

Ultrafast electronic and structural dynamics induced by XFEL pulses

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The advent of short-wavelength free-electron laser has opened new pathways in the investigations of electronic and structural dynamics in any form of matter. One of the most expected study using XFEL is single-shot X-ray imaging based on “before destroy” concept. However, the target samples should be affected by ultrafast reactions occurring during XFEL pulse duration. In order to utilize XFEL for “before destroy” imaging, it is indispensable to understand what happens in the matter during irradiation of the XFEL pulse. The ultrafast reaction dynamics in atoms, molecules and atomic clusters in the gas phase are not only fundamentally interesting but also important for this purpose.

In March 2012, SPring-8 Angstrom Compact free electron LASER (SACLA), started user operation in Japan [1]. We set up a program to investigate reaction dynamics in atoms, molecules and clusters induced by intense ($\sim 50 \mu\text{J}/\mu\text{m}^2$), ultrashort (~ 10 fs) pulses generated by SACLA. At photon energy of 5.0-5.5 keV, we could identify that Xe^{n+} with n up to 26 is produced, evidencing occurrence of deep inner-shell ionization and sequential electronic decay cycles repeated multiple times in the xenon atom within ~ 10 fs pulse duration [2]. The results for momentum-resolved multiple ion coincidence study on iodine-contained organic molecules (CH_3I , 5-iodouracil, CH_2I_2) illustrates that the charges are produced by the cycles of deep inner-shell ionization of the iodine atom and sequential electronic decay and spread over the entire molecule within 10 fs, leading to Coulomb explosion [3-5]. The measured momentum distributions and correlations are well reproduced by both classical and quantum mechanical MD simulations. The results for electron spectroscopy on argon and xenon clusters, with help of theoretical calculations, illustrate that nanoplasma are formed by the XFEL pulse, in tens of fs, and continuous thermal emission from the plasma occurs in ps [6]. The experiment also confirmed x-ray induced oligomer formation from rare-gas clusters [7]. We have carried out also XFEL pump-NIR probe experiments for xenon atoms and clusters [8], as well as iodine-contained organic molecules [9], at time resolution down to a few tens of femtoseconds, in order to probe the XFEL-induced ultrafast reaction in real time. The latest results will be shown and discussed.

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References

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