Quantum Crystallography-Driven Modular Design of Complex Co-Crystal Systems for Optoelectronic Applications (MolQCMat)

Crystals possess a multitude of fascinating **physical properties**, but their expression relies heavily on the careful selection of building blocks and on their arrangement, known as **symmetry**. By understanding this fundamental aspect, we can **harness unique effects in crystals**. For instance, **polar materials** can exhibit intriguing optical effects like **second harmonic generation (SHG), Pockels effect**, as well as **piezoelectric**, **pyroelectric**, and **ferroic properties**. However, symmetry alone cannot determine the magnitude of physical effects. Interestingly, molecules with significant dipole moments may aggregate to form **centrosymmetric structures, nullifying their polarity-driven effects**. Similarly, the aggregation of molecules can affect **fluorescence** and **absorption** properties. For example, many fluorescent dyes exhibit intensive emission in solution but experience quenching in the solid state due to unfavorable orientations of the crystal's building blocks. Recent years have seen significant advancements in **logic-based materials design**, primarily due to the development of **quantum crystallography tools** that enable us to control and predict interactions between molecules. In this project, we propose an **innovative approach using a redefined reverse crystal engineering method**, supported by quantum crystallography, to enhance the properties of materials. Our approach involves dividing crystal structures into subsections or **modules** that can be modified, removed, or exchanged. It is crucial to understand how these modules interact with one another to design smart devices. **Each module** can be a source of a distinct "property" (fluoro- chromo- or NLO-phores, chromogens, chiral components) which can be enhanced/utilised through an enforced arrangement of the building blocks in the crystal structure. Modules can range from molecular components to larger aggregates, essential to particular properties, and are connected through directional interactions. Using the **modular approach**, we aim to design **novel binary and ternary co-crystals suitable for optoelectronic applications**, including imaging and temperature sensing, optical modulators, and optical data storage devices. **Organic materials** play a vital role in various modern technologies, such as OLED, OLET, organic photodetectors, laser-driven medical diagnostics and telecommunication. However, there is still a need to improve their stability for long-life devices and enhance their efficiency. **Our project will focus** on two types of **materials with transferable properties** that can be modulated within the crystal structure. Firstly, we will explore NLO-phores, molecules with large hyperpolarizability, combined with components promoting noncentrosymmetric crystal structure formation. This combination allows us to tune linear and nonlinear optical properties, including **linear birefringence and SHG**. Secondly, we will investigate **chromogens or fluorophores** combined with co-formers that induce chromic effects, enabling selective changes in material colour based on external stimuli or **enhanced fluorescence quantum yield**. Finally, we will employ various experimental techniques and computational methods to achieve our goals in functional co-crystal synthesis. We will particularly emphasize the recent advancements in the field, **moving beyond traditional co-crystallization techniques** and incorporating **in-silico crystal design**. This holistic approach will provide insights into the properties of the resulting materials and establish correlations with the crystal structure. Ultimately, we aim to exchange one or more modules to further enhance the **electro-optic properties** of the studied crystals, pushing the boundaries of possible effects and opening up exciting new possibilities of applications.