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"Slow" Molecular Fragmentation after Ultrafast Interaction

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Synopsis We report experimental observation of "slow" fragmentation from ethylene molecules on nanosecond or even microsecond timescale after their interaction with femtosecond laser pulses. Our results indicate that such fragmentation process originates from meta-stable high-lying vibrational states of ethylene ions.

Molecular fragmentation from dissociative states in molecules is a common process after their interaction with strong laser fields [1]. In general, such process happens on a very short, *picosecond* or even *femtosecond*, timescale. In this contribution, we present experimental observation of "slow" fragmentation on *nanosecond* or even *microsecond* timescale from ethylene molecules which are ionized within a few femtosecond by few-cycle laser pulses.

In the experiments, we recorded ionic products in coincidence from the interaction of ethylene molecules with strong near-infrared femtosecond laser pulses (with duration of 4.5 fs and peak intensity from a few 10^{14} W/cm^2 up to 2×10^{15} W/cm^2) in a reaction microscope apparatus [2, 3]. During the strong field interaction, the molecules can be doubly ionized and may be further broken into two moieties. We observed stable dications of ethylene and three two-body fragmentation channels from dications, as indicated in the photoion-photoion coincidence (PIPICO) spectrum in Figure 1.

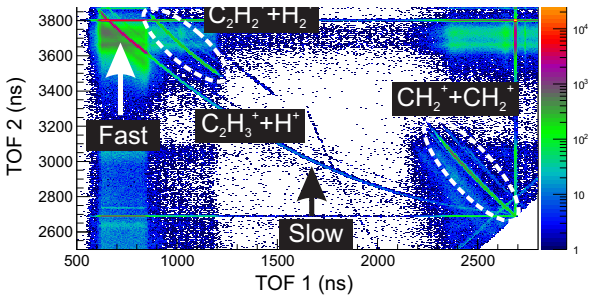

Figure 1. Measured PIPICO spectrum of ethylene molecule. The long tail of slow deprotonation of ethylene dication can be distinguished from the fast fragmentation process.

The two-body fragmentation channels are presented as parabolic lines in the PIPICO spectrum due to momentum conservation during the fragmentation process. Since the kinetic energy released (KER) of a two-body fragmentation from ethylene is on the order of a few electron volt, the corresponding PIPICO line is normally rather short. We found there is a long tail attached to the deprotonation PIPICO line, which is formed by a slow fragmentation process happening on nanosecond up to microsecond timescale after the interaction with ultrafast laser pulses. The KER of the slow fragmentation process and the lifetime of the corresponding ethylene dication is retrieved from the measurements for studying the origin of such process.

In the previous work, we found that the electronic ground state of ethylene dication is meta-stable [4]. Our observation indicates that the slow fragmentation of ethylene dication happens from meta-stable states with lifetime on the nanosecond timescale. In the quantum chemical simulations, we found that there are high-lying vibrational states on the electronic ground state of ethylene dication with nanosecond timescale lifetime. During the strong field interaction, such states might be populated through coupling with higher electronic excited states. After the laser pulse, dications can be trapped in such long lifetime meta-stable states which later lead to the observed slow fragmentation.

References

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