【Grant-in-Aid for Specially Promoted Research】

Science and Engineering

Title of Project： Strucutral visualization of photochemical reaction with time-resolved X-ray solution scattering

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【Purpose and Background of the Research】

As the saying goes "seeing is believing", it is a dream of chemists to directly observe molecular structural dynamics in solution. While time-resolved spectroscopies provide information on electronic states and molecular vibrations, they provide indirect and limited information on molecular structures in principle. The most promising measurement method that can provide direct information on molecular structure is the time-resolved X-ray scattering.

The biggest challenge of the current time-resolved X-ray solution scattering method is that heavy elements such as gold (Au) or iodine (I), which is located below the 5th and 6th period of the periodic table, must be included in samples due to the limitation of the signal-to-noise (S/N) ratio of the measurement. We will substantially improve the S/N ratio in this research project for expanding the range of molecules to which this method can be applied, and will promote a new research field in ultrafast molecular structure science.

【Research Methods】

In this study, we will improve the time-resolved X-ray solution scattering method (Figure 1) based on the results of our previous researches and developments as follows.

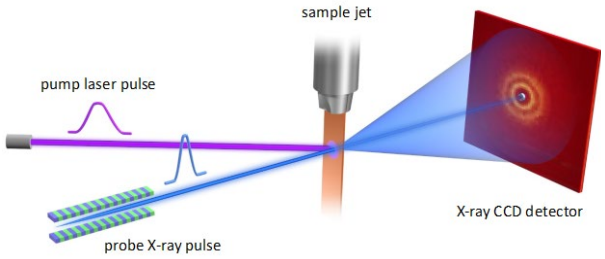

Figure 1. Schematic drawing of time-resolved X-ray solution scattering

The synchrotron radiation ring at KEK, PF-AR, is dedicated for single bunch mode operation, and the revolution frequency is set to 794 kHz. By using this frequency as the sampling frequency for measurement, the sampling rate will be improved by about three orders of magnitude (794 kHz) from the current sampling rate at 1 kHz. This will improve the signal-to-noise ratio of the measurement by a factor of about 30, and will make it possible to observe changes in the molecular structure even for molecules consisting only of light elements. We will appropriately address technical issues such as X-ray focusing and heat loads on X-ray optics based on the results of our previous researches and developments. We will also try to develop a measurement system in femtosecond order measurement using XFEL facilities, which are undergoing high repetition rate operation.

【Expected Research Achievements and Scientific Significance】

In recent years, there has been an increasing demand for structural information in the non-equilibrium state where the sample is actually functioning, and this has led to the development of new approaches to "understanding the non-equilibrium state at the atomic level" in various materials researches. At the same time, new attempts for understanding the non-equilibrium state at the atomic level are being made in various materials researches by increasing the brilliance of synchrotron radiation, improving the performance of detectors, and introducing new data analysis methods. This project aims to expand the range of application of time-resolved X-ray solution scattering by improving the signal-to-noise ratio of measurements for typical condensed system dynamics, *i.e.*, photochemical reactions in solution, and is of great significance and impact for a wide range of research fields.

【Publications Relevant to the Project】

・ J. G. Kim, S. Nozawa, H. Kim, E. H. Choi, T. Sato, T. W. Kim, K. H. Kim, H. Ki, J. Kim, M. Choi, Y. Lee, J. Heo, K. Y. Oang, K. Ichiyanagi, R. Fukaya, J. H. Lee, J. Park, I. Eom, S. H. Chun, S. Kim, M. Kim, T. Katayama, T. Togashi, S. Owada, M. Yabashi, S. J. Lee, S. Lee, C. W. Ahn, D.-S. Ahn, J. Moon, S. Choi, J. Kim, T. Joo, J. Kim, S. Adachi & H. Ihee, "Mapping the emergence of molecular vibrations mediating bond formation" ***Nature*****, 582**, 520–524 (2020).

・ K. H. Kim, J. G. Kim, S. Nozawa, T. Sato, K. Y. Oang, T. W. Kim, H. Ki, J. Jo, S. Park, C. Song, T. Sato, K. Ogawa, T. Togashi, K. Tono, M. Yabashi, T. Ishikawa, J. Kim, R. Ryoo, J. Kim, H. Ihee & S. Adachi, "Direct observation of bond formation in solution with femtosecond X-ray scattering", ***Nature***, **518**, 385-389 (2015).

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