

Controlling isomerization and fragmentation of polyatomic molecules by laser-sub-cycle electron recollision

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We present the first experimental demonstration of control of fragmentation pathways of polyatomic molecules (acetylene, ethylene, 1,3-butadiene) [1] using the sub-cycle shape of few-cycle laser pulse, characterized by the carrier-envelope phase (CEP), as the control parameter. Given the increased number of participating nuclei and a vastly more complex valence electron dynamics and the structure of energy surfaces, it is noteworthy that the CEP dependence of fragmentation pathways still survives in polyatomic molecules. We argue both experimentally and theoretically, that this is due to the existence of a universal attosecond mechanism of quasi-single-cycle fragmentation of large molecules, namely the field control of the tunneled-out electronic wave packet that is steered by the optical waveform before its recollision with the parent ion.

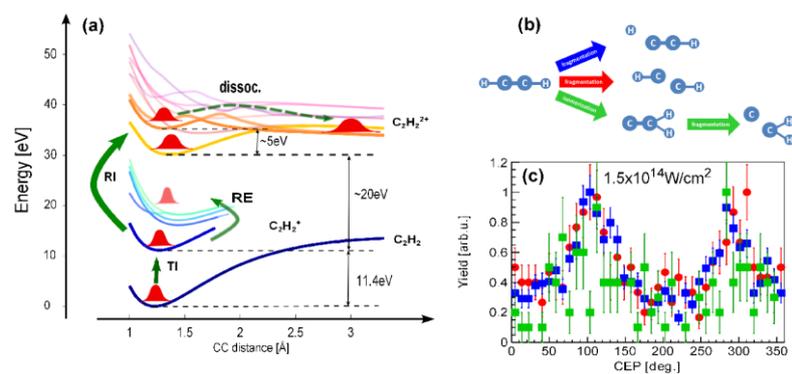


Fig. 1 (a) Electronic energy levels of the neutral, cation, and dication of acetylene as a function of C-C distance. TI, RI and RE denote tunnel ionization, recollision ionization, and recollision excitation, respectively. (b) Visualization of three fragmentation channels of the acetylene dication. (c) Measured fragmentation yields, normalized to 1, as a function of CEP for the three fragmentation channels depicted in (b) with the same color coding.

The process is sketched in Fig. 1(a) for the example of the acetylene molecule: Upon electron recollision a second electron is knocked out from an inner-valence orbital. This puts the molecule into an electronically excited ionic state from where the molecule might dissociate. In our experiments we measured the yields of several two-body fragmentation pathways of acetylene, ethylene and 1,3-butadiene molecules as a function of the CEP of ~4.5 fs laser pulses using COLTRIMS in combination with a phase-meter [2]. Fig. 1 exemplarily shows the strong modulation of the fragmentation yield measured for three different pathways of acetylene fragmentation. The results obtained for the other molecules are similar. At the conference we will present a detailed overview over all our experimental and theoretical results.

References:

- [1] X. Xie et al., Phys. Rev. Lett. 109, 243011, (2012)
 [2] A. M. Saylor et al, Opt. Lett. 36, 1 (2011)