Experimental report

17/02/2025

Proposal: 7-01-594 **Council:** 4/2023

Title: Phonon softening and lattice thermal conductivity in high entropy alloys of SnSe thermoelectrics: temperature dependent phonon softening

Research area: Materials

This proposal is a continuation of 7-01-508

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Samples: SnGeSeTe
Pb0.7Sn0.15Se0.15Te

Instrument	Requested days	Allocated days	From	To
PANTHER	5	2	15/03/2024	18/03/2024

Abstract:

Thermoelectric materials can contribute to be part of the alternative for a sustainable world, as they present the advantage of converting temperature differences into electrical power, mainly through the scavenging of waste heat with thermoelectric generators. A key limitation of thermoelectrics is their lattice thermal conductivity. Our goal is to understand and decrease the lattice contribution to the heat transport. Control of the phonon system, and phonon scattering, is crucial in improving thermoelectrics, and this is where neutrons can provide valuable insight unobtainable by other means. We will study the temperature dependent phonon spectrum of high entropy alloys based on SnSe, a recently discovered promising thermoelectric, compare with NPD and estimated phonon modes from ADP, and of unalloyed SnSe. In particular, we are interested in the tell-tale sign of the appearance of a softening low energy mode.
This proposal builds on the results [13] obtained in IN6, Experiment: 7-01-508, Instrument: IN6-SHARP from 11/10/2019 to 14/10/2019. The results were published in [12] as ¿High-Performance n-type SnSe Thermoelectric Polycrystal Prepared by Arc-Melting¿ in 2020

Experimental Report: Phonon softening and lattice thermal conductivity in high entropy alloys of SnSe thermoelectrics: temperature dependent phonon softening

Abstract

This study investigates the phonon properties of a high entropy alloy based on PbTe using inelastic neutron scattering (INS) at the Institut Laue-Langevin (ILL). Measurements were conducted on the PANTHER instrument with incident energies of 30 meV and 12.5 meV to explore the phonon density of states and dynamic structure factor. The phonon spectra reveal characteristic vibrational modes, phonon softening, and broadening effects that indicate strong anharmonicity. These results contribute to understanding the lattice thermal conductivity of PbTe-based alloys and their thermoelectric performance.

Experimental Report

The sample was a fine polycrystalline powder, synthesized at 850°C under 4 GPa pressure with nominal composition of $Pb_{2.5}SnSeTe_3$. It remained stable up to 600°C and was stored in a dry environment. Measurements were performed at low temperatures to obtain well-defined phonon spectra. Prior to data collection, the sample was conditioned in a vacuum furnace at 125°C for 4 hours to remove adsorbed moisture. The mass remained unchanged throughout the experiment, confirming its stability.

The phonon density of states (PDOS) was extracted from INS data, revealing key vibrational features of PbTe-based alloys. The dynamic structure factor $S(Q,\omega)$ was obtained for both incident energies, showing notable phonon contributions from alloying elements. Phonon softening was observed at low energies, consistent with previous studies on PbTe-based materials [1,2]. Broadening effects at higher temperatures suggest increased anharmonicity and phonon scattering, impacting thermal transport.

These results align with previous investigations of PbTe alloys, confirming the presence of low-energy phonon modes responsible for reduced lattice thermal conductivity. The study provides insight into vibrational dynamics, which are crucial for optimizing thermoelectric performance. Further theoretical modeling and high-temperature measurements will refine the understanding of phonon transport in these materials.

References

[1] Yi Zhang, Xhuezhi Ke, Changfeng Chen, J. Yang and P. R. C. Kent “Thermodynamic properties of PbTe, PbSe, and PbS:First principles study” Phys. Rev. B (2009) 80, 024304

[2] J. Gainza, F. Serrano-Sánchez, N. Biškup, N. M. Nemes, J. L. Martínez, M. T. Fernández-Díaz, J. A. Alonso “Influence of nanostructuration on PbTe alloys synthesized by arc-melting” Materials, (2019) 12, 3783-1-3783-15

[3] Delaire, O., Ma, J., Marty, K., May, A.F., McGuire, M.A., Du, M.-H., Singh, D.J., Podlesnyak, A., Ehlers, G., Lumsden, M.D., and Sales, B.C., "Giant Anharmonic Phonon Scattering in PbTe", Nature Materials (2011), 10, 614–619.

[4] Li, C.W., Ma, J., Lindsay, L., Bansal, D., Hong, J., Chiang, T.-C., and Delaire, O., "Phonon Self-Energy and Origin of Anomalous Neutron Scattering in SnTe and PbTe", Physical Review Letters (2014), 112, 175501.

[5] Stern, R., Thomas, S., Manley, M.E., Budai, J.D., and Delaire, O., "First-Principles Approach to Nonlinear Lattice Dynamics: Anomalous Spectra in PbTe", Physical Review Letters (2014), 113, 105501.

