

Invited talk

Precision spectroscopy of few-body atoms and molecules

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High-resolution spectroscopic measurements in few-electron atoms and molecules are increasingly used as a means to test the foundations of the theory of atomic and molecular structure. Modern first-principles calculations of the energy-level structure of few-electron atomic and molecular systems consider all interactions in the realm of the standard model of particle physics.^{1,2,3,4} Systematic comparisons of the results of such calculations with precise spectroscopic measurements in simple atoms and molecules such as H, He, H₂⁺, H₂ and He₂⁺ aim at searching for effects not yet included in the theory (see, e.g., Refs.^{5,6}) and at reducing the uncertainties of physical constants (see e.g., Refs.^{1,7,8}).

This talk will present precision spectroscopic measurements of transitions to high Rydberg states of H, He, and H₂, which we use to determine accurate values of their ionization energies and, in the case of H₂, also of the spin-rovibrational energy-level structure of H₂⁺. The talk will describe our experimental strategy to overcome limitations in the precision and accuracy of the measurements originating from the Doppler effect, the Stark effect, and the laser-frequency calibration. The experimental results will then be compared with the results of first-principles calculations that include the treatment of finite-nuclear-size effects and relativistic and quantum-electrodynamics corrections up to high order in the fine-structure constant. Recent aspects of these investigations include a new determination of the Rydberg constant⁹ as a contribution to the resolution of the proton-size puzzle¹⁰, a new method to record Doppler-free single-photon excitation spectra in the visible and UV spectral ranges¹¹ (see Figure 1), a “zero-quantum-defect” method to determine the energy-level structure of homonuclear diatomic molecular ions such as H₂⁺,¹² and a 9σ discrepancy between theory and experiment in the ionization energies of metastable (1s2s ³S₁) ³He and ⁴He.^{3,13}

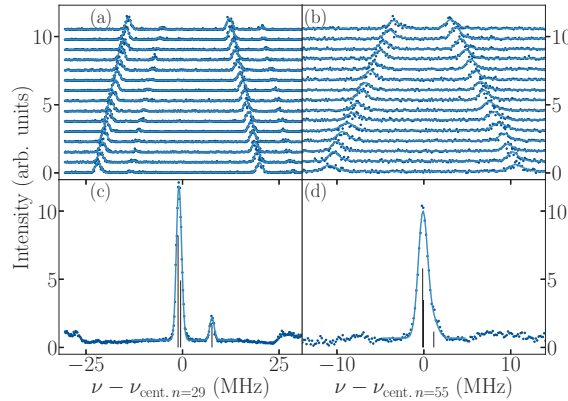


Figure 1: Imaging-assisted Doppler-free spectra of the (a) $(1s)(29p) {}^3P_J \leftarrow (1s)(2s) {}^3S_1$ and (b) $(1s)(55p) {}^3P_J \leftarrow (1s)(2s) {}^3S_1$ transitions in ⁴He in a supersonic beam, obtained by spatial imaging of atoms with different transverse-velocity components in the atomic beam (dots) and fits to the experimental spectra (solid lines). The photoexcitation laser beam is retroreflected so that each transition is recorded as a pair of lines with opposite Doppler shifts. (c) and (d): Sums of the corresponding cross-correlation spectra used to extract the first-order-Doppler-free line positions. The stick spectra give the positions of the $J = 2, 1$ and 0 fine-structure components in order of increasing frequency and the full line represents the spectrum calculated using the line-shape parameters determined in the least-squares fits. From Ref.¹⁴

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