

ULTRAFAST 2D SPECTROSCOPY WITH FREQUENCY COMBS: TOWARDS CAVITY-ENHANCED MULTIDIMENSIONAL SPECTROSCOPY IN MOLECULAR BEAMS

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Multidimensional spectroscopy has been shown to be a powerful tool to study dynamics of complex systems in the condensed phase. However, 2D spectroscopy in the gas phase, specifically of dilute species in molecular beams, has yet to be realized. There are many complex systems, such as small clusters or transient intermediates, for which the added information from 2D spectroscopy would aid in the understanding of structures and dynamics. We use the unique properties of frequency comb lasers to improve multidimensional spectroscopy with the goal of ultrafast, 2D-spectroscopy of dilute species in molecular beams. First, we are creating a novel 2D spectrometer utilizing a homebuilt Yb-fiber frequency comb laser and an electro-optic modulator-based frequency comb. Inspired by dual-comb spectroscopy, this converts the signal from optical to radio frequencies via heterodyne detection and eliminates the need for a traditional spectrometer. A second benefit of using frequency comb lasers is that the ultrafast pulses can be coupled into enhancement cavities, greatly increasing the sensitivity of the technique. By improving the sensitivity, ultrafast 2D spectroscopy of dilute species in molecular beams will be possible for the first time. Current progress towards cavity-enhanced 2D spectroscopy will be discussed.