

Abstract Submitted
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Coherent Chemistry with THz Pulses: TDDFT-Ehrenfest Simulations of Field-Induced LiNC Isomerization¹ LENSON PELLOUCHOUD, EVAN REED, Stanford Univ — The ability to coherently rearrange molecular structures is among the grand challenges of physical chemistry. Some of the primary obstacles are non-adiabatic increases in energy, such as intramolecular vibrational relaxation (IVR) and electronic excitations. Motivated by recent advances in the generation and control of strong terahertz (THz) pulses, we have investigated their potential to circumvent these obstacles. THz pulses are promising because their spectral content is well separated from electronic excitation frequencies, yet they may be fast enough to add and remove energy from the ionic system without allowing IVR to take place. In this work, we utilize simulations to discover that LiNC can be isomerized to two distinct metastable conformations with very low residual heating and ionization rates, pointing out a new route towards THz coherent control of chemical bonds and materials. We use time-dependent DFT (TDDFT)-based Ehrenfest molecular dynamics simulations to test a variety of strong time-varying THz pulses applied to the molecule. We find the limits of how quickly an activation barrier can be surmounted before the driving pulse becomes strong enough to ionize the molecule, and how well the target must be aligned in order for the final configuration to be stable.

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