect theory[c]. Choosing suitable double-well valence states of H_2, characterized by long lifetimes and favorable Franck-Condon factors to different vibrational states in the ion, allows us to excite Rydberg states that converge on selected rovibrational levels of H_2^+.

For the excitation of Rydberg states, a resonant three-photon excitation scheme was employed, using pulsed VUV and VIS laser sources to reach the intermediate valence state and a continuous-wave (cw) near-infrared laser source for the excitation to the Rydberg states. The valence state - Rydberg state intervals could be measured with a relative accuracy of 3E-10 using an optical frequency comb for the frequency calibration of the cw laser and minimizing systematic uncertainties[d].

[a] V. I. Korobov, L. Hilico, and J.-Ph. Karr, Phys. Rev. A 89, 032511 (2014)

[b] J.-Ph. Karr, L. Hilico, J. C. J. Koelemeij, and V. I. Korobov, Phys. Rev. A 94, 050501(R) (2016)

[c] D. Sprecher, Ch. Jungen and F. Merkt, J. Chem. Phys. 140, 104303:1-18 (2014)

[d] M. Beyer, N. Hölsch, J. A. Agner, J. Deiglmayr, H. Schmutz, and F. Merkt, Phys. Rev. A 97, 012501 (2018)