

PhD thesis – LLB

Low thermal conductivity mechanisms in rare-earth oxides

Understanding the parameters which determine the magnitude of thermal conductivity (κ) in solids is of both fundamental and technological interests. κ is sensitive to all quasiparticles carrying energy, whether charged or neutral. Foremost among these are *phonons*, the collective vibrations of atoms in crystals. Measurements of κ , however, have also identified more exotic carriers: emergent quasiparticles such as non-Abelian states that manifest in the fractional quantum Hall effect [1], or spinons in the antiferromagnetic Heisenberg chain [2]. In terms of applications, thermal properties of solids are at the heart of major social and environmental issues. The need, for instance, for highly efficient thermoelectric and thermal barrier devices to save energy has driven the quest for *low thermal conductors*. Over time, a range of strategies has thus been suggested to hinder phonon velocities and/or mean free paths: use of weak interatomic bonds, strong anharmonicity, nanoscale designs, or complex or disordered unit cells (see [3] for a review). Another promising concept to further impair the phonon mean-free path is based on magneto-elastic coupling.

Still in its infancy [3], this concept has emerged from the observation of a spin-phonon coupling in perovskites such as EuTiO_3 [4], but also in pyrochlores such as $\text{Tb}_2\text{Ti}_2\text{O}_7$ [5], garnets $\text{Tb}_3\text{Ga}_5\text{O}_{12}$ [6] or iridate compounds [7]. The magnetic excitations involved in the magneto-elastic coupling at play in those compounds are not standard magnons, but *low energy crystal field excitations* (CEF). Since the latter are local electronic excitations, they do not disperse and thus cannot be associated with propagating quasiparticles. In other words, they are not potential heat carriers hence do not contribute to κ , in contrast with dispersive magnetic quasiparticles like magnons. However, they can significantly reduce the phonon lifetime by opening a new scattering mechanism.

In addition, the CEF scheme is highly sensitive to the magnetic species involved, and to its chemical environment, which impacts the symmetry and strength of the ligand field. Secondly, CEF schemes are modified in a magnetic field of a few Teslas generally, making thermal conductivity κ in such systems naturally dependent on the direction and magnitude of the magnetic field.

The aim of the PhD thesis is therefore to investigate, both experimentally and theoretically, magnetoelastic coupling and its impact on thermal conductivity. The systems to be studied will be (but not restricted to) Tb perovskites, and will include high-entropy or entropy stabilized compositions, displaying glass-like thermal conductivity.

During this PhD you will:

- **Synthesize high-entropy crystalline samples**, in collaboration with ICMMO, a solid-state chemistry laboratory at Université Paris-Saclay
- **Carry out neutron scattering experiments**, to study crystal structures and lattice and magnetic dynamics
- **Perform comprehensive thermal conductivity and specific heat measurements** down to sub-kelvin temperature on state-of-the art experimental setups, at ICMMO or at ESPCI (Paris)
- **Confront experimental results with first principles calculations** to gain a better understanding of the microscopic mechanism responsible for the excitations spectrum and thermal conductivity observed in the experiments

The PhD work will be performed at the Léon Brillouin Laboratory (LLB), under the supervision of S. Petit and F. Damay, a laboratory known worldwide for its expertise in neutron scattering techniques. Beam-time on neutron spectrometers will be requested in large-scale facilities such as ILL (Grenoble, France), PSI (Villigen, Switzerland), NIST (Gaithersburg, USA), etc. Long standing collaborations with ICMMO (C. Decorse) and the Physics Institute of Collège de France (B. Fauqué), will provide access to sample synthesis and sub-kelvin thermal conductivity measurements, respectively. DFT calculations will be carried out in collaboration with A. Subedi at CPHT (CNRS/Polytechnique).

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