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### **Mapping molecular motions leading to charge delocalization with ultrabright electrons<sup>1</sup>**

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Ultrafast diffraction has broken the barrier to atomic exploration by combining the atomic spatial resolution of diffraction techniques with the temporal resolution of ultrafast spectroscopy. X-ray free electron lasers, slicing techniques and femtosecond laser-driven X-ray and electron sources have been successfully applied for the study of ultrafast structural dynamics in a variety of samples. Yet, the application of fs-diffraction to the study of rather sensitive organic molecular crystals remains unexplored. Organic crystals are composed by weak scattering centres, often present low melting points, poor heat conductivity and are, typically, radiation sensitive. Low repetition rates (about tens of Hertz) are therefore required to overcome accumulative heating effects from the laser excitation that can degrade the sample and mask the structural dynamics. This imparts tremendous constraints on source brightness to acquire enough diffraction data before adverse photo-degradation effects have played a non-negligible role in the crystalline structure. We implemented ultra-bright femtosecond electron diffraction to obtain a movie of the relevant molecular motions driving the photo-induced insulator-to-metal phase transition in the organic charge-transfer salt (EDO-TTF)<sub>2</sub>PF<sub>6</sub>. On the first few picoseconds (0 - 10 ps) the structural evolution, well-described by three main reaction coordinates, reaches a transient intermediate state (TIS). Model structural refinement calculations indicate that fast sliding of flat EDO-TTF molecules with consecutive motion of PF<sub>6</sub> counter-ions drive the formation of TS instead of the expected flattening of initially bent EDO-TTF moieties which seems to evolve through a slower thermal pathway that brings the system into a final high temperature-type state. These findings establish the potential of ultrabright femtosecond electron sources for probing the primary processes governing structural dynamics with atomic resolution in labile systems relevant to chemistry and biology. For more information vide-infra Gao et al., Nature 496, 343 (2013) and references there in.

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