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Strong field ionization of small hydrocarbon chains with full 3D momentum analysis

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Synopsis Strong field ionization of small hydrocarbon chains is studied in a kinematic complete experiment using a reaction microscope. By coincidence detection of ions and electrons different ionization continua populated during the ionization process are identified. In addition, photoelectron momentum distributions from laser-aligned molecules allow to characterize the electron wavepackets emerging from different Dyson orbitals.

Strong field driven spectroscopy of molecules is a very active field of research which promises to bring unprecedented time- and spatial resolution to molecular dynamics studies. It has become clear in recent years that a number of the initial simplifying assumptions, such as the adiabatic and the single-electron approximation, can break down and have to be replaced by a multi-electron picture. In particular the appearance of multiple ionization continua in Strong-Field Ionization and High Harmonic Generation of polyatomic molecules is being increasingly investigated theoretically and experimentally. The interpretation of the strong field ionization is largely facilitated by a complete experiment, where the full 3-D momentum vectors of all outgoing particles created during the photoionization process are registered. A so-called reaction microscope, which has already been developed more than a decade ago, enables this type of complete experiments by detecting the energy and angular distribution of photo electrons and ions in coincidence. Recently, we have set-up a laboratory where we combine a high repetition rate laser with a reaction microscope [1].

In a first experimental run we have studied small hydrocarbon chains like n-Butane and Butadiene. These molecules have already been investigated in similar experiments [2]. By correlated ion-electron detection it was shown that different ionization continua are excited during strong field ionization. In our experiment the hydrocarbon chains are ionized with a sub-7 fs, $4 \ \mu J$ pulses at 800 nm. These pulses are focused by an 80 mm focal length spherical mirror onto a supersonic molecular beam. The intensity in the interaction region is on the order of 10^{13} to 10^{14} W/cm^2 . Our experiments confirm the population of different excited ionic states. In addition, we were able to determine a complete energy and angular distribution of the photo electrons emerging from different populated states.

Presently, this study is extended by using laser-aligned hydrocarbon molecules [3]. In these experiments we are studying how multiple strong-field ionization continua manifest themselves in electron recollision driven by the strong laser field. Electron diffraction patterns for different fragments promise to characterize ionization channel resolved continuum electron wavepackets, emerging from different Dyson orbitals. This might evolve into an ultrafast dynamical probe of electronic coherences in molecules.

References

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- [2] A. E. Boguslavskiy et al. 2012 Science 335 1336
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