Figures

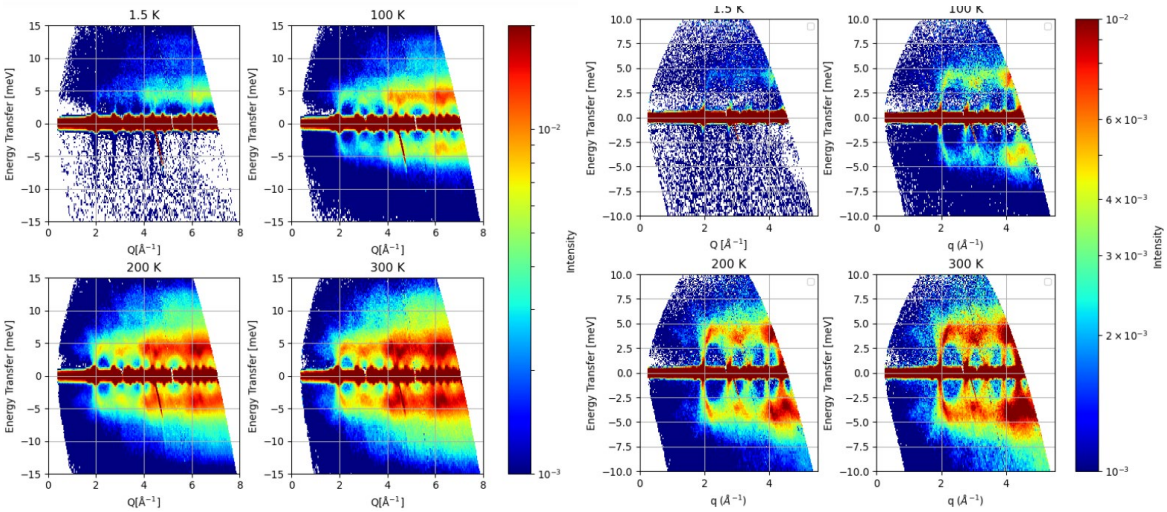

Figure 1: S(Q,ω,T) and elastic intensity with E_i = 30 meV (panels on left) and 12 meV (panels on right) at 1.5, 100, 200 and 300 K.

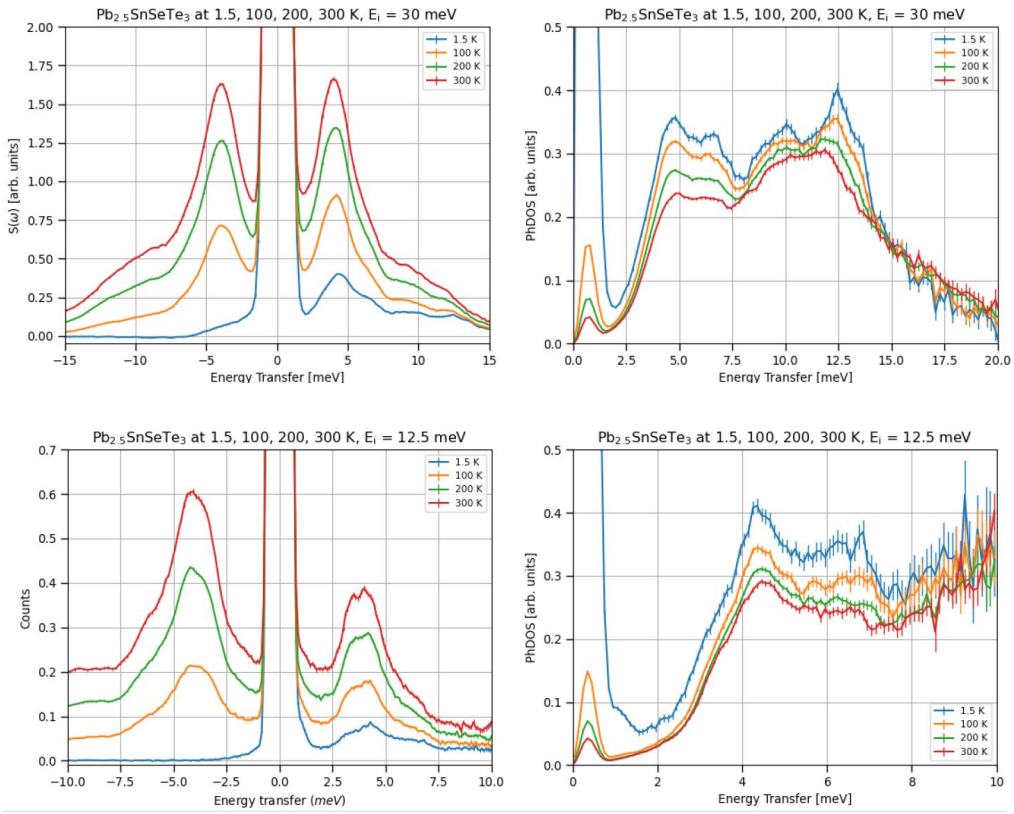

Figure 2: Dynamic structure factor (left) and phonon density of states (right) with E_i = 30 meV (panels on top) and 12 meV (panels on bottom) at 1.5, 100, 200 and 300 K.