TOWARDS THE QUEST OF DISSIPATIVE STRUCTURES AND AUTOCATALYTIC EFFECTS IN THE PHOTO-INDUCED SPATIOTEMPORAL BEHAVIORS OF SPIN-CROSSOVER SOLIDS

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This phd thesis will take place in the PMFM (Physics of Multifunctional Materials) group of GEMAC laboratory.

Spin-crossover materials switching between low-spin (LS) and high-spin (HS) states are prototypes of bistable molecular solids having serious potential applications such as reversible high density memories, actuators, displays or mini-sensors that are sensitive to pressure, temperature, gases, mechanical strain etc. In particular, first-order phase transitions in molecular spin-crossover single crystals manifest themselves in a spectacular way when observed by optical microscopy. The thermally-induced transformation between the LS and the HS states (having different unit cell volumes) is accompanied by the emergence of the unique HS domain, characterized by the presence of a regular HS/LS interface, propagating at constant velocity for regularly shaped single crystals. This phenomenon, still escaping to deep understanding, is nicely monitored by optical microscopy thanks to the different colors of the HS and LS phases. Similarly to a tsunami, the transformation starts at some point, usually at the border of the crystal (a stochastic process) and propagates in a deterministic way.

The objective of this thesis is to conduct deep experimental investigations on the conditions of appearance of the transformation fronts, and to study their physical properties (structure, shape, width, speed, orientation etc.). The second part will be devoted to the quest of non-linear dynamics of the front interface, leading to the observation of self-oscillations under the influence of a strong but constant illumination, causing large thermal gradients in the crystal. Coupled with theoretical investigations based on reaction diffusion description of the spatiotemporal dynamics of the front propagation, we aim to clarify the experimental perquisites (light intensity, crystal and bath temperatures) leading to the emergence of self-organization of the spin states and autocatalytic behaviors of the interface dynamics. At the end, photo-induced spin state switching will be investigated at low temperatures < 30 K) with the objective to bring to light the possible existence of dissipative structures along the first-order nonequilibrium photoinduced phase transitions where, competing effects between photoheating, light absorption and light switching of the spin states may lead to morphogenesis phenomena.