High energy proton ejection from hydrocarbon molecules driven by highly efficient field ionization

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We report on the observation of an ionization-fragmentation process that puts a polyatomic molecule into a very high charge state of +12 and beyond at laser intensities as low as a few 10^{14} W/cm^2 and subsequently leads to complete molecular decomposition. Our surprising experimental evidence shows that all of the molecule's protons are ejected with high kinetic energies, which is inconsistent with the explanation based on dynamic charge localization used in [1] to explain the proton energies. Furthermore, the energies of all the protons are very similar, which indicates that at the instance of decomposition the protons interact with the same molecular charge state, suggesting an all-at-once (concerted) fragmentation scenario.

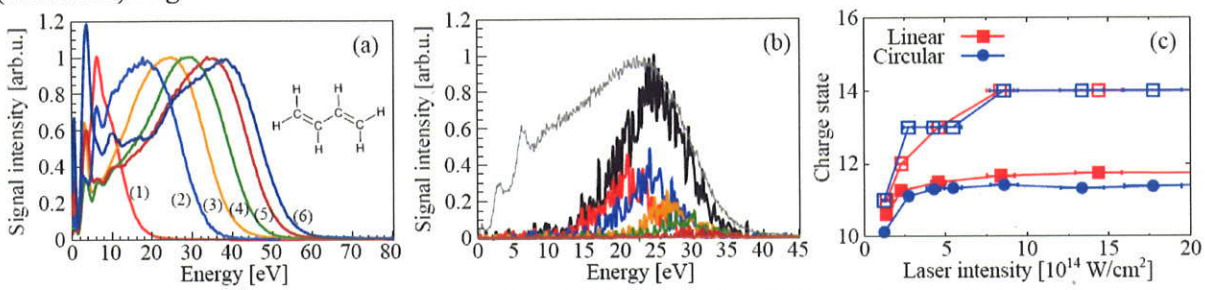

Fig. 1 (a) Measured proton energy spectra from 1,3-butadiene for different laser peak intensities; (b) Decomposition of a proton spectrum (grey) into different fragmentation channels; (c) Average and maximum (full, open symbols) charge state before fragmentation as a function of laser peak intensity.

In our experiments we measure the energy spectra of protons ejected from methane, CH_4, ethylene, C_2H_4, 1,3-butadiene, C_4H_6, or hexane, C_6H_{14}, molecules when irradiated by 27 fs Ti:Sapphire laser pulses. The spectra extend to surprisingly high energies, as shown in Fig. 1(a) for different laser peak intensities. Using multi-particle coincidence techniques [2] we are able to decompose the spectra into the contributions from different fragmentation channels, cf. Fig. 1(b) where the black line shows the sum of individual contributions. From the decomposition we can calculate the charge state that the molecules fragment from. An average and maximum charge state as a function of intensity is shown in Fig. 1(c) for linear and circular light. The surprisingly high charge state explains the measured proton energies, but raises other questions: Due to which mechanism can the molecules become ionized so many times? How can the molecules possibly absorb so much energy from the ionizing field? Careful analysis of the data suggests the following scenario: During the first field ionization stages at the leading edge of the pulse, the C-H bonds start to stretch until they reach the critical inter-nuclear distance for enhanced ionization (EI) [3, 4], upon which the molecule becomes ionized several times in parallel at many different C-H bonds within a short period. Subsequently, all of the protons are removed from the highly charged ionic complex by Coulomb explosions in a concerted multi-particle fragmentation. Thereafter, the remaining structure decomposes into bare atomic carbon ions.

In the particular case of hydrocarbon molecules, the very fast nuclear motion of C-H bonds on the order of 10 fs can make this process very efficient even for the interaction with very short (down to a few optical cycles) laser pulses. Therefore, the identified mechanism has implications for any strong-field experiment on molecules, including high harmonic generation experiments.

References

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