Experimental report

08/02/2016

Proposal: **7-01-409** **Council:** 10/2014

Title: Dispersions and lifetimes of low energy heat carrying phonons in the cage based materials Ba8Ge40.3Au5.25

Research area: Physics

This proposal is a new proposal

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Samples: Ba8Ge40.3Au5.25

Instrument	Requested days	Allocated days	From	To
IN5	7	2	07/07/2015	10/07/2015

Abstract:

In cage-based crystals, the guest-host interaction is widely accepted to be responsible for their exceptional low thermal conduction, opening new routes for the design of thermoelectric materials. We recently investigated the lattice dynamic of a Si-based [1] and Ge-based [2] clathrates and we demonstrated that the guest vibrations with the lowest energies act as a low pass filter for heat carrying phonons. A first step toward the understanding of the thermal conductivity in these materials would be to calculate the part of the conduction arising from the not filtered phonons. It requires the knowledge of their dispersions and lifetimes, a basic quantity, this latter, that is not accessible experimentally because of the finite sizes of the 4D instrumental resolution [1,2]. The high energy resolution setup of the IN5 cold TOF spectrometer offers an opportunity to overcome this limitation. Moreover, we grew a cubic centimeter single grain of the clathrate Ba8Ge40.3Au5.25 which has an exceptional crystalline quality (mosaïcity of 0.01°). In this proposal, we propose to map the dispersions of acoustic phonons at 300 K and 10 K in order to highlight the effect of the guest vibrations.

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Dispersions and lifetimes of low energy heat carrying phonons in the cage based materials $Ba_8Ge_{40.3}Au_{5.25}$

Clathrates are materials composed of covalently bonded polyhedra filled with guest atoms and are characterized by a low and weakly temperature dependent lattice thermal conductivity of typically $\kappa \sim$ 1-2 $W \cdot m^{-1} \cdot K^{-1}$ at room temperature. The understanding of this low lattice thermal conductivity, and its relation with the phonon properties of these complex systems is still a matter of debate.

We have shown recently that the low lying optical excitations related to the vibration of the guest atom lead to a filtering effect that limits the heat carrying acoustic phonons to a restricted q and energy range [1; 2].

One of the main challenges is to extract from inelastic neutron scattering the phonon lifetime, a parameter that is essential for a better understanding of the lattice thermal conductivity of clathrates.

First results have been obtained using a triple axis spectrometer, with a tight collimation. The interaction of the dispersion slope and the resolution ellipsoid, leads to a resolution of the order 0.2 meV only when the dispersion slope approaches the value of 10 meV/ $Å^{-1}$, whereas it is 0.8 to 1 meV when the slope is zero (unpublished results).

We have carried out a feasibility experiment on the IN5 time of flight spectrometer. The instrument was set up so as to: (i) reach the strong 600 Bragg reflections where the acoustic signal is well visible; (ii) achieve a resolution between 0.5 and 0.3 meV for the transverse acoustic modes below 10 meV.

The sample, a cm size single crystal with composition $Ba_{7.81}Ge_{40.67}Au_{5.33}$, was mounted with a (01-1) axis vertical, and data collected by rotating the sample by steps of 1° (0 to 30°) . The sample was oriented such that the TA and LA modes could be measured around the (600) Bragg peak.

The figure 1 displays the result LA and TA 2D maps, as obtained from a section of the 4D dataset. In this procedure data have been integrated in a slab of +- 0.04 $Å^{-1}$ in the direction perpendicular to the plane. The extracted scattering function S(Q,E) function has been multiplied by the normalization factor E^2 so that the acoustic mode intensity should remain constant.

Both LA and TA modes are well visible. The TA dispersion displays a gap at about 5 meV, as previously observed. The LA modes are well defined up to 6 meV where a gap is also observed.

Of special interest is the measured width of the acoustic phonon. Unlike the case of the triple axis, the resolution function depends essentially on two parameters: the phonon energy, and the slope of the acoustic branch. The latter one leads to a contribution to the broadening equal to DQxslope. The transverse resolution is equal to 0.03 $Å^{-1}$.

The figure 2 displays two constant q energy scans for the TA branch. The measured FWHM of the mode is equal to 0.38 and 0.3 meV respectively, i-e they are resolution limited. Impressively, the width of the longitudinal excitations is of the order 0.7 meV, again close to what is expected once the q resolution is taken into account. This latter results is especially encouraging, since LA modes are usually difficult to measure on a TAS experiment, due to the respective orientation of the resolution ellipsoid and the dispersion. The 0.7 meV resolution achieved close to the 600 Bragg peaks is at least 3 times better than what we measured on a TAS instrument.

