Abstract Submitted for the DAMOP18 Meeting of The American Physical Society

Cavity-enhanced two-dimensional spectroscopy using higherorder modes THOMAS ALLISON, Stony Brook University — We describe methods using frequency combs and optical resonators for recording two-dimensional (2D) ultrafast spectroscopy signals with high sensitivity. By coupling multiple frequency combs to higher-order modes of one or more optical cavities, background-free, cavityenhanced 2D spectroscopy signals are naturally generated via phase cycling. As in cavity-enhanced ultrafast transient absorption spectroscopy, the signal to noise is enhanced by a factor proportional to the cavity finesse squared, so even using cavities of modest finesse, a very high sensitivity is expected, enabling ultrafast 2D spectroscopy experiments in dilute molecular beams.

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Date submitted: 06 Feb 2018

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