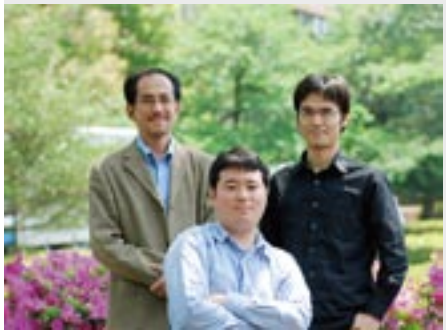
Atoms & Molecules in Few-Cycle Intense Laser Fields

Department of Photo-Molecular Science
Division of Photo-Molecular Science III



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Intense laser fields, comparable in magnitude with the Coulomb field within atoms and molecules, can be generated by focussing high-energy and ultrashort laser pulses. When exposed to such an intense laser field, molecules exhibit various exotic features that cannot be observed in weak laser fields. We are seeking a deeper understanding of the behavior of molecules in intense laser fields, to elucidate how molecules interact with light, as well as to apply the new features they exhibit to the real-time visualization of ultrafast chemical reactions and their control. In particular, we focus on the following research themes:

(1) Understanding of atomic and molecular dynamics in intense laser fields
(2) Ultrafast reaction imaging by laser Coulomb explosion imaging
(3) Generation and application of ultrashort soft-X-ray pulses by laser high-order harmonics.

1. Visualizing Recurrently Migrating Hydrogen in Acetylene Dication by Ultrashort Intense Laser Pulses[1,2)]

Highly charged molecular ions formed in ultrashort intense laser fields subsequently explode into fragments due to the strong electrostatic repulsion among the positive charges. The structure of the molecules at the instance of the laser irradiation can be studied by measuring precisely the momenta of the fragments. This can be compared to fireworks exploded in the night sky, which reflect how the "stars" containing sparklers are embedded in the shells. We studied the location of hydrogen atoms in deuterated acetylene dication, by igniting the "molecular fireworks" at different times after the creation of dication. We demonstrate the visualization of ultrafast hydrogen migration in deuterated acetylene dication ($C_2D_2^{2+}$) by employing the pump-probe Coulomb explosion imaging with sub-10-fs intense laser pulses (9 fs, 0.13 PW/cm^2, 800 nm). It is shown, from the temporal evolution of the momenta of the fragment ions produced by the three-body explosion, $C_2D_2^{3+} \rightarrow D^+ + C^+ + CD^+$, that the migration proceeds in a recurrent manner: The deuterium atom first shifts from one carbon site to the other in a short time scale (~90 fs) and then migrates back to the original carbon site by 280 fs, in competition with the molecular dissociation. Hydrogen migration plays important roles in various chemical reactions such as the synthesis of vitamin D in skin. The direct visualization demonstrated here will provide a deeper understanding of such chemical reactions as well as new prospects for their control.

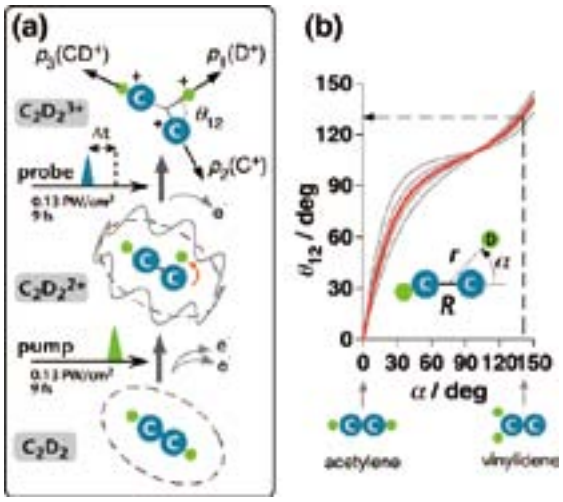

Figure 1. (a) Pump-probe Coulomb explosion imaging employed in the present study. The pump pulse creates $C_2D_2^{2+}$ and triggers the hydrogen migration. The instantaneous location of migrating deuterium atoms is determined from the momenta of fragment ions, D^+, C^+, CD^+, ejected in the Coulomb explosion of $C_2D_2^{3+}$ induced by the probe pulse. (b) Momentum angle θ_{12} as a function of the polar angle α obtained by classical simulation of the three-body Coulomb explosion.

