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Influence of linear chirp on non-vertical transitions in a dye solution ELIZABETH CARROLL, ANDREI FLOREAN, PHILIP BUCKSBAUM, ROSEANNE SENSION, FOCUS Center, University of Michigan, Ann Arbor, MI, KEN SPEARS, Department of Chemistry, Northwestern University, Evanston, IL
— Chirped laser pulses can manipulate vibrational coherences in dye solutions. We show that the effect of linear chirp also depends strongly on the power spectrum of the ultrafast pulse. We use a programmable phase mask to control the spectral phase of 25-fs visible pulses generated in a noncollinear optical parametric amplifier (NOPA). Following chirped pulse excitation of the oxazine laser dye LD690, coherent oscillations are observed in the time-resolved transient absorption. When a vertical transition is excited, negative chirp leads to strong ground state vibrational coherences as resonant stimulated Raman processes are enhanced by the frequency sweep of the laser pulse. When the chirped pulse excitation is higher frequency than the Franck-Condon transition, the optical response of the dye is significantly affected by excited state absorption. In this case, coherences are established in excited states but no chirp-enhancement of the ground state wavepacket is observed.

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