Disentangling electronic & structural dynamics with X-ray lasers for shining new light on ultrafast photoinduced transitions

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The advent of control science for directing matter and energy represents an important challenge for material science. It is now possible to control materials by light for generating remarkable properties on ultrafast timescale (ferroelecticity, conductivity, magnetism, photochromism…). These result from complex correlations between electronic and atomic constituents of matter. X-ray free electron lasers (X-FEL) open new possibilities for probing ultrafast photoinduced phenomena in order to disentangle, understand and control electronic and structural dynamics [1,2] (Figure 1). Ultrafast photoswitching in bistable molecular crystals is associated with a complex transformation pathway, multiscale in nature, where both molecular photo-switching (100 fs) and macroscopic elastic (ns) or thermal (µs) transformation of the crystal play their role [3]. We have studied the basic mechanisms allowing light to switch molecular materials between different magnetic states, by using femtosecond x-ray diffraction & absorption and optical spectroscopy. The stabilization of the photoinduced magnetic state results from the activation and damping of a molecular breathing mode. We gained experimental insights of this process, beyond the Born–Oppenheimer approximation, by disentangling the electronic charge-transfer excitation decay from the structural trapping dynamics [4-6]. We have demonstrated that in the active crystalline medium cooperative elastic effects can drive self-amplified and coherent response to light excitation [3]. The self-amplification process results from the elastic field induced by light and coupled to the molecular volume change, allowing the transformation of several molecules from a single photon.

X-ray free electron lasers broaden the range of methods for investigating ultrafast dynamics in mater, which will boost our capabilities to gather new insight into electronic and structural changes involved during light-induced phenomena.

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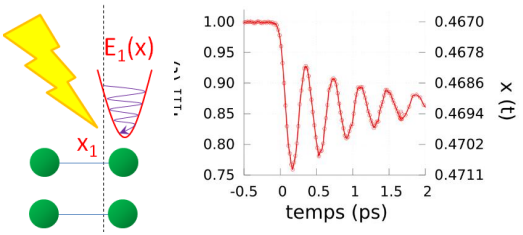

Figure 1 : Femtosecond laser excitation of Bi modifies instantaneously its potential energy surface and the equilibrium distance x between atoms, launching so coherent phonon. Ultrafast X-ray diffraction allows monitoring atomic motions in real time [1].