Computational design of magnetic metal-organic frameworks using *ab initio* crystal structure prediction

Magnetic materials are used in a number of devices we sue every day: electric engines in home appliances and transport; data storage in smartphones and computers; medical devices such as MRI. All these different applications require magnets with different field strength, operating temperatures, size and weight parameters. Conventional magnets are most commonly based on inorganic materials, including transition metals and their oxides as well as rare-earth elements. While inorganic magnetic materials offer strong fields and room-temperature magnetization, their tuneability is generally limited. That is why, synthetic magnetic materials increasingly include organic components, either in the form of purely organic materials, or inorganic-organic hybrid systems.

In this project we are looking at the design of magnetic materials based on metal-organic frameworks (MOFs). MOFs are modular materials constructed from metal atoms connected by organic linker molecules. While metal atoms with unpaired electrons carry magnetic moment , it is the organic linkers that allow to control the distance between magnetic centres and their orientation, thereby influencing the overall magnetic performance of the material. However, it is by no means clear how to chose linkers which would produce MOFs with desired magnetic performance. That is because there are numerous ways how metals and linkers can arrange into a crystal structure, producing so-called polymorphs, which are materials with different magnetic behaviour.

It is possible to explore different packing arrangements experimentally and select the structures offering the kind of magnetic performance we are looking for. Yet, it may take a lot of work till a suitable structure is found in these kind of experimental trials.

The purpose of this project is to make the design of magnetic MOFs efficient by using a method of *crystal structure prediction* (CSP) to explore the variety of possible MOF structures on a computer, find which ones are predicted to offer desired magnetic properties, and subsequently synthesize those in the laboratory. This approach shall reduce the time and cost of magnetic MOF design, while also adding to our understanding of how these materials operate.