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Monitoring coherent vibrational control with transient multidimensional infrared spectroscopy. MATTHEW ROSS, KEVIN KUBARYCH, Department of Chemistry, University of Michigan — Multidimensional infrared (MDIR) Fourier transform spectroscopy is a powerful experimental technique to relieve spectral congestion, revealing coherence transfer, energy relaxation, and coupling between states. Using novel non-equilibrium, transient MDIR sequences we monitor the effects of high vibrational excitation ($v > 2$) on intramolecular dynamics and system-bath interactions. MDIR spectroscopy offers a structurally sensitive probe of coherent control enabling a more detailed molecular interpretation of control mechanisms. A phase shaped, high-energy IR control pulse is timed to arrive either before or during a three-pulse echo sequence. We investigate the spectral phase dependence of this control pulse on the evolving MDIR spectrum in a system of strongly coupled CO oscillators in metal carbonyls. A key goal is controlling ground electronic state dynamics by exciting the system with optical excitation that pushes beyond the perturbative limit in order to enhance or suppress specific transitions in analogy to NMR spectroscopy.

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