Experimental report

09/12/2018

Proposal: 7-01-446 **Council:** 4/2016

Title: Inelastic neutron scattering studyof the lattice dynamics of the homologous series (PbSe)5(Bi2Se3)3m (m= 1, 2 and 3)

Research area: Materials

This proposal is a new proposal

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Samples: Pb5Bi12Se23
Pb5Bi18Se32
Pb5Bi6Se14

Instrument	Requested days	Allocated days	From	To
IN4	0	2	02/07/2016	04/07/2016
IN6	8	5	31/08/2016	05/09/2016

Abstract:

The search for efficient thermoelectric materials has witnessed a resurgence of interest over the last two decades. An ideal thermoelectric compound should meet two conflicting requirements: the thermal transport should mimic that of a glass while the electrical properties should be those of crystalline solids. One possible strategy resides in choosing materials which show intrinsically low lattice thermal conductivity values. The homologous series (PbSe)5(Bi2Se3)3m (m = 1, 2 and 3) were shown to harbor a transition from trivial band insulator (m = 1) to bulk topological insulating properties (m = 2) by simply varying the number of Bi2Se3 layers and to exhibit extremely low lattice thermal conductivity values. We recently initiated a detailed study of these compounds with the aim of assessing the potential of these compounds for thermoelectric applications. In this context, we would like to investigate their lattice dynamics via inelastic neutron scattering experiments on IN6 between 100 and 500 K to shed light on the microscopic mechanisms that disrupt efficiently the heat-carrying phonons and on the influence of the number of Bi2Se3 layers on the thermal transport.

Experimental report 7-01-446 "Inelastic neutron scattering study of the lattice dynamics of the homologous series $(PbSe)_5(Bi_2Se_3)_{3m}$ (m = 1, 2 and 3)"

The vibrational dynamics of the homologous series $(PbSe)_5(Bi_2Se_3)_{3m}$ for m = 1, 2 and 3 have been investigated using the cold time-of-flight spectrometer IN6 between 50 and 500 K. Figure 1 shows a comparison of the generalized density of states $G(\omega)$ measured at 300 K.

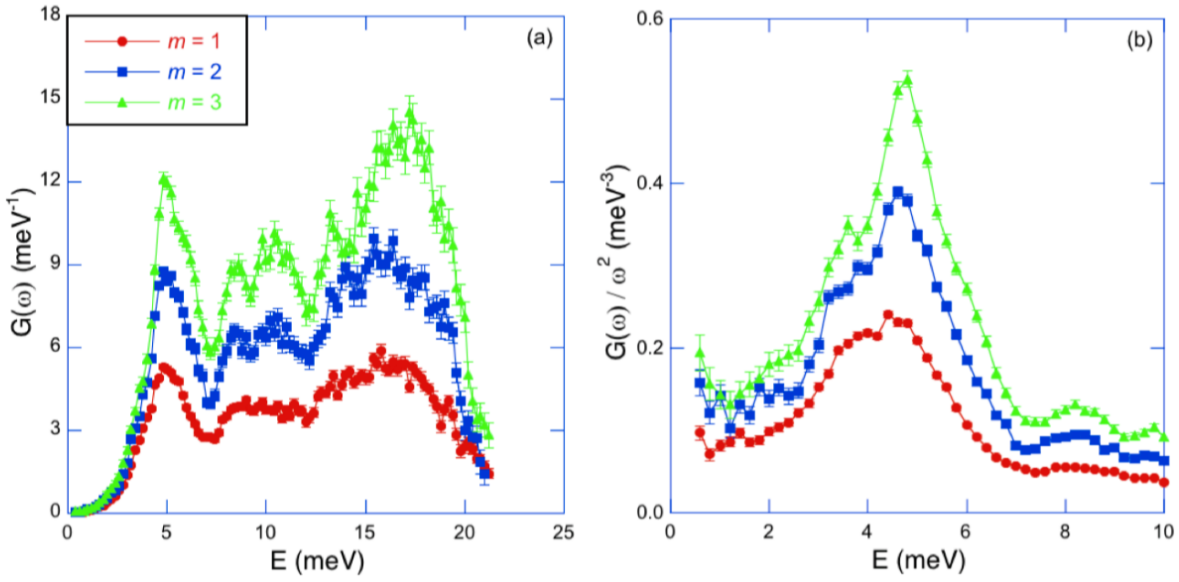

Figure 1: (a) Temperature dependence of the generalized phonon density of states $G(\omega)$ of $(PbSe)_5(Bi_2Se_3)_{3m}$ for m = 1, 2 and 3. **(b)** Debye plot of the data to stress the low-energy range.

We clearly observed an excess of vibrational density of states at low energies near 5 meV in all compounds. The fine details are, however, dependent on the number of Bi_2Se_3 layers.

Upon warming (Figure 2), this excess does not show a significant renormalization of its characteristic energy suggesting a quasi-harmonic behaviour, regardless of the number of Bi_2Se_3 layers. The lack of strong anharmonicity suggest that the peculiar evolution of the

lattice thermal conductivity with m (it increases despite an increase in the structural complexity of the unit cell) is rather tied to the number of interfaces between PbSe and Bi_2Se_3 layers.

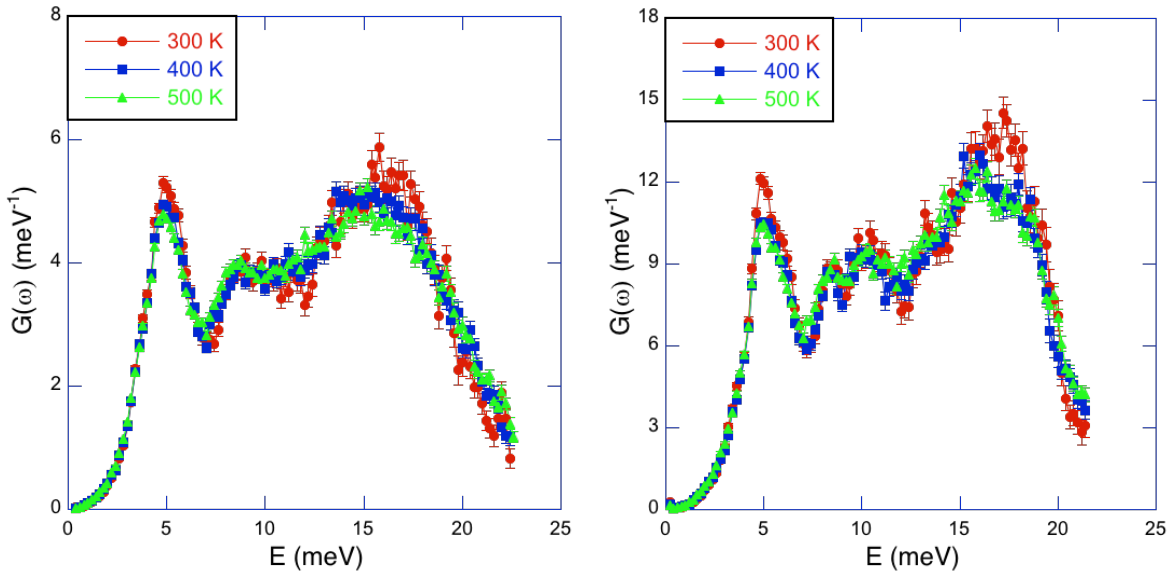

Figure 2: Temperature dependence of $G(\omega)$ for the m = 1 (left panel) and 3 (right panel) compound.

This work has been published in *Phys. Chem. Chem. Phys.* **20**, 14597 (2018).