Experimental Report

Proposal: **7-01-365** **Council:** 10/2012

Title: Study of lattice dynamics of energy-harvesting thermoelectrics based on Mg2Si(1-x)Snx

This proposal is a new proposal

Researh Area: Physics

Main proposer: **PRYTULIAK Anastasiia**

Experimental Team: PRYTULIAK Anastasiia
RECOUR Quentin
BOURGEOIS Julie

Local Contact: KOZA Michael Marek

Samples: Mg2Si
Mg2Si0.75Sn0.25
Mg2Si0.5Sn0.5
Mg2Si0.25Sn0.75
Mg2Sn

Instrument	Req. Days	All. Days	From	To
IN6	5	5	30/05/2013	04/06/2013

Abstract:

In order to improve the thermoelectric (TE) performance semiconducting material their thermal energy transport should be minimized. The understanding of vibrational eigenstates, phonons, of the TE materials and their interaction is a fundamental step towards a thorough comprehension of heat transport. We carried out extensive ab initio calculations of phonon properties of bulk and doped Mg2Si solid solutions. Our extensive calculations require now a careful experimental checkup. As a first step our interest is focused on a series of Mg2Si(1-x)Snx specimen. For which the complete phonon spectra and lifetimes have been calculated. We ask for beamtime at the time of flight instruments IN4 and IN6 and for help with the computing of powder averaged spectra.

Experimental report

Experiment 7-01-365
Title: Study of lattice dynamics of energy-harvesting thermoelectrics based on $Mg_2Si_{(1-x)}Sn_x$
Experimental team: A. Prytuliak, J. Bourgeois, Q. Recour
Local contact: M. Koza

Abstract:
The Mg_2Si based alloys belong to the most promising group of materials for developing TE modules of high performance due to their low thermal conductivity (e.g. when alloyed with tin). The materials' low price together with non-toxicity and thermal stability are very important and attractive for industrial applications. In order to improve the thermoelectric (TE) performance semiconducting material their thermal energy transport should be minimized.
The understanding of vibrational eigenstates, phonons, of the TE materials and their interaction is a fundamental step towards a thorough comprehension of heat transport. An inelastic neutron scattering experiments together with *ab initio* computational work on phonons, including powder average lattice dynamics calculations (PALD) allowed us to get information on the vibrational dynamics of investigated solid solutions and their temperature dependencies.

Possibilities of the direct conversion of waste heat into electricity attract significant attention, specially now, when limits of classical energy resources are rapidly being reached. I was shown [1] that solid solutions in the system Mg_2Si-Mg_2Sn exhibit the lowest lattice thermal conductivity among Mg_2Si based materials due to the maximum difference in molecular mass. We carried out extensive *ab initio* calculations of phonon properties of bulk and doped Mg_2Si solid solutions. For which the complete phonon spectra and lifetimes have been calculated. The application of inelastic scattering from powder samples combined with the computational lattice dynamics gives a possibility to determine the powder average lattice dynamics (PALD) [2].

Inelastic neutron scattering were provided in ILL on time of flight IN6 instrument on 5 powder samples, with following compositions: Mg_2Si, $Mg_2Si_{0.75}Sn_{0.25}$, $Mg_2Si_{0.5}Sn_{0.5}$, $Mg_2Si_{0.25}Sn_{0.75}$, Mg_2Sn. Al the samples were analyzed from room temperature to 300°C, with Mg_2Si sample it was possible to go up to 600°C, as the sample is more stable. IN6 was used in configuration with wavelength of 4.14 Å.

Inelastic neutron scattering experiment on IN6 instrument in ILL allowed us to get multi-phonon corrected generalised densities of states (GDOS) for Mg_2Sn, $Mg_2Si_{0.25}Sn_{0.75}$, $Mg_2Si_{0.5}Sn_{0.5}$, $Mg_2Si_{0.75}Sn_{0.25}$, Mg_2Si at room temperature (Fig.1) and of the GDOS evolution at different temperatures (FIG.2). Based on the results of the experiment calculations are running in order to determine a level of order or disorder in the provided compositional range from Mg_2Si to Mg_2Sn.

