Quantum optimal control of photoelectron dynamics by interfering resonantly enhanced multiphoton ionization pathways

R. ESTEBAN GOETZ, LOREN GREENMAN, Department of Physics, Kansas State University — We present a model [Phys. Rev. Lett. 122, 013204 (2019)] based on many-body electron dynamics, variational scattering theory and optimal control theory to efficiently describe photoelectron continuum states and control photoionization dynamics in randomly oriented molecules. We show that a finite manifold of indistinguishable even-parity \((1 + 1')\) REMPI pathways interfering at a common photoelectron energy but probing different intermediate molecular states results in a significant enhancement of the photoelectron circular dichroism (PECD) in chiral molecules, outperforming widely used schemes including interference between opposite-parity photoionization pathways driven by bichromatic \((\omega, 2\omega)\) fields and sequential pump-probe ionization. Based on the Reconstruction of Attosecond Beating by Interference of Two-photon Transition (RABITT) technique, we also demonstrate coherent control over the PECD at a given RABITT sideband by manipulating the quantum interference arising from different continuum-continuum photon pathways. Finally, we consider a combination of different photon polarization directions in the driven field and analyze the parity of the interfering photoionization pathways to achieve 100% of anisotropy in the photoelectron emission probability.