PROBING QUANTUM COHERENCE IN MOLECULAR SYSTEMS

SURESH YARLAGADDA, *Department of Chemistry, Wayne State University, Detroit, MI, USA*; TEMITAYO OLOWOLAFE, *Chemistry, Wayne State University, Detroit, MI, USA*; SUK KYOUNG LEE, *Chemistry Department, Wayne State University, Detroit, MI, USA*; H. BERNHARD SCHLEGEL, WEN LI, *Department of Chemistry, Wayne State University, Detroit, MI, USA*.

Ultrafast spectroscopy can initiate and probe electronic dynamics in molecules within femtoseconds to attoseconds timescale. Here, we report the use of strong field ionization pump-probe technique to detect multimode vibrational motions and electronic coherence in methyl iodide cation (CH3I+). For the first time, the coherence between the spin-orbit components of methyl iodide cation ground states and all symmetric vibrational modes were captured. The periodicities of the detected quantum beats vary between 6.0 fs and 117.0 fs. A few vibrational beatings from the first excited state (A state) were also detected. Furthermore, our approach reveals the time evolution of the quantum coherence in methyl iodide cation. The electronic coherence decays in the first picosecond while vibrational quantum beats persist. Our study further showed that rotational revival does not revive electronic coherence, suggesting both vibrational and rotational dephasings play a role in the decay of electronic quantum beatings. Theoretical analysis using a quantum model reveals intimate interaction between electronic and vibrational coherence in polyatomic systems.