SPECTROSCOPIC-NETWORK-ASSISTED PRECISION SPECTROSCOPY AND ITS APPLICATION TO WATER: THE EXPERIMENT

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Frequency combs and cavity-enhanced optical techniques have revolutionized molecular spectroscopy: their combination allows recording saturated Doppler-free lines with ultrahigh precision. Despite these advancements, precision spectroscopy has not been employed systematically to improve the quality of comprehensive spectroscopic databases as individual observations are often time consuming and lacks impact.

Here we present a new spectroscopic method using network theory, based on the generalized Ritz principle, which offers a powerful tool for the intelligent design and validation of precision-spectroscopy experiments and the subsequent derivation of accurate energy differences.

As a proof of concept, 351 carefully-selected near-infrared transitions (at $1.4 \mu m$) are detected for both $H_2^{16}O^a$ and $H_2^{18}O^b$, two benchmark species of molecular spectroscopy. Transition frequencies of the Lamb-dips are measured up to kHz accuracy using our frequency comb referenced NICE-OHMS spectrometer. These measurements, augmented with extremely-accurate literature lines to ensure overall connectivity, allow the precise determination of the lowest *ortho*-energy and 348 energy levels of both isotopes with unprecedented accuracy. Based on the limited number of observed transitions, 2765 calibration-quality lines are obtained in a wide wavenumber interval. These can be used to improve spectroscopic information systems and applied to frequency metrology, astrophysics, atmospheric sensing, and combustion chemistry.

^aR. Tóbiás et al., Spectroscopic-network-assisted precision spectroscopy and its application to water, Nature Comm. 11, 1708 (2020)

^bM.L. Diouf *et al.*, Network-based design of near-infrared Lamb-dip experiments and determination of pure rotational energies of H₂¹⁸O at kHz accuracy, in preparation (2021)