Ultra-fast light-induced structural order evidenced by time-resolved crystallography

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An important and fascinating feature of some photo-active materials is the possibility of tuning their optical, magnetic, conduction or other physical properties by light stimuli. This arises by virtue of photo-induced co-operative phenomena, where the structural relaxation of the electronic excited state following the absorption of a photon is not localized on one atom or molecule but entails a drastic structural distortion involving several electrons and atoms or molecules, yielding a transformed nano-domain. When a large enough number of nano-domains is created at the same moment, the material switches between two different macroscopic states, and a photo-induced phase transformationtowards a new lattice structure and electronic order. This opens the way to a new type of manipulation of matter by light, where for instance light can induce order, i.e. this differs from well-known light-induced disordering phenomena, such as surface melting or demagnetisation. This new type of photo-induced effects is exemplified by molecular charge-transfer materials that are readily tuned between competing neutral (N) and ionic (I) ground states. The photo-induced phase transformation in these materials, taking place on a ps time-scale, are highly co-operative and highly non-linear. The present birth of time-resolved photo-crystallography at synchrotron source gives a golden opportunity to directly observe the photo-induced structural change and the symmetry breaking by recording “complete” diffraction pattern with a ps time resolution. Here we present this direct evidence of the N-to-I photo-induced structural phase transformation, i.e. after photo-irradiation of the high-symmetry N phase, stable at high temperature. The monochromatic X-ray diffraction data establish unambiguously the ps time-evolution of the intensity of some Bragg peaks and the appearance of a new Bragg peak characterising the ferroelectric order in the photo-induced I state.