Femtosecond Dynamics and Multiphoton Ionization driven with an Intense High Order Harmonic Source

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Synopsis We have constructed a high intensity high order harmonic source delivering $\sim 10^9$ extreme ultraviolet photons/shot on a gas target and used it to observe multiphoton ionization and conduct femtosecond EUV-pump IR-probe experiments. Following excitation by 20-25 eV photons, we observed that the excited ethylene cation $(H_2C\text{-}CH_2)^+$ experienced isomerization to the ethylidene configuration $(HC\text{-}CH_3)^+$ in 50±25 fs, followed by an H_2 stretch motion. Experimental data and analysis from several other performed and planned experiments will be presented as well.

At the Lawrence Berkeley National Lab, we have developed an intense high harmonic generation (HHG) system, delivering around 10^9 photons per harmonic per shot into an experimental chamber. The drive laser (λ_0 = 800 nm) has a pulse duration of 35 fs, a pulse energy of around 30 mJ, and operates at 10 Hz. In a 5-cm-long gas cell (filled with krypton, xenon, or argon) the laser produces radiation at odd harmonic frequencies of the fundamental, after which phase matching conditions and optical reflective properties limit the range of photon energies available in the chamber to around 5-50 eV (EUV regime). Insertion of additional foils as well as a multi-layer coating on the final focusing mirror allow for further control of the delivered photon energy.

In a first series of experiments we used 20-25 eV photons to pump ethylene ($H_2C\text{-}CH_2$) to excited cationic states. We probed the subsequent isomerization channels with a NIR probe laser by looking at ion fragments, see Fig. 1. We observed directly the formation of the hydrogen-migrated isomer ethylidene $(HC\text{-}CH_3)^+$ by measuring CH^+ and CH_3^+ fragments. The ethylidene geometry is known to play a role in the internal conversion of electronic to nuclear energy, making ethylene a model molecule to study such processes. Optimum CH^+ and CH_3^+ yields occurred 80 fs after pumping. We explored several other ionic species (for example H_2^+ from a subsequent H_2 stretch), and have applied a model to match the isomerization data.

We will also discuss results on EUV-driven non-linear optics. At our high EUV intensity we find measurable quantities of multiphoton-driven products. For example, we have observed Ar^{2+}, Ne^{2+}, Xe^{3+}, $C_2H_2^{2+}$ and $C_2H_4^{2+}$ from multiphoton excitations. Recently we have finished the construction and implementation of a split-mirror stage, allowing for multi-color EUV-pump EUV-probe experiments with sub-fs temporal control. The latest results from this setup will be addressed.

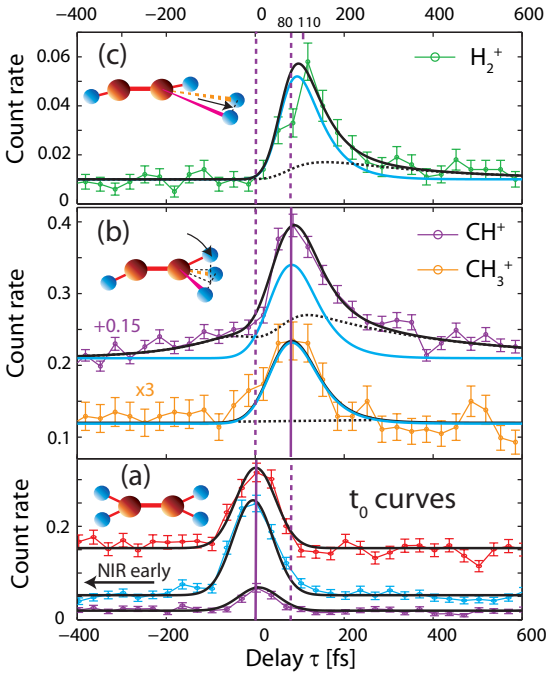

Fig. 1. (a) Ion yields to determine t_0, namely H_2O^+, $C_2H_3^{2+}$ and the isotope $^{29}C_2H_4^{2+}$. (b) Ion yields for the ethylidene fragments CH_3^+ and CH^+. Optimum yield occurs at $\tau = 80$ fs. (c) Ion yield for the fragment H_2^+ (optimum at $\tau = 110$ fs), believed to represent fragmentation from a transient configuration succeeding ethylidene. The solid black curves in (b) and (c) represent modeled curves, consisting of a background (black dotted curves) and a transient signal $S(\tau)$ (solid blue curves).

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