The investigation of phonon lifetime and thermal transport mechanisms in complex and disordered crystalline systems by means of inelastic neutron and X-ray spectroscopy

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Amphitheater Besson (Phelma Campus)

Jury :

Olivier Delaire, Associate Professor, Duke University, Rapporteur
Matthieu Le Tacon, Professeur des Universités, Karlsruhe Institute of Technology, Rapporteur
Virginie Simonet, Directrice de recherche CNRS, Institut Néel, Examinatrice
Jelena Sjakste, Chargée de recherche CNRS, Laboratoire des Solides Irradiées, Examinatrice

Abstract: Interest in the engineering of thermal process mechanisms has grown significantly in recent years, particularly for applications involving thermal functionalization of a material for use as a thermal barrier, thermoelectric converter, thermal diode, etc., which requires the control of thermal conductivity. The thermal manipulation and design of a material in turn requires knowledge of the fundamental transport properties of the elementary heat carrier particle, the phonon.

This is easier said than done, however, since models that would allow us to comprehend heat transport in complex materials are still under development, and because we lack systematic energy and temperature dependencies of experimentally measured phonon dispersions and lifetimes on a wide range of materials. This presents a barrier to our understanding of more complex and disordered systems, which are typically the ones needed to produce the thermal conductivity spectrum for the applications listed above. With the introduction of the many more atoms per unit cell and/or the disorder, we must now connect the dots between the effects of increasing structural complexity and defects on the phonon spectrum, and, ultimately, on thermal conductivity.

Within the context of overcoming these challenges, I present the phonon spectra in three families of materials that exhibit different types of structural disorder and complexity. These contributions include the inorganic type-I clathrate Ba7.81Ge40.67Au5.33, defined by its cage structure and many atoms per unit cell; the equimolar high-entropy alloy FeCoCrMnNi, represented by a simple and averaged monatomic FCC unit cell with significant chemical disorder; and cubic Yttria-Stabilized Zirconia, characterized by an extended and correlated defect structure brought on by the inclusion of oxygen vacancies. Towards this end, I have experimentally measured their phonon dispersions, lifetimes, and mean free paths using inelastic neutron and X-ray scattering techniques found at large-scale facilities.

My results confirm the use of the ab-initio self-consistent phonon method calculations for clathrates, emphasizing the importance of quartic anharmonic terms in our understanding of both the hardening of the lowest-lying optical branch with increasing temperature, and in the reproduction of the weak temperature dependence of the lattice thermal conductivity. The FeCoCrMnNi alloy study establishes the lattice dynamics of a random five-element alloy, demonstrating that the factor limiting phonon lifetimes is associated to force-constant fluctuations. Finally, I revisit the lattice dynamics of cubic Yttria-Stabilized Zirconia with higher resolution measurements, bringing new insight into the acoustic-optic interaction within the phonon spectrum, and to the linewidth broadening that results from the extended defect structure in this material.

Through each of my case studies, I provide the energy dependence of a specific type of phonon scattering mechanism. Then, by methodically detailing these features in each system, we can work upwards from their microscopic phonon properties to their macroscopic material properties, bringing us one step closer to understanding heat transport in complex and disordered crystalline systems.