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Distinguishing excitonic from vibronic oscillations in ultrafast spectroscopy JACOB KRICH, University of Ottawa, JOEL YUEN, Harvard University, ALLAN JOHNSON, University of Ottawa, JOSEPH GOODKNIGHT, ALÁN ASPURU-GUZIK, Harvard University — Ultrafast experiments on photosynthetic and conjugated organic systems have indicated that coherent delocalization of exciton states significantly contributes to exciton transport, even up to room temperature. Oscillations in 2D spectra due to excitonic delocalization can be similar to those from vibronic oscillations, which are not important for exciton transport. We describe a straightforward experiment – broadband pump-probe – to distinguish between ultrafast oscillations from excitonic or vibronic-only sources. We consider anharmonic molecular oscillators and consider the requirements for how broadband (short in time) the experimental pulses must be to distinguish excitonic from vibronic oscillations.

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