Site-specific lattice dynamics in Sn-based chalcogenides thermoelectrics: an XAFS and far-infrared spectroscopic study

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Thermoelectric materials that can be used to convert heat into power or vice versa have been found in technological applications such as electrical refrigeration for solid-state detectors, power regeneration utilizing waste-heat from vehicles. The record of the figure of merit ZT value have been improved towards 3, a dreamland for decades of thermoelectric researches worldwide. The discovery of the record high ZT ~2.1 in binary SnSe single crystal stirred interests not only in technological applications and commercial marketing, but also in fundamental sciences, such as the anharmonicity in lattice heat transfer. Meanwhile, it was also discovered that quaternary compounds $Cu_2ZnSnSe_4$ compounds can be optimized to achieve a record high ZT up to 0.9, half of the value achieved in SnSe. The reason why the two Sn-based chalcogenides thermoelectrics exhibit distinctive ZT properties are not well understood. With better understanding of the structure and dynamics in the two materials, one can expect to design materials with building blocks (atomic chains, atomic pairs, atomic aggregates, etc.) bearing great potential for high thermoelectric performance.

By using the site-specific and local structural probe of X-ray Absorption spectroscopy and far-infrared/THz spectroscopy for the collective lattice excitations, we investigated the interplay of atomic structure, electronic structure and lattice dynamics in quaternary $Cu_2ZnSnSe_4$ and SnSe, respectively. The synergy of the two techniques can address the open issue regarding the local structural manipulation of the thermoelectric properties in a simple (binary SnSe) and complex (quaternary $Cu_2ZnSnSe_4$) compounds.

Moreover, the hierarchical functionalization of the heat/charge transport in micro to nano-scale structures are far beyond full understanding. For instance, the point defects, stack faults and grain boundaries, which hamper the charge transport, were found to be essential for scattering acoustic phonons thus suppressing lattice thermal conductivity, only if the scattering of the charge carriers is not dominant.

In this contribution, we outline the profound understanding of the lattice dynamics, pivotally influencing the thermoelectric performance, for the two systems. We illustrate the powerful tool for exploit the structure and dynamics of thermoelectrics as well as other functional materials.

This work was financially supported by the National Science Foundation of China (Grants no. U1532128) and the INFN&IHEP collaborative framework