Contribution ID: **11** Type: **Talk**

Molecular Orbital Imprint in Laser-Driven Electron Recollision

Friday 16 February 2018 11:10 (30 minutes)

Electrons released by strong-field ionization from atoms, molecules, or in solids can be accelerated in the oscillating laser field and driven back to their ion core. The ensuing interaction, phase-locked to the optical cycle, initiates the central processes underlying attosecond science. A key long-standing assumption regards the returning electron wavepacket as a plane wave. Here we study laser-induced electron rescattering associated with two different ionization continua in the same, spatially aligned, polyatomic molecule. We show by experiment and theory that the electron return probability is in fact molecular-frame dependent and carries structural information on the ionized orbital. Pronounced deviations of the returning wavepacket from plane-wave character have to be accounted for in analyzing attosecond experiments based on strong laser fields.

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Session Classification: Strong-field ionization

Track Classification: Invited talk