

UV Dual Comb Spectroscopy – Ultra-High Resolution Molecular Spectra in Milliseconds

E.A. Ehl¹

¹ Institute of Experimental Physics, Graz University of Technology

Dual comb spectroscopy (DCS) is a novel method that enables the measurement of atomic and molecular spectra at ultra-high spectral (MHz - GHz) and temporal resolution (fs pulses) with measurement times of 500 ms to a few seconds. The principle of DCS is based on the difference in repetition rate between two laser frequency combs generating a beat note that down-converts the optical signals into the radio regime, enabling detection with standard photodiodes. Spectral information is then reconstructed via fast Fourier transformation (FFT). This method offers high stability due to its lack of moving parts, rapid acquisition times, broad spectral coverage, and precise wavelength accuracy. Recent advancements have expanded this method into the UV regime using nonlinear frequency-upconversion, allowing access to not only rotational and vibrational, but also electronic transitions.

The UV regime is critical for the investigation of exoplanet atmospheres, because it drives atmospheric chemistry through photodissociation and provides an unique view into the upper layers of the atmosphere, enabling the detection of trace gases that are difficult to detect in the IR. Investigating this by atmospheric modelling requires precise knowledge of both reaction rates and spectral signatures to be able to predict the abundances and spectral detectability of atmospheric constituents. Many key species for atmospheric chemistry, including potential biosignatures, are expected to be present in such atmospheres only in trace amounts or have weak spectral features in the IR. One such example, NO₂, is difficult to observe in the MIR due to being overshadowed by H₂O. Here, the UV/VIS regime offers a promising alternative due to higher absorption cross-sections and less interference. With upcoming missions like the habitable worlds observatory (HWO), such molecules may be detected.

Analysing observed spectra, accurately simulating expected absorption and modelling their atmospheric chemistry require expansive molecular databases at a wide range of temperatures. Current databases in the UV regime are often incomplete or lack high resolution data, particularly at higher temperatures. Simulated spectra derived from theoretical line list calculations come with significant uncertainties, especially in the absence of high-quality reference data. This method was used to investigate formaldehyde (HCHO) in the NUV region around 350 nm with a resolution of 1 GHz. We were able to detect over 100 previously unresolved lines, with the measurements being in excellent agreement with ab-initio calculations. Subsequently, benzene (C₆H₆) and SO₂ were studied at 262 nm. Like with formaldehyde, we were able to resolve previously unseen weak lines of benzene. All of these measurements are comparable to or even improve existing state-of-the-art literature with the spectral resolution being Doppler-limited. Good SNR can be achieved with measurement times from a few seconds to as low as 500 ms. By providing high-precision laboratory measurements, DCS helps refine line positions, intensities, and broadening parameters, improving databases such as HITRAN, ExoMol, and UV spectral atlases. Our high resolution data provides experimental validation to existing line lists and can also provide a basis for new models.