

Abstract Submitted
for the MAR16 Meeting of
The American Physical Society

Catching Conical Intersections in the Act; Monitoring Transient Electronic Coherences by Attosecond Stimulated X-Ray Raman Signals¹

KOCHISE BENNETT, MARKUS KOWALEWSKI, KONSTANTIN DORFMAN, SHAUL MUKAMEL, University of California, Irvine — Conical intersections (CIs) dominate the pathways and outcomes of virtually all photochemical molecular processes. Despite extensive experimental and theoretical effort, CIs have not been directly observed yet and the experimental evidence is inferred from fast reaction rates and vibrational signatures. We show that short X-ray pulses can directly detect the passage through a CI with the adequate temporal and spectral sensitivity. The non-adiabatic coupling that exists in the region of a CI redistributes electronic population but also generates electronic coherence. This coherent oscillation can then be detected via a coherent Raman process that employs a composite femtosecond/attosecond X-ray pulse. This technique, dubbed Transient Redistribution of Ultrafast Electronic Coherences (TRUECARS) is reminiscent of Coherent Anti-Stokes Raman Spectroscopy (CARS) in that a coherent oscillation is set in motion and then monitored, but differs in that the dynamics is electronic (CARS generally observes nuclear dynamics) and the coherence is generated internally by passage through a region of non-adiabatic coupling rather than by an externally applied laser.

¹Support provided by U.S. Department of Energy through Award No. DE-FG02-04ER15571, the National Science Foundation (Grant No CHE-1361516), and the Alexander von Humboldt foundation through the Feodor Lynen program.

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Date submitted: 06 Nov 2015

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