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Molecular Spectroscopy with Frequency Combs

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Pulsed femtosecond frequency combs are rapidly developing as a powerful spectroscopic tool. As a spectroscopic source stabilized frequency combs potentially offer broad spectral coverage, near perfect frequency accuracy, low timing jitter and broadband compatibility with resonant cavities. This talk will focus on the first three advantages in a dual comb spectroscopic technique that is highly analogous to traditional Fourier transform spectroscopy. In the dual comb approach, (pioneered in the THz by Keilmann, Van der Weide and coworkers under the name multi-heterodyne spectroscopy), one comb is used to sample a gas and a second frequency comb serves as a local oscillator (LO) that samples the first comb. The LO is held at a slightly different repetition rate than the first comb. When viewed in the time domain, the comb sources each emit a train of pulses. With the difference in repetition rates, for each successive pair of pulses, the timing between the sample and LO laser pulses shifts slightly. Through successive measurements, the LO pulses read out the entire time domain structure of the transmitted sample pulse. Through a Fourier transform, we recover the broadband, complex, absorption profile of the sample gas. In analogy to a Fourier transform spectrometer the LO serves as a scanning interferometer arm. The removal of moving parts from the system along with the addition of high brightness collimated sources brings new flexibility to FTIR spectroscopy. This talk will focus on strengths and limitations of the dual comb technique. Specifically we focus on comb stabilization techniques that allow for long averaging periods, firmware based averaging techniques that keep data sizes manageable and allow for realtime data processing, time domain multiplexing of signal and reference data for continuous removal of system drift, and difference frequency generation techniques to extend this system into the mid IR. We will also discuss methods to improve the sensitivity of this technique as well as tradeoffs between sensitivity and resolution, while maintaining the inherent frequency accuracy of this system.