Crystals make it possible to design chemical processes and reactions that cannot occur in liquids or in gases. They open the door for a deeper understanding of reaction mechanisms and for the exploration and design of solvent-free synthesis and applications in materials science. While molecules in crystals do not have the freedom of motion that is expected for chemical reactions to take place, reactions in crystals can be engineered in a reliable manner by taking advantage of a strategy used by nature, based on structures that can capture and guide the energy contained in sunlight. A few inspiring examples of such sunlight-induced photobiological processes include photosynthesis, DNA repair, and the biological compasses used by birds to guide their navigation across the planet. Similar to those systems, reactions in crystals rely on the capture of light to generate excited states that go on to form highly reactive intermediates whose fate is controlled by the crystal’s rigidity, homogeneity, and order. In this lecture I will describe examples on the use of crystals in complex chemical synthesis and the application of pulsed lasers to analyze the generation of triplet radical pairs consisting of two extremely close unpaired electrons formed by the cleavage of a single chemical bond, which causes their magnetic spins to be strongly entangled and potentially useful for applications in Quantum Information Science (QIS). Examples will also include a relatively exotic type of chemical reaction where a single photon, leads to many chemical events. Such “quantum chain reactions” are enabled by the strong energetic interactions between neighboring molecules, which leads to the transformation of photons into “excitons” and are helped by reactants that contain and unleash large amounts of internal energy.