【Grant-in-Aid for Specially Promoted Research】

Science and Engineering (Chemistry)

Title of Project： Sub-femtosecond molecular imaging

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Research Project Number：15H05696　Researcher Number：40182597
Research Area：Basic chemistry, Physical chemistry
Keyword：Ultrafast chemistry, Reaction dynamics

【Purpose and Background of the Research】

Recent advances in the ultrashort pulsed laser technologies enabled us to generate laser pulses with the duration of ~5 fs, with which a variety of characteristic ultrafast molecular processes have been elucidated. However, the temporal resolution of ~5 fs is not short enough to probe in real time the intramolecular charge transfer occurring within molecules in response to an ultrashort pulsed laser field as well as the subsequent correlated motion of hydrogen atoms within those molecules. Currently, elucidation of such early stage dynamics proceeding within the sub-femtosecond (<1 fs) timescale is considered as the most urgent issue in ultrafast chemistry.

In this project, on the basis of experimental techniques and quantal dynamics theories we have developed by ourselves, we will investigate ultrafast intramolecular charge transfer processes and subsequent changes in the geometrical structure of molecules with the sub-femtosecond temporal resolution.

【Research Methods】 (See Fig. 1)

In order to clarify how the electronic response to an intense laser field triggers motion of light atoms such as hydrogen atoms and the other heavier atoms within a molecule, leading to breaking and rearrangement of chemical bonds, we will develop two experimental techniques; (i) coincidence momentum imaging (CMI) by which geometrical structures of molecules are determined from momenta of photoelectrons and fragment ions generated through multiple ionization of molecules followed by Coulomb explosion processes and (ii) laser-assisted electron diffraction (LAED) by which snapshots of instantaneous geometrical structures of molecules are obtained. Furthermore, by constructing a theoretical framework beyond the Born-Oppenheimer approximation, we will investigate mechanisms of ultrafast intra-molecular charge transfer occurring within a laser field and the subsequent ultrafast hydrogen migration.

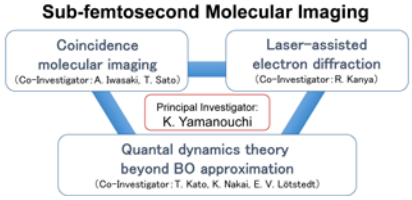

Fig. 1. Our Research Organization

【Expected Research Achievements and Scientific Significance】

By the CMI, the electronic excitation processes of molecules occurring within the duration of laser pulses as well as the accompanying ultrafast nuclear motion leading to chemical bond breaking and rearrangement will be elucidated. By the LAED, ultrafast variations of the electron density distribution as well as the distribution of nuclei within a molecule will be visualized with the sub-femtosecond temporal resolution. On the other hand, by developing non-adiabatic theories, correlated motion among electrons and nuclei in a sub-femtosecond timescale will be investigated, through which procedures for extracting the information of the electron-nuclear correlation from experimental data will be proposed.

By the experimental and theoretical studies described above, we will be able to understand the very early stage molecular dynamics, in the sub-femtosecond time domain in which electrons and nuclei within a molecule are correlated, leading to the chemical bond breaking and rearrangement processes occurring in the later stage in the femtosecond time domain.

【Publications Relevant to the Project】

K. Kurihara, H. Kono, H. Fukumura, K. Yamanouchi, *eds.*, "Kyokoushiba no Kagaku," *CSJ Current Review* **18**, pp. 1-28, 65-76, and 98-102 (Kagaku Dojin, 2015).

【Term of Project】 FY2015-2019

【Budget Allocation】 399,600 Thousand Yen

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