

Spatial and temporal localization of the vibronic and photoelectron wave packets in LiH photoexcited by intense few cycle IR pulses

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Few-cycle ultrashort IR pulses allow excitation of coherently coupled electronic states towards steering nuclear motions in molecules. The progress of the non stationary vibronic density towards the products can be monitored using a second pulse, by transient absorption spectroscopy or photoionization. The Hamiltonian includes the pump and probe pulses which provides a quantum mechanical description of both multiphoton excitation and ionization. For pumping, we use an IR pulse with a controlled phase between its envelope and carrier wave. We report on the interplay between photoexcitation and photoionization in shaping the ensuing coupled electronic-nuclear dynamics in both the neutral excited electronic states and the cationic states of the diatomic molecule LiH. The dynamics is described by solving the time-dependent Schrodinger equation at nuclear grid points using the partitioning technique with a subspace of ten coupled bound states and a subspace of discretized continuous states for the photoionization continua. We show that the coherent dynamics in the neutral subspace is strongly affected by the amplitude exchanges with the ionization continua during the pulse, as well as by the onset of nuclear motion which drives non adiabatic interactions in the Franck-Condon region and on the way to the reaction products. Our methodology provides a visualization in space and in time of the entanglement between vibronic wave packet in the neutral states and the wave packet of the outgoing photoelectron.