Control of electron localization in a molecule using XUV and IR pulses

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Synopsis: We demonstrate an experimental control of electron localization in the deuterium molecular ion created and dissociated by the combined action of an attosecond pulse train (APT) and a multicycle IR laser pulse. A left-right asymmetric ejection of the deuterium ions, characterized by an asymmetry parameter A, exhibits oscillations with a full IR period when the time-delay between the APT and IR pulses is scanned with 300as resolution.

The control of electron localization in molecules is crucial in order to manipulate and steer many chemical reactions [1]. A key experimental step in this direction has been achieved by a recent experiment whereby a few cycle IR laser pulse was used to control the electron localization in molecular dissociation [2]. This technique requires the generation of strong few fs long phase stabilized pulses.

Here we experimentally demonstrate an alternative but powerful approach to control the electron localization using a combined action of linearly polarized attosecond pulse train (APT) and 50fs long IR pulse. The APT is synthesized using a two color technique by combining 800nm IR pulse with its second harmonic. The resulting APT contains one attosecond pulse per IR laser cycle which is necessary in order to observe the electron localization effect [3]. Moreover, because second harmonic is phase-locked to the fundamental 800nm pulse, any need for carrier phase stabilization is eliminated. Both the APT and the fundamental IR pulse are focused simultaneously on the deuterium target gas inside the COLTRIMS reaction microscope and the time-delay between them is scanned in steps of 300as.

The produced ionic fragments are ejected either parallel (left) or anti-parallel (right) to the laser polarization axis. The asymmetry between left-right ejection of the D^{+} ions is quantified by the asymmetry parameter, A, defined as

$$A = \frac{C_{left} - C_{right}}{C_{left} + C_{right}},$$

where C_{left}, C_{right} are the corresponding counts.

The Fig. 1 shows the parameter A versus the time-delay between the IR and APT pulses (zero delay corresponds to overlapping pulses).

The parameter A clearly oscillates with full laser cycle period of 2.7fs with about 5% contrast. Furthermore, an analysis of the kinetic energies of the released ionic fragments suggests that the underlying mechanism of the localization control takes place primarily as the IR induces the molecular dissociation for selected internuclear distances.
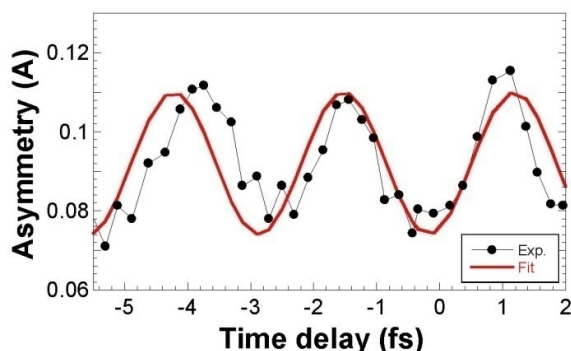
Fig. 1. The asymmetry parameter A versus the time delay between APT and IR pulses. Red solid line is a fit to sinusoidal function with the laser period.

Our data are supported by theoretical calculations based on numerically solving time-dependent Schrodinger equation for the system, which reproduces well the observed oscillations of the asymmetry.

References

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