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Mainstream and alternative routes to photoinduced phase transitions

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The general excitement born out of the ultrafast clock caused rush of new ideas, new materials, and new instruments. One feat in particular has been draining a lot of effort, namely the control of materials with an ultrashort laser pulse. There is ample evidence now that materials can be directed between different macroscopic states by using appropriate electronic, or structural, excitations. The switching with a laser pulse of such materials can severely change their macroscopic properties (electric conductivity, magnetism, colour, etc.), whereby emerging cooperativity and coherence of different degrees of freedom underpin the resulting phase transitions of various sorts. However, the pertinent time scales for photo-switching processes in materials have been rather difficult to scrutinise. The pioneering investigations dealt mainly with the electron/phonon dynamics immediately following the femtosecond excitation, or the kinetics of recovery to the thermally stable states. Time-resolved X-ray diffraction and ultrafast VIS-IR spectroscopy reveal that the degrees of freedom triggered by a femtosecond laser pulse in a spin-crossover (SCO) material follow a sequence in the out-of-equilibrium dynamics. Those steps dissected in time, provided a mechanistic picture of a material transformation driven under different regimes (coherent or stochastic). The role of coherent optical phonons has been intensively investigated, whereas that of acoustic phonons and cell deformations, albeit looked upon, has not benefited from the same surge of effort. They involve propagation of a strain waves, essentially determined by sound velocity. The coupling between the strain wave and the order parameter field raises a challenging question whether such coupling can lead to material transformation. SCO crystals composed of bistable molecules where the cooperativity is mainly governed by the change of molecular “volume” between the two states provide an excellent test bed for addressing this question. We have investigated such materials over several time decades, and on samples of very different size, from nano- to macro-crystals. These studies bring the photo-switching of materials into new perspective, notwithstanding its common perception, uniquely related to electronic or optical phonon dynamics.

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