Electron localization in molecular fragmentation with CEP stabilized laser pulses

Bettina Fischer*[,1], Manuel Kremer*[,2], Bernold Feuerstein*, Vitor L.B. de Jesus[‡], Artem Rudenko[†], Claus Dieter Schröter*, Robert Moshammer* and Joachim Ullrich*

* Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany
[†] Max-Planck Advanced Study Group at CFEL, 22607 Hamburg, Germany
[‡] Instituto Federal do Rio de Janeiro – IFRJ, 26530-060 Nilópolis-RJ, Brazil

Fully differential data on dissociating H2 molecules in ultrashort (~6 fs), linearly polarized, intense (~0,4 PWcm-2) laser pulses with stabilized carrier-envelope-phase (CEP) have been measured using a reaction microscope. Depending on the CEP of the laser pulses we see a clear asymmetry in the emission direction of the created protons. Contrary to earlier measurements by Kling et al. [1] we observe the largest asymmetry in a kinetic energy release (KER) range between 0-3 eV. This excludes the recollision mechanism suggested in [1] and requires another explanation.

Recent experiments [1] showed the possibility to measure and even control electron localization in the dissociating D_2^+ molecular ion by intense, ultrashort, CEP controlled laser pulses.

In contrast to these earlier asymmetry measurements that have been performed by using velocity map imaging, we used a reaction microscope [3] that allows us to reconstruct the full three dimensional momentum vectors of all outgoing (charged) particles. Furthermore, measuring electrons and ions in coincidence facilitates a channel separation that is not accessible by other techniques.

For measuring the asymmetry in the proton (deuteron) emission direction, it is useful to define the so called asymmetry parameter $A=(N_{up}-N_{down})/(N_{up}+N_{down})$, where N_{up} (N_{down}) is the number of protons emitted to the upper (lower) hemisphere, respectively.

The result of our experiment is shown in Fig. 1. Here, the asymmetry parameter is plotted as a function of the KER of the dissociating H_2^+ and the CEP of the ultrashort laser pulse. Contrary to [1] we see clear changes in the asymmetry for different CEP in a KER range between 0-3 eV similar to predictions in [3]. Furthermore we see a tilt in the asymmetry stripes that was not observed for the fragments at higher energy measured in [1], where the stripes were vertical. By looking at different emission angles α of the H_2 molecule relative to the laser polarization axis the tilted stripes shift with respect to the CEP.

This behavior can not be explained by the model introduced by Kling et al. who observed KERs between 6-16 eV resulting from the rescattering of the first electron with the H_2^+.

The low energy fragments observed here however can be attributed to bond softening. The CEP-dependent asymmetry is introduced by the coupling of the $1s\sigma_g$ and $2p\sigma_{u\ s}$ states of the H_2^+.

We performed one dimensional TDSE calculations, which qualitatively reproduce our experimental results.

An asymmetry in the electron emission was also observed but did not show any relation to the asymmetry observed in the proton emission.

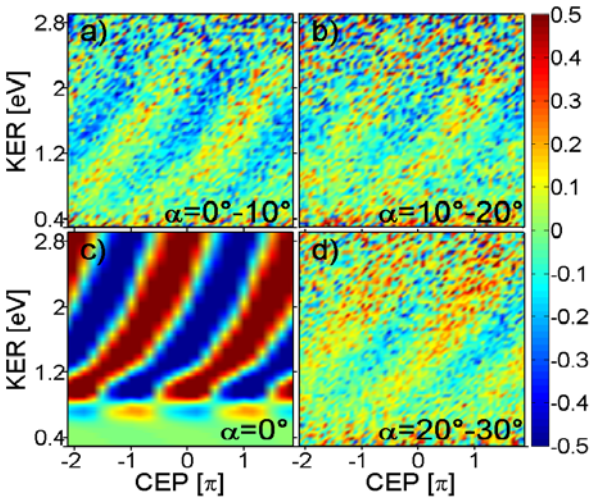

Fig. 1. Asymmetry parameter A in dependence of the proton energy and the CEP for (a) emission angles between 0°-10° and (b) between 10°-20° (d) between 20°-30° with respect to the laser polarization axis. (c) Result of the TDSE calculation for an emission angle of 0° .

References

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[1] E-mail: Bettina.Fischer@mpi-hd.mpg.de
[2] E-mail: Manuel.Kremer@mpi-hd.mpg.de