

Abstract number: O21

Monday 10:00-10:20

Oral

Coherent Raman Metrology of Molecular Hydrogen: application to the Q(1) 1-0 line

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We introduce a comb-calibrated spectrometer for frequency metrology of Raman-active transitions and thus of quadrupole transitions. It may operate in a two-octave spanning range, from 50 to 5000 cm⁻¹, covering all fundamental rovibrational bands as well as purely rotational bands. The spectrometer exploits a stimulated-Raman-scattering interaction of the gas target with two comb-calibrated near-infrared cw lasers in a multi-pass cell. We demonstrate its metrological quality by measuring the transition frequency of the most famous line of molecular hydrogen, namely the Q(1) 1-0 line at 4155 cm⁻¹, with a few-parts-per-billion accuracy of 1×10⁻⁵ cm⁻¹. This improves by an order of magnitude the experimental state of the art obtained by Resonantly-Enhanced Multiphoton-Ionization¹ and overcomes by a factor of about 2 the current theoretical benchmark obtained by initio-calculations².

The Raman spectra are acquired over a 2-decades-spanning pressure range, from 40 mbar to 4 bar, and globally fitted with a β -corrected Hartmann-Tran profile to retrieve the zero-pressure line center ν_0 . The fitting uses a properly chosen set of collisional parameters fixed against ab-initio values inferred from quantum-scattering calculations on H₂-H₂ collisions³. To mitigate the impact on ν_0 of an inadequate modeling over such a large range of relatively high pressures we investigate how the inferred ν_0 modifies upon restricting the fit to experimental data at lower and lower pressures (i.e., progressively discarding high-pressure spectra). Interestingly, the analysis of ν_0 as a function of the maximum pressure used in the global fit for a variety of conditions - namely linear pressure shifts and ab-initio parameters varied in their confidence intervals - returns a family of converging trajectories that define ν_0 with a systematic uncertainty as low as 235 kHz. The overall uncertainty, summing up in quadrature a statistical error of 205 kHz, is 310 kHz. The experimental line center agrees with theory within 1- σ . Prospectively, further reducing both experimental and theoretical uncertainties, highly stringent tests of molecular quantum electrodynamics can be performed in a robust way with this highly versatile spectrometer on a large number of transitions.

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