In conclusion, the IN5 experiment opens up new possibilities for the study of phonon modes in complex systems. It allows an overview of the dispersion relation in different directions together with a very good phonon energy resolution equal to 0.3-0.5 meV.

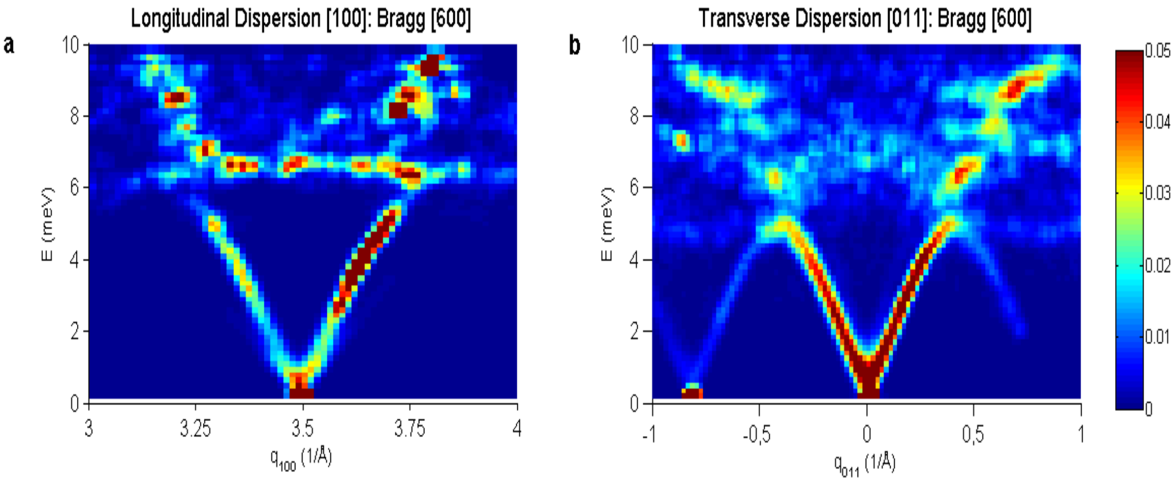

Figure 1: LA and TA spectra measured on IN5. The color code indicates the intensity of the measured signal. The maps have been obtained from integration of the 4D:(u,v,w,e) data with a step of 0.035 rlu or 0.02 $Å^{-1}$.

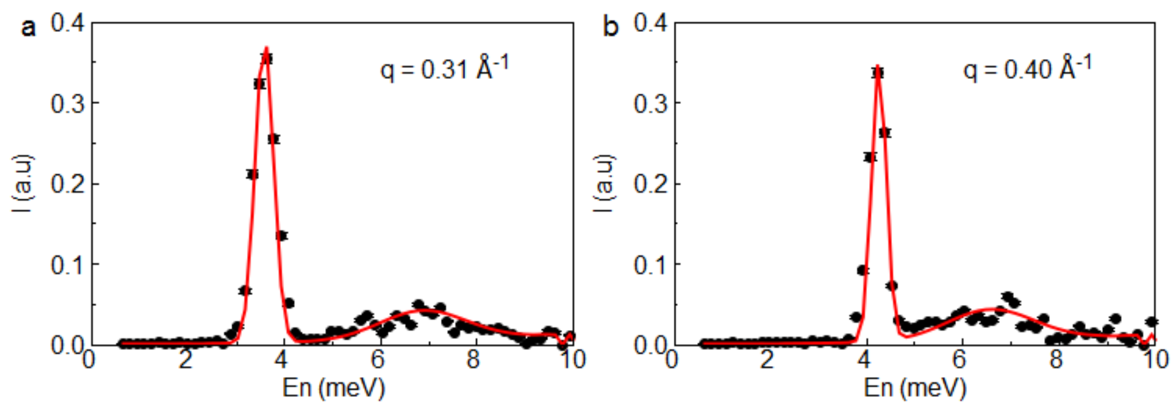

Figure 2 : Constant q, Energy scan extracted from the 4D data, for TA modes. These two wavevectors, the measured phonon width is resolution limited and equal to 0.38 and 0.3 MeV respectively.

1 . Euchner H, Pailhes S, Nguyen L T K, et al. 2012 Phys. Rev. B **86** 224303.
2 . Pailhes S, Euchner H, Giordano V M, et al. 2014 Physical Review Letters **113** 25506.