

Transient charge density maps of ionic materials from femtosecond x-ray powder diffraction

Thomas Elsaesser

Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Max-Born-Str. 2a,
D-12489 Berlin, Germany, elsasser@mbi-berlin.de

Relocation of electronic charge plays a key role for functional processes in condensed-phase molecular materials. X-ray diffraction with a femtosecond time resolution allows for spatially resolving transient atomic arrangements and charge distributions [1]. In particular, time-dependent spatial maps of electron density have been derived from x-ray powder diffraction patterns measured with a 100 fs time resolution. In this talk, recent progress in this field will be discussed [2-5]. Crystals containing a dense array of Fe(II)-tris(bipyridine) ($[\text{Fe}(\text{bpy})_3]^{2+}$) complexes and their $(\text{PF}_6)^-$ counterions display pronounced changes of electron density that occur within the first 100 fs after two photon excitation of a small fraction of the complexes [3]. Electron density maps demonstrate a transfer of electronic charge from the Fe atoms and - so far unknown - from the $(\text{PF}_6)^-$ counterions to the bipyridine units. The charge transfer displays pronounced many-body features originating from the Coulomb interaction between the ionic molecular entities and affecting approximately 30 complexes around the directly excited one. As a second topic, electron relocations induced by strong external optical fields will be discussed [4,5]. This interaction mechanism allows for generating coherent superpositions of valence and conduction band quantum states and inducing fully reversible charge dynamics. While the materials LiBH_4 and NaBH_4 display electron relocations from the $(\text{BH}_4)^-$ ions to the neighboring Li^+ and Na^+ ions, LiH exhibits an electron transfer from Li to H. The latter is a manifestation of electron correlations and in agreement with theoretical calculations.

- [1] T. Elsaesser, M. Woerner, J. Chem. Phys. 140, 020901 (2014)
- [2] J. Weißhaupt et al., Nature Photonics 8, 927 (2014)
- [3] B. Freyer et al., J. Chem. Phys. 138, 144504 (2013)
- [4] J. Stingl et al., Phys. Rev. Lett. 109, 147402 (2012)
- [5] V. Juvé et al., Phys. Rev. Lett. 111, 217401 (2013)