ULTRAFAST SPECTROSCOPY WITH FREQUENCY COMBS: ENABLING NEW MEASUREMENTS OF DILUTE SPECIES IN MOLECULAR BEAMS

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Initially developed as a tool for metrology, frequency combs are most often used for precision, frequency-resolved spectroscopy. The utility of frequency combs in ultrafast spectroscopy is just beginning to be explored. By exploiting the properties of frequency combs, we are improving the sensitivity, spectral resolution, and detection of ultrafast spectroscopies. The first technique discussed will be cavity-enhanced transient absorption spectroscopy, which uses fiber-laser frequency combs coupled to external enhancement cavities to increase the sensitivity of ultrafast transient absorption spectroscopy. A home-built Ytterbium fiber-laser frequency comb and amplifier system provide a stable source of ultrafast pulses. External enhancement cavities increase both the laser power and effective absorption path length, thus improving the signal by several orders of magnitude over traditional transient absorption spectroscopy. Altogether the sensitivity is more than four orders-of-magnitude better than the previous best transient absorption techniques, which allows for the study dilute samples in molecular beams on the femtosecond timescale with transient absorption spectroscopy. The second technique discussed will be the application of cavity-enhancement and frequency-comb techniques, including dual-comb spectroscopy, to two-dimensional spectroscopy. Initial results and current progress towards ultrafast two-dimensional spectroscopy of dilute species in molecular beams will be presented.