Experimental report

18/02/2025

Proposal: 5-21-1199 **Council:** 4/2024

Title: Composition, structure and phonon softening in high entropy alloys based on PbTe thermoelectrics

Research area: Materials

This proposal is a new proposal

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Samples: PbTe
Pb2.5SnSbTe3
Pb2.5SnTe3Se
Pb2.5SnSbTe3Se

Instrument	Requested days	Allocated days	From	To
D20	2	0		
D2B	2	2	24/06/2024	26/06/2024

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 static disorder and the phonon system (phonon scattering) is crucial to improving thermoelectrics, and this is where neutrons can provide valuable insight unobtainable by other means. We have successfully prepared high entropy alloys based on PbTe, alloyed with Sn, Sb and Se. However, SXRD and STEM-EELS alone were not sufficient to characterize the actual composition of the cubic crystalline phase. We will compare them with high resolution low temperature NPD and estimated phonon modes from temperature dependent ADPs. The latter will also complement an imminent experiment on PANTHER on the temperature dependent phonon spectrum of high entropy alloys based on PbTe.
This proposal accompanies Experiment: 7-01-594, Instrument: PANTHER, near future

Experimental Report: Composition, structure and phonon softening in high entropy alloys based on PbTe thermoelectrics

Abstract

This study presents neutron diffraction measurements conducted on a $Pb_{2.5}SbSnTe_3$ sample to elucidate its crystallographic structure and phase composition. The experiment was performed at the Institut Laue-Langevin (ILL) using the D2B diffractometer. The obtained diffraction patterns were analyzed to determine lattice parameters, identify present phases, and assess structural distortions. The results contribute to a deeper understanding of the structural characteristics of PbTe-based alloys, which is essential for optimizing their thermoelectric properties.

Experimental Report

The $Pb_{2.5}SbSnTe_3$ sample was synthesized through a solid-state reaction method, involving high-purity elemental precursors. The homogenized mixture was sealed in a niobium, subjected to a temperature profile reaching 850°C under 4 GPA pressure, and then cooled to room temperature. The resulting ingot was ground into fine powder for neutron diffraction analysis.

Neutron diffraction experiments were conducted at the ILL on the D2B high-intensity two-axis diffractometer. The instrument was configured with a neutron wavelength of 1.3 Å, and data were collected over a 2θ range of 10° to 150° with a step size of 0.1°. Measurements were performed at various temperatures.

The diffraction data were analyzed using the Rietveld refinement method implemented in the FullProf software suite. Initial structural models were based on the rock-salt structure of PbTe, with considerations for possible substitutions and vacancies due to the presence of Sb and Sn. The refinement process yielded lattice parameters and phase fractions, providing insights into the sample's structural composition.

The analysis confirmed that the primary phase of the $Pb_{2.5}SbSnTe_3$ sample crystallizes in a cubic rock-salt structure, characteristic of PbTe, with lattice parameters slightly deviating from pure PbTe, indicating successful incorporation of Sb and Sn into the lattice. No secondary phases were detected within the sensitivity limits of the measurement.

The refined lattice parameter was found to be consistent with values reported in literature for similar compositions. The absence of impurity phases suggests a homogeneous solid solution, which is favorable for thermoelectric applications as it ensures uniformity in electronic and thermal transport properties.

The neutron diffraction patterns of the $Pb_{2.5}SbSnTe_3$ sample is presented in Figure 1:

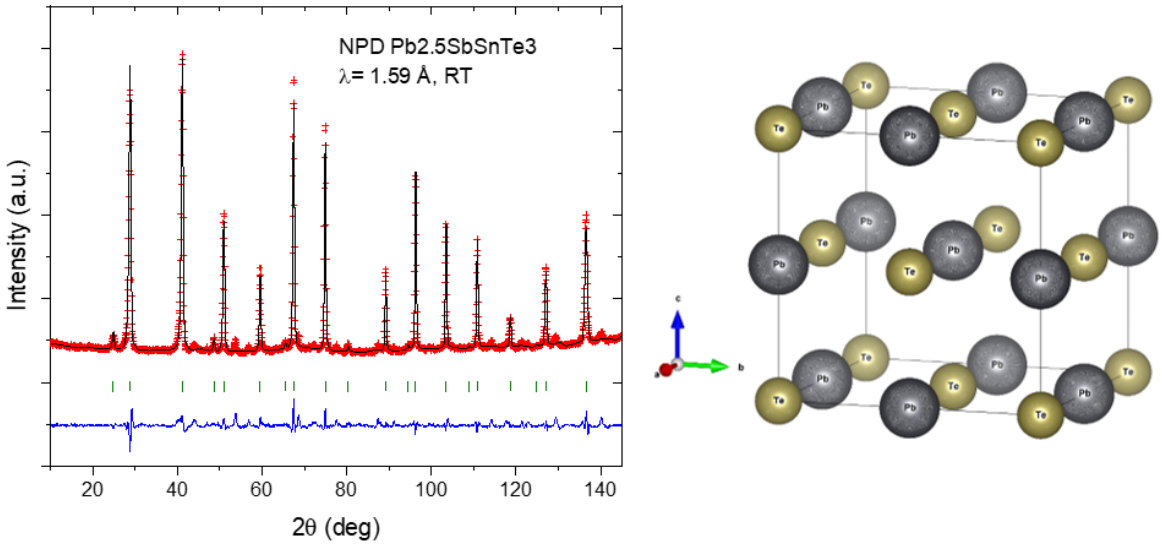

Figure 1: Neutron diffraction pattern of $Pb_{2.5}SbSnTe_3$ measured at room temperature and the corresponding refined structure.

These findings align with previous neutron diffraction studies on PbTe-based materials, such as the work by Jensen et al.[1], which reported on lattice dynamics and local symmetry breaking in PbTe. Additionally, studies on high-entropy alloys, like the in situ neutron diffraction investigation by Wu et al., have demonstrated the utility of neutron diffraction in elucidating complex structural behaviors in multi-component systems[2-4].

In conclusion, the neutron diffraction analysis of the $Pb_{2.5}SbSnTe_3$ sample reveals a single-phase cubic structure with lattice parameters indicative of successful alloying. These structural insights are crucial for understanding and optimizing the thermoelectric performance of PbTe-based materials.

References

1. Jensen, K. M. Ø., Božin, E. S., Malliakas, C. D., Stone, M. B., Lumsden, M. D., Kanatzidis, M. G., Shapiro, S. M., & Billinge, S. J. L. "Lattice dynamics reveals a local symmetry breaking in the emergent dipole phase of PbTe." *Physical Review B* **(2012)**, **86**, 085313.
2. Wu, Y., Liu, W. H., Wang, X. L., Ma, D., Stoica, A. D., Nieh, T. G., He, Z. B., & Lu, Z. P. "In-situ neutron diffraction study of deformation behavior of a multi-component high-entropy alloy." *Applied Physics Letters* **(2014)**, **104**, 051910.
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4. Stern, R., Thomas, S., Manley, M. E., Budai, J. D., & Delaire, O. "First-Principles Approach to Nonlinear Lattice Dynamics: Anomalous Spectra in PbTe." *Physical Review Letters* **(2014)**, **113**, 105501.