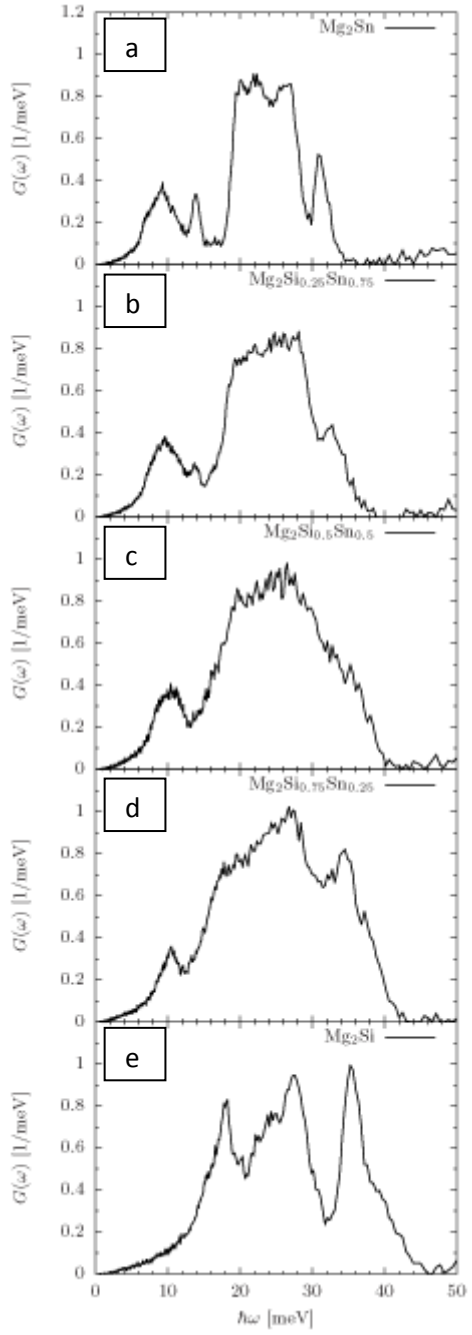

Fig. 1 Generalized phonon density of states (GDOS) obtained from IN6 spectra at room temperature from: (a) Mg_2Sn; (b) $Mg_2Si_{0.25}Sn_{0.75}$; (c) $Mg_2Si_{0.5}Sn_{0.5}$; (d) $Mg_2Si_{0.75}Sn_{0.25}$; (e) Mg_2Si.

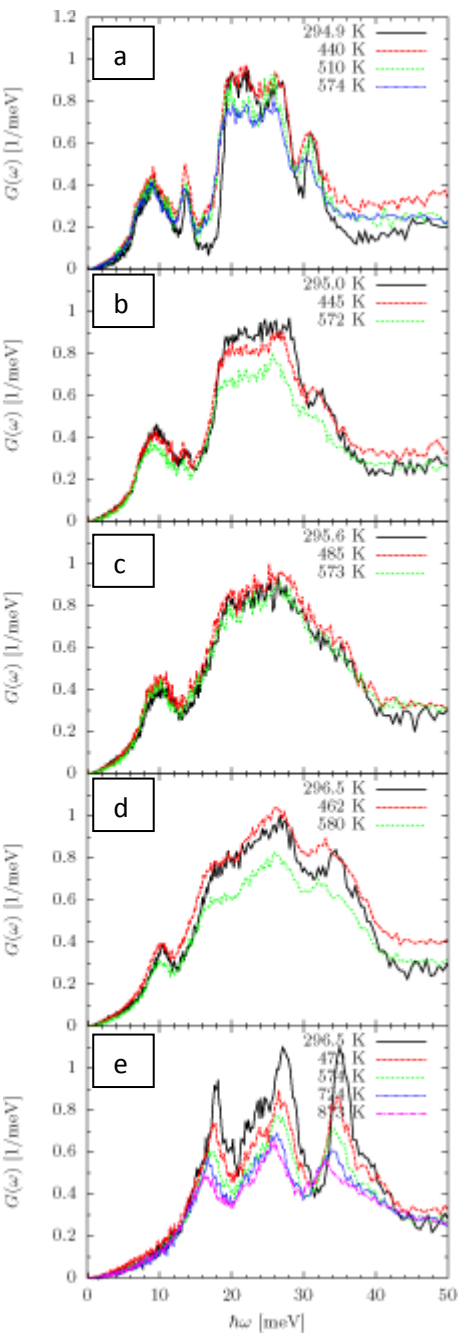

Fig. 2 Temperature dependence of GDOS obtained from IN6 spectra from: (a) Mg_2Sn; (b) $Mg_2Si_{0.25}Sn_{0.75}$; (c) $Mg_2Si_{0.5}Sn_{0.5}$; (d) $Mg_2Si_{0.75}Sn_{0.25}$; (e) Mg_2Si, temperatures are color-coded and reported in the figures.

We faced the difficulties to extract phonon lifetimes from the experimental results because of to the phonon renormalization due to the thermal expansion, which do not allow concluding about the lifetimes from the peaks broadening. We have pointed out a high level of disorder in the solid solutions, especially $Mg_2Si_{0.5}Sn_{0.5}$. Further data interpretation requires additional calculations for the solid solutions, both with initial assumption of the systems being ordered and disordered.

References:

1. Zaitsev, V. K. *et al.* Thermoelectrics of n-type with ZT > 1 based on Mg 2Si-Mg2Sn solid solutions. *ICT 2005. 24th Int. Conf. Thermoelectr. 2005.* 204–210 (2005).

2. Koza, M. M. *et al.* Vibrational dynamics of the filled skutterudites M1−xFe4Sb12 (M=Ca, Sr, Ba, and Yb): Temperature response, dispersion relation, and material properties. *Phys. Rev. B* **84,** 014306 (2011).