

Optical Frequency Comb Fourier Transform Spectroscopy with Resolution Beyond the Path Difference Limit

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Fourier transform spectrometers (FTS) based on optical frequency combs (OFC) allow detection of broadband molecular spectra with high signal-to-noise ratios within acquisition times orders of magnitude shorter than traditional FTIRs based on thermal sources^[1]. Moreover, high absorption sensitivity is obtained using optical enhancement cavities^[2]. Due to the pulsed nature of OFCs the interferogram consists of a series of bursts rather than a single burst at zero optical path difference (OPD). The comb mode structure can be resolved by acquiring multiple bursts in both mechanical FTS systems^[3] and dual-comb spectroscopy^[4]. However, in all existing demonstrations the resolution was limited either by the maximum OPD between the interferometer arms or by the total acquisition time. We show that it is sufficient to acquire an interferogram in a symmetric range around a single burst with length precisely matched to the comb line spacing in order to exceed the spectrometer's OPD-limited resolution and measure the intensity change of the individual comb lines. Our method allows measurements of broadband spectra with absorption lines narrower than the OPD-limited resolution without any influence of the instrumental lineshape function, and reduces the acquisition time and interferometer length by orders of magnitude. We demonstrate this by measuring undistorted low pressure CO₂ and CO absorption lines with linewidths narrower than the OPD-limited resolution using OFC-based mechanical FTS in the near- and mid-infrared wavelength ranges. The near-infrared system is based on an Er: fiber femtosecond laser locked to a high finesse cavity, while the mid-infrared system is based on a Tm: fiber-laser-pumped optical parametric oscillator coupled to a multi-pass cell.

References

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