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Ultrafast molecular processes mapped by femtosecond x-ray diffraction

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X-ray diffraction with a femtosecond time resolution allows for mapping photoinduced structural dynamics on the length scale of a chemical bond and in the time domain of atomic and molecular motion. In a pump-probe approach, a femtosecond excitation pulse induces structural changes which are probed by diffracting a femtosecond hard x-ray pulse from the excited sample. The transient angular positions and intensities of diffraction peaks give insight into the momentary atomic or molecular positions and into the distribution of electronic charge density. The simultaneous measurement of changes on different diffraction peaks is essential for determining atom positions and charge density maps with high accuracy. Recent progress in the generation of ultrashort hard x-ray pulses (Cu K_α, wavelength $\lambda = 0.154$ nm) in laser-driven plasma sources has led to the implementation of the powder diffraction and the rotating crystal method with a time resolution of 100 fs. In this contribution, we report new results from powder diffraction studies of molecular materials. A first series of experiments gives evidence of a so far unknown concerted transfer of electrons and protons in ammonium sulfate [$(NH_4)_2SO_4$], a centrosymmetric structure. Charge transfer from the sulfate groups results in the sub-100 fs generation of a confined electron channel along the c-axis of the unit cell which is stabilized by transferring protons from the adjacent ammonium groups into the channel. Time-dependent charge density maps display a periodic modulation of the channel's charge density by low-frequency lattice motions with a concerted electron and proton motion between the channel and the initial proton binding site. A second study addresses atomic rearrangements and charge dislocations in the non-centrosymmetric potassium dihydrogen phosphate [KH_2PO_4, KDP]. Photoexcitation generates coherent low-frequency motions along the LO and TO phonon coordinates, leaving the average atomic positions unchanged. The time-dependent maps of electron density demonstrate a concomitant oscillatory relocation of electronic charge with a spatial amplitude of the order of a chemical bond length, two orders of magnitude larger than the vibrational amplitudes. The coherent phonon motions drive the charge relocation, similar to a soft mode driven phase transition between the ferro- and paraelectric phase of KDP.