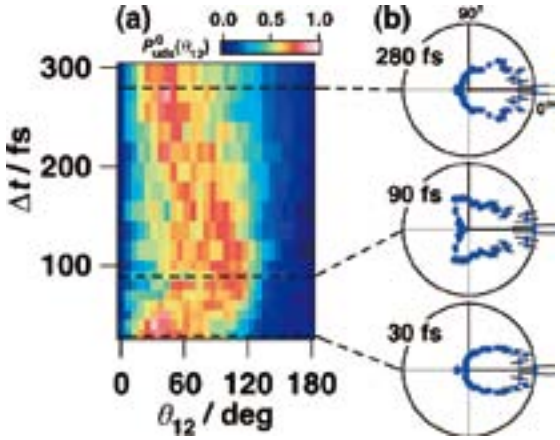

Figure 2. (a) Evolution of the momentum angle (θ_{12}) distribution and (b) the polar plot, showing the recurrent hydrogen migration in $C_2D_2^{2+}$.

2. Acetylene-Vinylidene Isomerization in Ultrashort Intense Laser Fields Studied by Triple-Ion Coincidence Momentum Imaging[3)]

Polyatomic molecules exposed to intense laser fields exhibit a variety of characteristic features associated with their many degrees of vibrational freedom. Due to the strong interaction with the laser fields, the geometrical structure can be strongly deformed by bond stretching or bending, and by isomerization through the rearrangement of chemical bonds. Hydrogen migration is one such processes, in which hydrogen atoms shift from one site to another within a molecule. Because of their large mobility, hydrogen atoms can explore the potential energy surfaces over long distances within the ultrashort laser pulse, leading to the formation of molecular hydrogen ion and the emission of energetic protons. In the present study, the isomerization of acetylene via hydrogen migration in intense laser fields (8×10^{14} W/cm^2) has been investigated by coincidence momentum imaging of the three-body Coulomb explosion process, $C_2H_2^{3+} \rightarrow H^+ + C^+ + CH^+$.

When ultrashort (9 fs) laser pulse are used, the angle between the momenta of C^+ and H^+ fragments exhibits a sharp distribution peaked at a small angle (~20°), showing that the hydrogen atom remains near the original carbon site in the acetylene configuration. On the other hand, a significantly broad distribution extending to larger momentum angles (~120°) is observed when the pulse duration is increased to 35 fs, showing that the ultrafast isomerization to vinylidene is induced in the longer laser pulse.

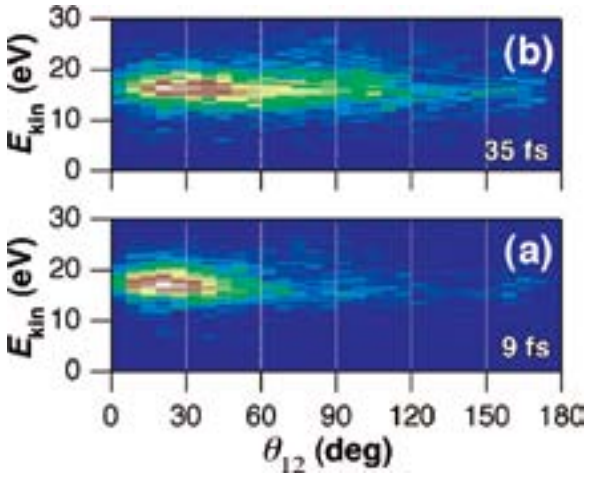

Figure 3. Two-dimensional maps of the momentum angle 12 between $\boldsymbol{p}_1(H^+)$ and $\boldsymbol{p}_2(C^+)$ and the total kinetic energy release for the asymmetric three-body Coulomb explosion process, $C_2H_2^{3+} \rightarrow H^+ + C^+ + CH^+$, observed for the (a) 9 fs and (b) 35 fs intense laser fields 8×10^{14} W/cm^2.

References

1) A. Hishikawa. A. Matsuda, M. Fushitani and E. J. Takahashi, *Phys. Rev. Lett.* **97**, 258302 (2007).
2) A. Hishikawa. A. Matsuda, M. Fushitani and E. J. Takahashi, *J. Phys.: Conf. Series* **88**, 012056 (2007).
3) A. Hishikawa, A. Matsuda, E. J. Takahashi and M. Fushitani, *J. Chem. Phys.* **128**, 084302 (2008).

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