

Quantum critical refrigeration in molecular materials

Global demand for helium is around 170 mln cubic meters per year, especially in research, industrial, and medical facilities with the need to use liquid helium — 50,000 magnetic resonance imaging (MRI) systems are installed globally. Moreover, almost all helium today is a by-product of methane or natural gas production, thus increasing the carbon footprint. The world will **eventually run out of helium**, gradually increasing its costs, which is **dangerous for hospitals and progress in research and industry**. Hence, finding helium-free alternatives for cooling is an important task to address current challenges.

The project aims to investigate the **quantum spin liquids (QSL)** states and **magnetocaloric effect (MCE)** in strongly frustrated molecule-based materials for the demagnetization cooling processes. QSL is an exotic state of matter characterized by no magnetic ordering, highly entangled magnetic moments, and quasiparticles, which behave differently from the particles making up ordinary matter. QSL can be found only in frustrated magnets, i.e., when competing interactions between magnetic moments (pictured as tiny magnets) cannot be simultaneously satisfied in the material. For example, three magnetic moments in triangle corners would like to align antiparallel to each other. If two magnetic moments are already oriented antiparallel, upward, and downward, the third one would be unable to find an optimal orientation (Fig. 1, right).

In the MCE, the **material heats up or cools down while applying or removing a magnetic field**, respectively, which can be used in a helium-free thermodynamic cycle to reach low temperatures. An example of such a cycle is presented in Fig. 1. Magnetic cooling is up to **40% more efficient** than conventional gas compression solutions used in household refrigerators. Moreover, MCE-based cooling is free of fluorinated greenhouse gases with **no direct CO₂ emissions**, is nearly noise-free, and can be scaled without any loss in efficiency. In science, magnetic cooling is vital for **space applications and quantum technologies** as it allows us to obtain even sub-kelvin temperatures near absolute zero. Theoretical predictions indicate that the temperature reduction related to MCE is **significantly larger for strongly frustrated magnets** than for ordinary nonfrustrated magnets. Furthermore, QSL candidates are proposed to become **superior magnetocaloric materials**, especially near the point where materials undergo the transition to the QSL. This type of cooling is called **quantum critical refrigeration**, which is related to the abrupt temperature decrease near the transition point, which is much more significant than conventional magnetic coolants.

QSLs have been rarely studied in molecular magnetic materials. Molecular magnets are coordination compounds composed of two types of building blocks, **magnetic metal centers** and **ligands** (usually organic molecules), that can be designed in a wide range by selecting different combinations of building blocks during the synthesis. It resembles playing with blocks for children, where atoms or molecules take the place of blocks. Despite the promising potential of employing coordination chemistry to realize QSL, **only 10% of QSL candidates'** reports consider organic or molecular compounds. At the same time, molecular magnets are well-known for their pronounced MCE, especially in the cryogenic temperatures. Still, only a few reports on the frustration-enhanced MCE in molecular materials exist. The proposed project will fill a gap in this area.

The project's innovation is based on planned, systematic studies that can lead to the development of strategies to engineer and manipulate molecular systems with frustration-enhanced properties. The activities planned in the project will allow for the selection of a suitable combination of building blocks for QSL and the preparation of guidelines for further studies. The ultimate goal is to **find superior magnetocaloric coolants that will be low-cost, helium-free, and environmentally friendly**. Embarking on this topic in Poland holds excellent potential for pioneering research, as it remains largely unexplored within the country's scientific landscape. Exploring this field may propel scientific progress and pave the way for other researchers.

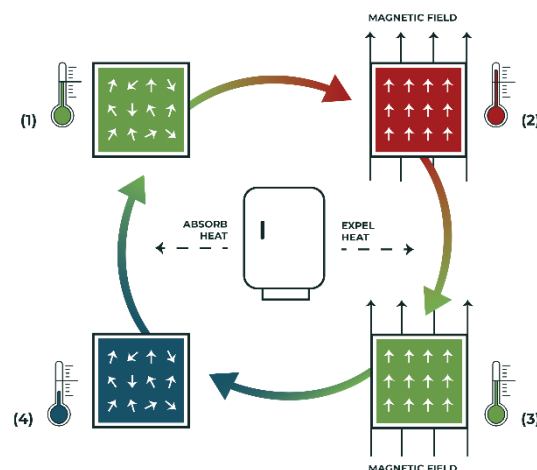
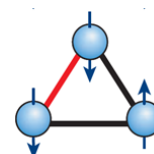


Fig. 1. Top: magnetic cooling principle¹:

- (1) heating the material,
- (2) expelling the heat,
- (3) cooling the material,
- (4) absorbing heat.



Right: magnetic frustration example².

¹ MagnoTherm®. Magnetic Cooling Cycle. <https://www.magnotherm.com/technology> (access: 01-03-2024)

² Balents, L. Spin liquids in frustrated magnets. *Nature* **464**, 199–208 (2010).