COMB-ASSISTED QCL DOPPLER-BROADENED THERMOMETRY IN NH_3 SAMPLES

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The thermodynamic temperature of a gaseous medium at thermodynamic equilibrium can be extracted from molecular spectra by examining: the relative intensities of rotational structures, the relative intensity of vibrational satellites, or the Doppler width of individual rovibrational lines. Of these methods, Doppler broadening thermometry (DBT), measuring the Doppler width of a single, well-isolated absorption profile, represents a primary method providing the most accurate optical determinations. DBT requires highly precise, accurate and repeatable frequency scanning of a narrow-linewidth probe laser around a given center frequency. This requirement often calls for complicated sideband-based approaches or frequency-locking techniques.

We describe here an elegant implementation of Doppler broadening thermometry in which a room-temperature continuous-wave quantum cascade laser at 9.07 μm is coherently phase-locked to a thulium frequency comb centered at 2 μm. Repeated tuning of the repetition-rate enables wide and highly accurate frequency scans throughout multiple rovibrational lines of ammonia. Hence, the thermodynamic temperature is retrieved from a manifold of profiles, rather than from a single, isolated line, taking advantage of the simple scaling law that relates Doppler width and line-center frequency. This is realized, regardless of the complexity of the adopted lineshape model, by implementing a multiple-line fitting procedure with some relevant physical constraints.

The precision attained for the retrieved gas temperature, even in the absence of an accurate control of the thermodynamic conditions of the ammonia sample, is as high as 50 ppm, even from an analysis of a restricted number of spectra ($\sim$100), acquired at different gas pressures. This represents a particularly relevant result as compared to previous DBT based upon Doppler-width retrievals from an isolated spectral line.