5 ERATO Non-Equilibrium Dynamics Project

5-1 Outline

Introduction

The Molecular Dynamic Imaging Group, which belongs to the Non-equilibrium dynamics project under the ERATO program of the Japan Science and Technology Agency (JST), has been actively working for two years at KEK. The project is financially supported by the Ministry of Education, Culture, Sports, Science and Technology (MEXT). The project director is Prof. Shin-ya Koshihara, a professor at the department of chemistry and materials science, Tokyo Institute of Technology.

The ERATO project will continue for 5 years with a total budget of 1.5 billion yen. The goal of the project is to reveal the physics of ultrafast non-equilibrium dynamics of novel condensed matters that undergo photo-induced phase transitions. The project consists of the following fields; (1) precise structure analysis of non-equilibrium states based on picosecond-resolved X-ray diffraction/scattering by using single-bunch mode of the PF-AR, (2) development of a new beamline NW14 fully dedicated to X-ray diffraction/scattering measurements, (3) synthesis of organic or non-organic crystals that show strongly correlated dynamics between photon, electrons, and lattice structure when triggered by femtosecond laser pulses, (4) characterization of phase transitions with conventional optical measurements, (5) feasibility studies to realize dynamic imaging with femtosecond resolution using newly-developed femtosecond X-ray sources.

In addition to the research groups under the project, we are also collaborating with other groups. For crystal preparation, we have branches at the Tokyo Institute of Technology for the preparation of inorganic materials and at Kyoto University for organic ones. We are also collaborating with a group at Osaka City University for protein crystals with photosynthetic reaction centers. Another branch in Tokyo Institute of Technology is working on crystal characterization using optical methods. The administrative section of the project is also located at KEK. Collaboration with Rennes University in France and LBNL in the USA also started in 2004.

Members

In order to work on a range of photo-induced phase transition phenomena, researchers from various fields such as ultrafast laser science, solid state physics, crystallography, sample preparation and synchrotron radiation instrumentation are necessary. In 2004, several researchers joined the project who specialize in ultrafast lasers, X-ray spectroscopy, X-ray crystallography, and theory of solid state physics. Three students on master's and doctoral courses joined the project as well.

Figure 1
Members of the project.

Office

The PF-AR User Room, next to the PF-AR North-West Building, is currently occupied by the ERATO members: ERATO researchers, the research manager, the administrative manager, and an administrative assistant. The project will be managed as "an independent mini institute" for 5 years at the project office for administration.

5-2 Research Subjects

Photo-induced phase transitions

Since photo-induced phase transitions in condensed matters is one of the most promising phenomena for demonstrating molecular dynamic imaging, we are focusing our efforts on pump-probe X-ray measurements of photo-induced structural transitions in solids. Preliminary experiments on an organic charge transfer complex crystal succeeded with the combination of 100 picosecond synchrotron pulses and femtosecond laser pulses at the ESRF [1]. Since the X-ray and laser pulses must be synchronized by 1:1 during the pump-probe X-ray diffraction experiments, the repetition rate of the X-ray pulses from synchrotron ring must be reduced to 1 kHz by using a high-speed chopper (X-ray pulse selector). Thus, single-bunch operation (i.e., relatively low repetition rate operation) of synchrotrons is crucial for pump-probe diffraction experiments. The PF-AR is a unique facility in that single bunch mode is the normal operation mode, and this is why the Molecular Dynamic Imaging Group is located at KEK.

Many efforts on the study of photo-induced phase transitions of organic charge transfer complex materials such as TTF-CA, and $(EDO\text{-}TTF)_2PF_6$ are underway to obtain molecular dynamic imaging at beamline AR-NW2A. So far, a gigantic photo-response at the photo-induced phase transition was observed in $(EDO\text{-}TTF)_2PF_6$

in the infrared region [2].

References

[1] E. Collet, M.-H. Lemee-Cailleau, M. Buron-Le Cointe, H. Cailleau, M. Wulff, T. Luty, S. Koshihara, M. Meyer, L. Toupet, P. Rabiller and S. Techert, *Science,* **300** (2003) 612.
[2] M.Chollet, L. Guerin, N. Uchida, S. Fukaya, H. Shimoda, T. Ishikawa, K. Matsuda, T. Hasegawa, A. Ota, H. Yamochi, G. Saito, R.Tazaki, S. Adachi and S. Koshihara, *Science*, **307** (2005) 86.

5-3 Beamline Development

NW2 beamline

Installation of a laser hutch near the experimental hutch of PF-AR NW2 was completed in May 2004. An X-ray pulse selector, a Ti:sapphire femtosecond laser (wavelength 800 nm, average output power 800mW, repetition rate 1 kHz) and a nanosecond Nd:YVO4 laser (wavelength 532 nm, output power 50 mW) were installed. Another pulsed light source, a femtosecond fiber laser (508 MHz repetition, 1500 nm) was purchased in March 2005. These lasers allow us to perform various types of pump-probe experiments using femtosecond or nanosecond lasers at different wavelengths.

Figure 2
The experimental hutch (green) and laser hutch (black). The hutches are connected by a black pipe for transport of the laser beam.

NW14 beamline

A new beamline, NW14, for time-resolved X-ray diffraction/scattering experiments is under construction. The current status is described elsewhere in this volume (See pp. 69-70).

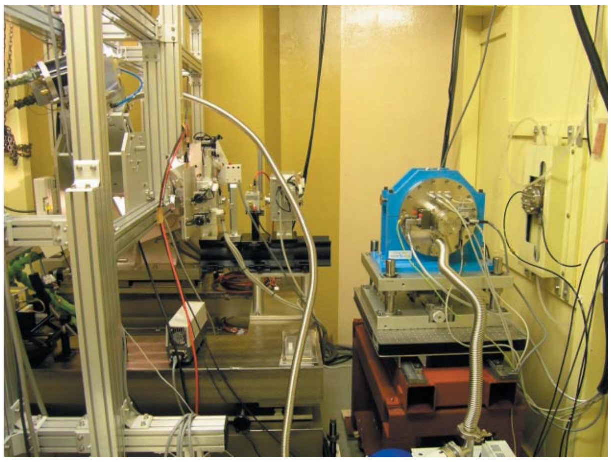

Figure 3
In the experimental hutch, the repetition rate of the X-ray pulses is reduced by a mechanical chopper (X-ray pulse selector) (blue) from 794 kHz to 945 Hz to match the repetition rate of the laser