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Femtosecond Isomerization Dynamics in the Ethylene Molecule[1]

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The ethylene molecule plays a fundamental and prototypical role for the understanding of photo-isomerizaton processes and particularly for ultrafast energy conversion through nonadiabatic transitions and state crossing via conical intersections. We have developed a high power femtosecond laser based pump-probe system to study femtosecond isomerization dynamics in various model molecules. By focusing 25-mJ laser pulses into a 5-cm-long xenon-filled gas cell, we can deliver about 10^9 photons per harmonic per pulse onto a target gas, with the photons ranging in energy from 8 to 40 eV. In this talk I will present the results of our studies of the dynamics in the excited ethylene cation ($C_2H_4^+$) using a high intensity high harmonic source. The dynamics in the excited ethylene cation leads, among other channels, to isomerization to the ethyledene configuration (CH_3CH^+), which is predicted to be a transient configuration for electronic relaxation. With an intense femtosecond EUV pulse as pump, and a NIR (near infra-red) pulse as probe, we measure a time scale of 45 ± 10 fs for formation of the transient ethylidene configuration (lifetime of 60 ± 15 fs) through detection of the NIR-induced fragmentation to CH_3^+ and CH^+. Also, a H_2-stretch transient configuration (believed to succeed ethylidene), yielding H_2^+, is found to be populated after 100 ± 10 fs. These studies were also extended to excited state dynamics in the neutral ethylene using a recently developed split mirror technique enabling XUV pump - XUV probe capability. In order to achieve this we optimized our high harmonic system for high power in order to produce a very intense source of high harmonics that allows multiphoton (XUV) absorption by a single molecule. In particular, we were able to measure two-photon double-ionization of Ethelyne and argon and three-photon double ionization of neon.

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