doi:10.1017/S1431927615006820 Paper No. 0603
Microsc. Microanal. 21 (Suppl 3), 2015
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Ultrabright Femtosecond Electron Sources: Ultrafast Structural Dynamics in Labile Organic Crystals

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The progress in the development of fs-structural probes during the last twenty years has been tremendous. Current ultrafast structural techniques provide the temporal and spatial resolutions required for the stroboscopic observation of atoms in motion. In regards to femtosecond electron sources, different compression approaches and ultra-compact designs have made possible the generation of ultrashort and ultrabright electron pulses. With an effective brightness only one hundredfold below that of fs-hard X-ray Free Electron Lasers, ultrabright femtosecond electron sources have revealed unprecedented results in the study of photoinduced ultrafast structural dynamics [1, 2]. A brief overview of field along with a recent femtosecond electron diffraction (FED) study of the photoinduced insulator-to-metal phase transition of organic charge-transfer salt $(EDO\text{-}TTF)_2PF_6$ [3] will be presented. Here, we implemented a low repetition rate (10 Hz) and ultra-bright femtosecond electron source in order to avoid cumulative heating effects, and obtain a movie of the relevant molecular motions driving this photo-induced insulator-to-metal phase transition. We were able to record time-delayed diffraction patterns that allow us to identify time-dependent changes over hundreds of Bragg peaks (see figure 1). Model structural refinement calculations indicate the formation of a transient intermediate structure (TIS) in the early stage of charge delocalization (during the initial 2 ps). The molecular motions directing the formation of TIS were found to be distinct from those that, assisted by thermal relaxation, convert the system into a metallic-like state on the 100-ps timescale.

In terms of design engineering, ultra-compact femtosecond electron diffraction setups still offer the most convenient and economic means to obtain 100-fs electron pulses with an areal electron density similar to that achievable by DC-RF compression schemes ($\approx 5\ e^-/\mu m^2$) [4-6]. The most compact and simplest

FED setup is comprised of a photocathode, an anode aperture and a cylindrical magnetic lens that is placed right after the sample [7]. N-body simulations show that the extracting DC electric field strength (E) plays a major role. There is a great reduction in the electron pulse duration when going from E = 10 MV/m to E = 20 MV/m. Reaching a DC field strength of 20 MV/m is challenging but possible [8], and under this condition such ultra-compact setup would enable sub-100 fs (FWHM) temporal resolution [9].

The presented findings illustrate the potential of ultrabright femtosecond electron sources for capturing, with atomic resolution, dynamical processes of relevance for the understanding of chemical reactions, phase transitions, and protein structure-function correlations.

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[9] Funding for this project was provided by the Natural Sciences and Engineering Research Council of Canada and the Canada Foundation for Innovation. This work was supported in part by a Grant-in-Aid for Scientific Research on Innovative Areas (grant number 20110006) and the Global Centre of Excellence (G-COE) programme for Chemistry from The Ministry of Education, Culture, Sports, Science and Technology in Japan and by Creative Scientific Research (grant number 18GS0208) from The Japan Society for the Promotion of Science.

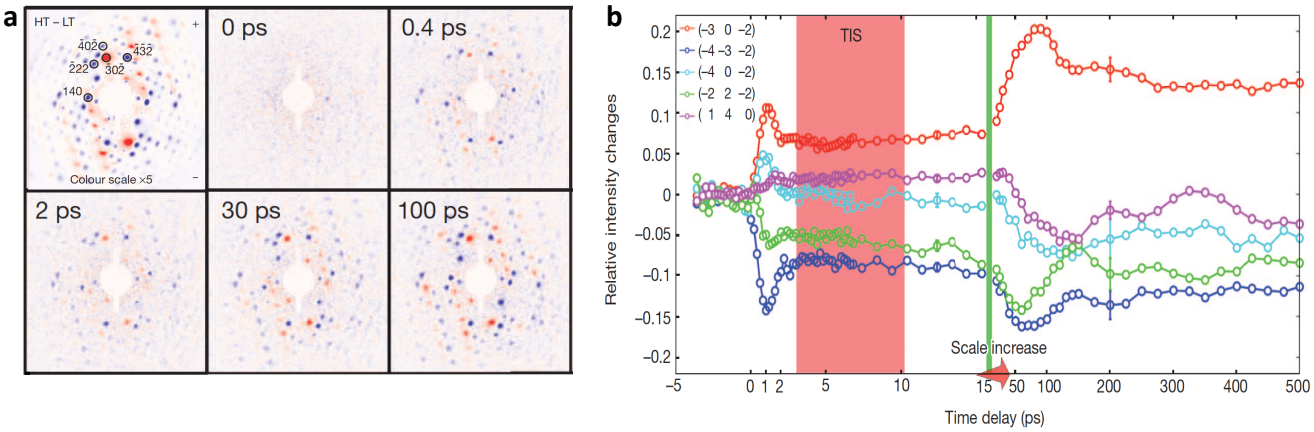

Figure 1. a) Top left panel: HT-LT denotes the difference between the diffraction pattern of high-temperature (HT) phase and that of the low-temperature (LT) phase. The rest of the panels show the difference between the diffraction patterns of the photoinduced and the initial LT phases as a function of the time delay between the optical excitation and electron probe pulses. **b)** Relative intensity changes for a few selected Bragg reflections. This figure was adapted from reference 3.