Abstract for an Invited Paper
for the DAMOP09 Meeting of
The American Physical Society

Molecular dynamics probed using high harmonic generation and strong field ionization

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The attosecond time-scale electron recollision process that underlies high harmonic generation has uncovered extremely rapid electronic dynamics in atoms and diatomics. We show that high harmonic generation can reveal coupled electronic and nuclear dynamics in polyatomic molecules. By exciting large amplitude vibrations in dinitrogen tetraoxide, we show that tunnel ionization accesses the ground state of the ion at the outer turning point of the vibration, but populates the first excited state at the inner turning point. This state switching mechanism is manifested as bursts of high harmonic light emitted mostly at the outer turning point. Theoretical calculations attribute the large modulation to suppressed emission from the first excited state of the ion. More broadly, these results show that high harmonic generation and strong field ionization in polyatomic molecules undergoing bonding or configurational changes involve the participation of multiple molecular orbitals. We also probe the electron rearrangement in a chemical reaction using strong field ionization. Electronic dynamics play the central role in a chemical reaction. It is extremely desirable for a detection technique to have the capability of probing the ultrafast electronic motion. Time resolved photoelectron spectroscopy can provide some insights. However, the most direct information of electronic dynamics such as the electron configurational change remains elusive. Recently, strong field ionization has been demonstrated to probe the static electron density distribution of the HOMO orbital. We report the preliminary results of applying strong field ionization as a probe to study the electron rearrangement in the photodissociation of bromine. This technique provides more complete information on reaction dynamics and is very promising of making *movies* of chemical reactions in atomic level.