

Control at a Conical Intersection through Adiabatic Ionization: Creating a “Pure” Wave Packet in the Strong Field Regime

Timothy Bohinski, Katherine Moore Tibbetts, Dmitri Romanov, Maryam Tarazkar, Spiridoula Matsika, Robert J. Levis

Center for Advanced Photonics Research, Department of Chemistry, Temple University,
Philadelphia PA 19122

The transition from nonadiabatic (multiphoton) to adiabatic (tunnel) ionization in the strong field regime as a means for high fidelity control of photoionization/dissociation in polyatomic molecules will be discussed. The time-resolved dynamics for a family of acetophenone radical cations measured as a function of the pump laser wavelength from 790 nm to 1500 nm has been measured experimentally and investigated theoretically. Coherence in the resulting ion signals as a function of time is enhanced by a factor of up to 40 in the adiabatic regime as compared with the nonadiabatic regime suggesting a new launch state for quantum control. The elimination of the launch state is achieved upon excitation with a 1370 nm pump pulse, revealing coherent motion on an excited ionic (D_2) surface with passage through a conical intersection seam multiple times. These measurements demonstrate the ability to prepare and manipulate multiple wavepackets in the vicinity of a conical intersection for polyatomic molecules on the time scale of picoseconds with up to six recurrences. The potential surfaces are calculated using the equations of motion approach and molecular dynamics on these surfaces reproduce the experimental measurements.