

Time-resolved X-ray diffraction at the Photon Factory Advanced Ring (PF-AR) to Probe Photo-Induced Phase Transition in Organic CT Crystals

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Time-resolved X-ray diffraction technique using synchrotron radiation sources has been successfully applied to various dynamics studies of condensed matters (organic materials [1], protein crystals [2], and liquids [3]). For such experiment that uses two-dimensional area detectors to record a series of time-dependent diffraction patterns, isolation of a single X-ray pulse from X-ray pulse train is a must, because such two-dimensional area detectors (CCD or imaging plate) do not have fast gating capabilities so far. Therefore, in order to isolate a single pulse by using a fast rotating shutter, relatively sparse bunch-filling mode (single-bunch or hybrid mode), which most synchrotron users dislike for its low ring current and short life time, is strongly needed for the time-resolved diffraction experiments.

The Photon Factory Advanced Ring (PF-AR) at the High Energy Accelerator Research Organization (KEK), Tsukuba, Japan is an unique 6.5-GeV storage ring operated only in single-bunch mode, which is deliberately designed for time-resolved X-ray studies [4]. The ring current is 60 mA at the revolution frequency of 794 kHz, and the life time is ~ 20 hours in the single-bunch mode. We have constructed an insertion device beamline, NW2, at the PF-AR that is designed for time-resolved X-ray diffraction and XAFS experiments. Primary scientific targets of the NW2 are systems which can be triggered reversibly by a laser pulse in condensed matters such as organic and inorganic materials, protein crystals, and liquids. In particular, photo-induced phase transition (PIPT) in molecular charge-transfer (CT) crystals is one of the candidates of our research. The remarkable feature of PIPT is its cooperativity, that is, the structural relaxation of the electronic excited state of a molecule causes large-scale photo-induced phase transformation toward new lattice, electronic order and physical properties. Time-resolved X-ray diffraction enables direct access to the dynamics of electronic, atomic and molecular motions in such systems. We are currently working on photo-induced insulator-to-metal transition of quarter-filled A_2B salts such as $(EDO-TTF)_2PF_6$ in picosecond-to-microsecond time scale.

The beamline has an undulator with period length of 40 mm, which covers an energy range of 5-30 keV with 1st, 3rd, and 5th harmonics, and is used as a tunable and intense

monochromatic X-ray source [5]. A double-crystal monochromator, which uses flat Si(111) crystals and has liquid nitrogen cooling system, is installed. The cooling system can handle the incoming heat load up to 300W. The mirror system consists of three mirrors, which is a bent cylindrical mirror for focusing, and a double-mirror system (two parallel cut-off mirrors) to reduce a contamination of the higher harmonics. The typical photon flux of the monochromatic beam is estimated to be $\sim 10^{12}$ photons/sec.

The timing control of the laser and x-ray pulse is based on the radio frequency master clock (508 MHz) that drives electron bunches in the storage ring. A high speed chopper (X-ray pulse selector, XPS) made by Forschungszentrum Jülich is used for isolation of a single X-ray pulse from X-ray pulse train. The XPS is synchronized at 946 Hz to a subharmonic ($1/537600$) of the radio frequency. The physical opening window of the chopper is 1.2 μ sec but when it is phased to select a single bunch in the PF-AR single bunch mode, the exposure time becomes ~ 100 psec duration of the x-ray pulse. One can thus produce a 946 Hz pulse train of ~ 100 psec pulses from X-ray pulse trains at 794 kHz emitted from PF-AR ring.

A femtosecond regenerative amplifier system seeded by a Ti:sapphire femtosecond laser is also operational. The seeding laser pulses are phase-locked to the reference signals of $1/6$ of the radio frequency master clock by controlling the cavity length of the laser externally. The jitter is about a few ps. Then the laser pulses are chopped, amplified by the regenerative amplifier system and synchronized with the XPS. Finally, light pulses with 150 fs duration and a few hundred μ J per pulse at 800 nm phase-locked to the X-ray pulses are delivered. Double pulse train of exciting laser pulses followed by probing x-ray pulses can be produced in which the relative delay time can be varied by the use of an optical delay line.

In the talk, current status of the time-resolved X-ray diffraction at the PF-AR and its application to photo-induced phase transition in organic charge-transfer crystals will be presented.

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