Experimental report

05/09/2019

Proposal: 7-01-492 **Council:** 10/2018

Title: Disentangling anharmonic from disorder dominated thermal transport inYttria stabilized cubic Zirconia

Research area: Physics

This proposal is a new proposal

Main proposer: Shelby TURNER

Experimental team: Shelby TURNER
Marc DE BOISSIEU
Valentina GIORDANO

Local contacts: Alexandre IVANOV
Andrea PIOVANO
Stephane RAYMOND

Samples: ((ZrO2)1-x(Al2O3)x, YSZ)

Instrument	Requested days	Allocated days	From	To
IN8	7	6	08/07/2019	11/07/2019
			10/09/2019	14/09/2019
IN12	7	0		

Abstract:

Yttria stabilized cubic zirconia (YSZ) is a ceramic of high technological interest, due to its ultra-low lattice thermal conductivity, large high temperature ion conductivity and thermal stability. We focus here on the measurement of the acoustic phonons lifetime. We first investigated at room temperature the energy dependence of the phonon lifetime and evidenced a crossover at q~0.25 rlu between a w^4 dependence at low-q (measured on 4f1@LLB), as expected in a disorder-dominated dynamics, and the w^2 dependence assumed to come from Umklapp scattering at higher q (measured on 1T@LLB). Surprisingly, we also observed a strong temperature dependence of the acoustic phonon lifetime in the w^4 regime, at odds with the observed glass-like temperature independence of the thermal conductivity. Thus, in the continuity of our work, we propose here to thoroughly investigate the temperature dependence of the lifetimes of phonons on both sides of the crossover over a wide temperature range [10 K:570 K]. We thus ask for 7 days on IN8. This work is part of the PhD thesis of S. Turner, focused on the phonon lifetime measurement for a microscopic understanding of thermal transport.

Quantifying the Effect of Chemical Disorder on Phonon Behavior of Yttria-Stabilized Zirconia

As stated in the original proposal, the goal of this experiment was to study the phonon lifetimes in Yttria-Stabilized Zirconia (YSZ) and find evidence for low-lying optical branches. Optical branches had not previously been measured experimentally for this material, even though they were predicted in theory (see original proposal for more information). This was mainly due to the resolution used in previous experiments. Triple axis spectrometers were used, but with a k_F of 4 Å^{-1}.

Now, due to better resolution, namely a k_F of 2.662 Å^{-1}, Si monochromator, and graphite analyzer, we are able to experimentally see the premature end of the acoustic dispersion due to low-lying optical branches. Since the flux of IN8 is well adapted for having good resolution on weaker signals like optical phonons, this became the main focus of the experiment. Measurements were done using our single crystal sample of ZrO_2 with 10 mol.% Y_2O_3 doping. The crystal was aligned into the [100][010] scattering plane.

Fig. 1 shows several energy scans taken of the transverse acoustic (TA) phonon dispersion along the [010] direction, polarized along [100], from the 200 Bragg. (Similar scans were done for the transverse dispersion along the [-110] direction, polarized along [110], from the 220 Bragg.) The TA mode stops dispersing at 9 meV, and we see three distinct optical branches at 17 meV, 25 meV, and 32 meV. The fact that the mode is cut off at 9 meV means that there is actually a low lying optical

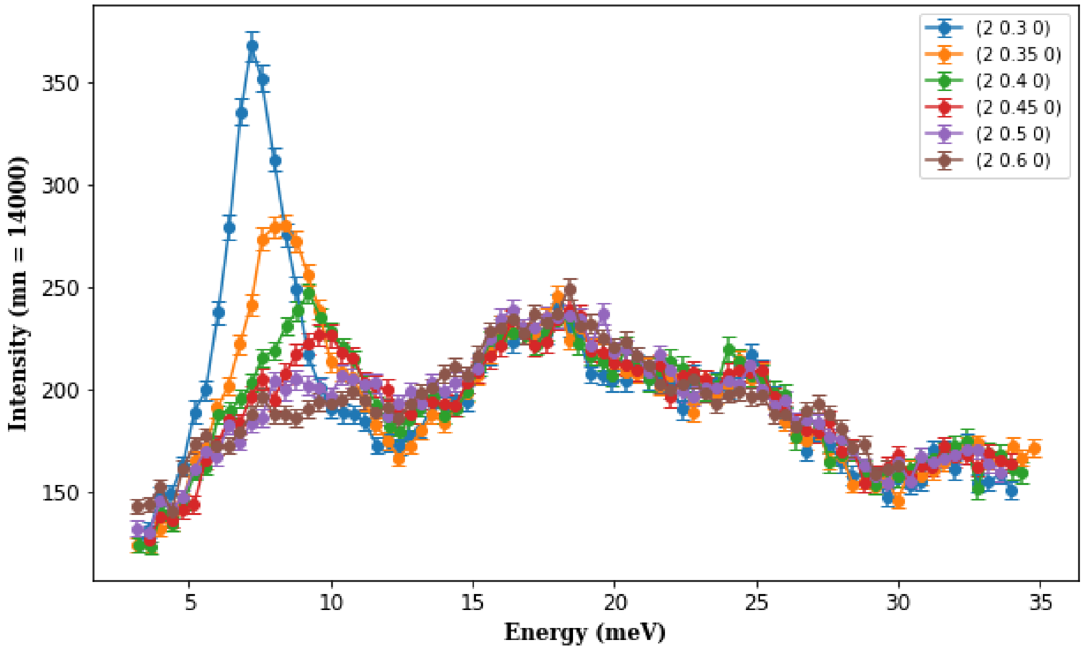

Figure 1: Raw energy scans of several q's of the transverse acoustic (TA) phonon dispersion. TA phonons are along the [010] direction, polarized along [100], stemming from the 200 Bragg in the scattering plane [100][010].

branch that intersects the mode at this energy as well, making there be 4 total optical branches that we could measure during this experiment. This branch at 9 meV in particular is evidence of the chemical disorder caused by doping pure ZrO_2 with Y_2O_3.

These findings change our understanding of the lattice dynamics of YSZ, as the transverse acoustic modes are not allowed to fully disperse across the Brillouin zone. Instead of dispersing until ~18 meV as was previously thought, the maximum energy of the dispersion is cut in half. Since the acoustic phonons account for the heat-carrying particles in a material, this means that we must find ways of describing the lattice thermal conductivity (κ_L) of YSZ in terms of this disorder, rather than simply by the dispersions previously used to calculate κ_L.