Experimental report

03/09/2016

Proposal: 7-01-398 **Council:** 4/2014

Title: Inelastic neutron scattering studyof the lattice dynamics of the tetrahedrite compounds Cu12Sb2Te2S13 and Cu10Te4S13

Research area: Materials

This proposal is a new proposal

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Samples: Cu10Te4S13
Cu12Sb2Te2S13

Instrument	Requested days	Allocated days	From	To
IN6	5	4	04/09/2014	08/09/2014

Abstract:

The search for efficient thermoelectric compounds has witnessed a resurgence of interest over the last two decades. These materials 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 thermal conductivity values. Natural minerals known as tetrahedrite show thermal conductivity values close to the amorphous limit i.e. when the phonon mean free path is reduced to the interatomic distances. We recently initiated a study on the Cu12Sb4-xTexS13 system with the aim at tuning the thermoelectric properties through varying the Te concentration. Preliminary results show that this is indeed the case opening up new possibilities to reach higher thermoelectric efficiency. In this context, we would like to investigate the lattice dynamics of these compounds via inelastic neutron scattering experiments to shed light on the origin of their poor ability to conduct heat.

Experimental report 7-01-398 "Inelastic neutron scattering study of the tetrahedrite compounds $Cu_{12}Sb_2Te_2S_{13}$ and $Cu_{10}Te_4S_{13}$"

The vibrational dynamics of the two tetrahedrites of compositions $Cu_{12}Sb_2Te_2S_{13}$ and $Cu_{10}Te_4S_{13}$ have been investigated at the time-of-flight spectrometer IN6 between 50 and 500 K. Figure 1 shows the generalized density of states $G(\omega)$ measured at different temperatures for both compounds.

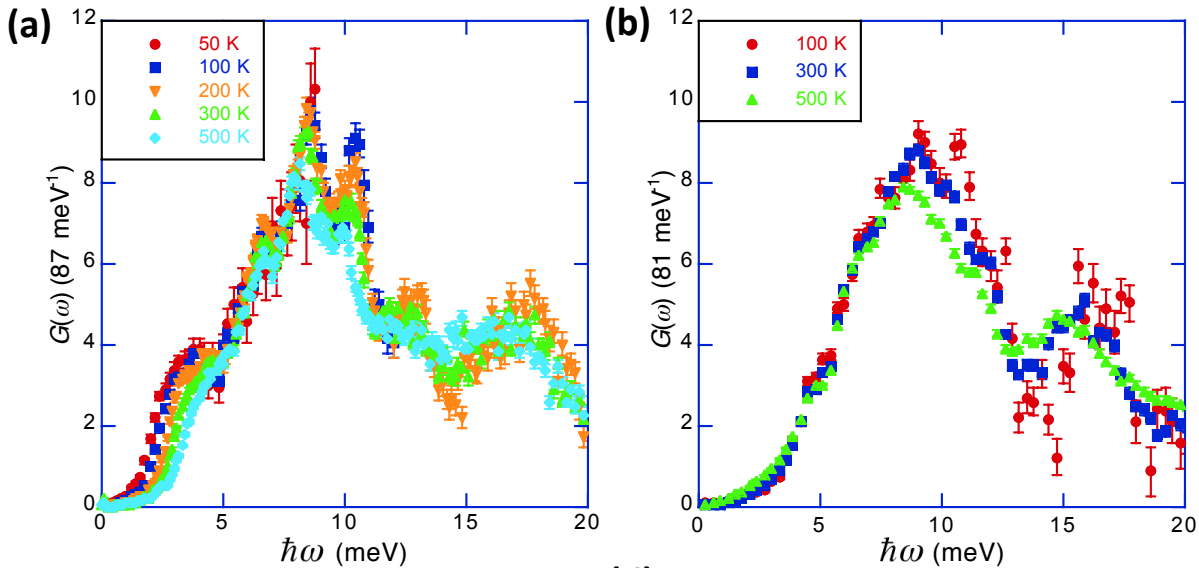

Figure 1: Temperature dependence of the generalized phonon density of states $G(\omega)$ of $Cu_{12}Sb_2Te_2S_{13}$ **(a)** and $Cu_{10}Te_4S_{13}$ **(b)** measured at different temperatures on IN6.

We clearly observed an excess of vibrational density of states at low energy in $Cu_{12}Sb_2Te_2S_{13}$, which is absent in the isostructural compound $Cu_{10}Te_4S_{13}$. Upon cooling, this excess shifts significantly towards lower energies evidencing a strongly anharmonic behavior.

Figure 2 shows the temperature dependence of the maxima of this excess. An estimation of the β parameter following the model proposed by Dahm and Ueda indicates that

the anharmonicity of this excess is similar to that observed in strongly-anharmonic compounds such as KOs_2O_6. The data taken on IN6 thus revealed the microscopic origin of the extremely low thermal conductivity observed in tetrahedrites. This work has been published in *Phys. Chem. Chem. Phys.* **17**, 19751 (2015).

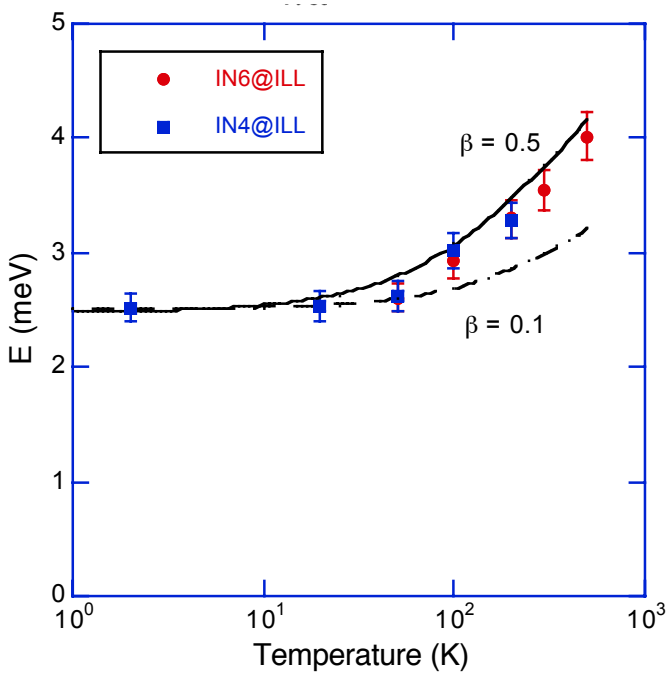

Figure 2: Temperature dependence of the maxima of the low-energy peak observed in the $Cu_{12}Sb_2Te_2S_{13}$ compound on a logarithmic scale. Data collected on IN4 are also added.