LOCKING FREQUENCY COMBS TO OPTICAL CAVITIES FOR SIGNAL ENHANCEMENT OF TWO-DIMENSIONAL SPECTROSCOPY

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Ultrafast Two-Dimensional (2D) Spectroscopy is a powerful technique that has provided valuable insight into diverse systems from protein folding to isomerization of metal complexes. We aim to bring this technique to the realm of small molecules in molecular beams. 2D spectroscopy is unique because it allows for the direct measurement of coupled excitations, represented by off diagonal peaks within the spectra. However, because 2D spectroscopy is a third-order process, the signals are necessarily weak, requiring a combination of higher laser power and concentrated sample to obtain detectable signals. To increase the sensitivity of this technique, our lab built a frequency comb laser and coupled it to an external enhancement cavity. This combination of a frequency comb and optical cavity allows for Cavity Enhanced 2D Spectroscopy to be performed with broadband ultrafast pulses. Even with a modest cavity finesse, the resonant cavity pulses see a 2-3 order increase in their electric fields, greatly increasing the sensitivity of the 2D experiment. We explore the use of enhancement cavities on 2D spectroscopy signal levels, as well as trade-offs inherent to the technique.