Abstract Submitted for the DAMOP19 Meeting of The American Physical Society

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 pumpprobe 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.

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Date submitted: 01 Feb 2019

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