The branch of chemistry that studies various reactions of chemical compounds triggered by irradiation with light is called **photochemistry**. Without any exaggeration, it can be said that each of us experiences photochemical effects on a daily basis. The very ability to see is due to the so-called photoisomerization process in which absorption of a photon by *cis*-retinal molecule causes its transformation to the all-*trans* retinal, which in turn influences the structural organization of rhodopsin, a biological pigment found in the rods of the retina. Photosynthesis is an excellent example of a photochemical process. Photochemical reactions are thought to be responsible for the creation of the first organic molecules which could spark the evolution of life on planet earth. Perhaps a more tangible example of photochemistry is that of 3D printing in which intense light is used to induce the photopolymerization process to manufacture objects with precisely controlled shape and functional properties. Photochemistry is also a very useful tool for chemists, since it allows one to perform chemical reactions that are not easily accessible with the wet-chemistry methods, or simply provide unique synthetic results (such as [2+2] photocycloaddition).

Chemists and physicists working in the field of functional materials such as metal-organic frameworks (MOFs) and coordination polymers (CPs) are interested in **photochemical reactions that can be performed post-synthetically on single crystals**. Single-crystal-to-single-crystal photochemical conversions are extremely interesting because it is possible to take ‘snapshots’ of the crystal structure with the use of X-ray diffraction experiments so that observation of what happens in a crystal is made with atomic precision. Moreover, photochemical reactions are effective in modifying the properties of MOFs and CPs: their crystal structure will change and this, in turn, can result in the modification of optical (e.g. emission of light), electrical or mechanical properties - to just name a few. In order to perform these photochemical reactions ultraviolet radiation is used because of its high efficiency – basically, all molecules that are capable of photoreaction will react. But what if we want to obtain a functional material that contains two kinds of photoreactive molecules but would like to trigger a photochemical reaction only of one, selected kind? How to “select” molecules which we want to perform a photochemical transformation while leaving other photochemical molecules intentionally intact? What are the physicochemical properties of such unusual systems? The traditional solution which employs ultraviolet radiation is not helpful in that regard.

PhotoReactMat2 project proposes a new approach to the design of photoreactive functional compounds which draws from nonlinear optics. Nonlinear optics, the optics of ultra-high light intensities is governed by different rules than conventional, linear optics. One of the main nonlinear optical processes is **two-photon absorption (2PA)**. It is a known fact that 2PA has different requirements for the structure of absorbing material compared to linear absorption (i.e. absorption of low-intensity light). For example, 2PA is strongly related to the electronic conjugation of molecules. This fact is important because it follows that chemists, through the design of chemical molecules, can “indicate” which molecules are stronger two-photon absorbers and should preferentially undergo a photochemical reaction, and which molecules will not photoreact due to low two-photon response.

In this project, the **synthesis of MOFs, CPs, and hydrogen-bonded networks containing two different photoreactive molecules will be performed and their 2PA-induced site-selective photoreactivity will be investigated**. By the combination of structural and spectroscopic results, we will **uncover the fundamental properties of this new nonlinear optical pathway to photoreactivity**. In particular, critical is the understanding of the kinetic differences between linear and two-photon triggered photochemical reactions. Another essential research direction is the exploration of **how the two-photon-induced photochemical reactions can affect physicochemical properties of crystalline materials such as second harmonic generation, photoluminescence, dielectric properties, or ferroelectricity.**

All in all, the outcome of this work will be crucial for the development of the new generation of two-photon photoreactive materials with unique characteristics and therefore will have far-reaching implications for the crystal engineering of such systems.