Experimental report

21/08/2017

Proposal: 7-01-440 **Council:** 4/2016

Title: Measurements of phonon density of states in Yb:CaF2 transparent ceramics

Research area: Materials

This proposal is a new proposal

Main proposer: Julia SARTHOU

Experimental team: Patrick GREDIN
Julia SARTHOU

Local contacts: Stephane ROLS
Michael Marek KOZA

Samples: Yb:CaF2 Transparent ceramic
Yb:CaF2 Single crystal

Instrument	Requested days	Allocated days	From	To
IN4	5	5	24/08/2016	29/08/2016
IN6	5	0		

Abstract:

In the past decade, transparent ceramics as solid-state lasers hosts have represented a major technological breakthrough in the field of laser materials. Ytterbium doped calcium fluoride (Yb:CaF2) represent one of the most promising compound for this application. Our team has recently developed a new fabrication process that leads to Yb:CaF2 transparent ceramics with excellent optical properties close to single crystals quality. Since thermal conductivity is an important parameter to take into account when it comes to laser host materials, we need to study the thermal properties of our ceramics and compare them with single crystal. We are currently performing thermal conductivity measurements on our ceramics and computing modelizations based on phonon scattering theory. However, we need to validate the relevance of our model with measurements of the phonon density of states of our ceramics and compare it with the single crystal. This kind of measurement can only be performed on a time of flight inelastic neutron scattering facility.

Experimental report

***Proposal**: 7-01-440*
***Title**: Measurements of phonon density of states in Yb:CaF_2 transparent ceramics*
***Experimental team**: Julia Sarthou, Patrick Gredin, Michel Mortier*
***Local contacts**: Stephane Rols*

The aim of this proposal was to compare the phonons density of states of Yb:CaF_2 transparent ceramics and single crystals, in order to validate the use of theoretical models based on single crystals to predict ceramics thermal conductivity. A secondary aspect of this work was to investigate the influence of the doping rate on the phonons density of states, using samples with various ytterbium doping rates.

The time-of-flight measurements were performed on transparent ceramics samples with four different doping rates – 1.5% at., 2% at., 3% at. and 5% at. – and one non-doped CaF_2 ceramic sample. In order to provide comparison material, single crystals samples with similar ytterbium doping rates – 1.7% at., 3% at. and 5% at. – were also measured. These last samples were crushed to a micronic powder in order to eliminate the crystal orientation anisotropy, while the ceramics samples remained under solid form. The experiments were carried out at T=320 K and λ=2.41 Å, and some samples were also measured at T=10 K and λ=1.1 Å. The samples were wrapped in an aluminium foil and inserted in a cadmium sample holder. For each sample, the acquisition time was comprised between four and ten hours depending on the sample mass.

Firstly, we compared the phonons densities of states measured at low (Stokes) and room (anti-Stokes) temperatures for the same sample. The results for a 1.5% at. ytterbium doped ceramic are displayed on Figure 1.

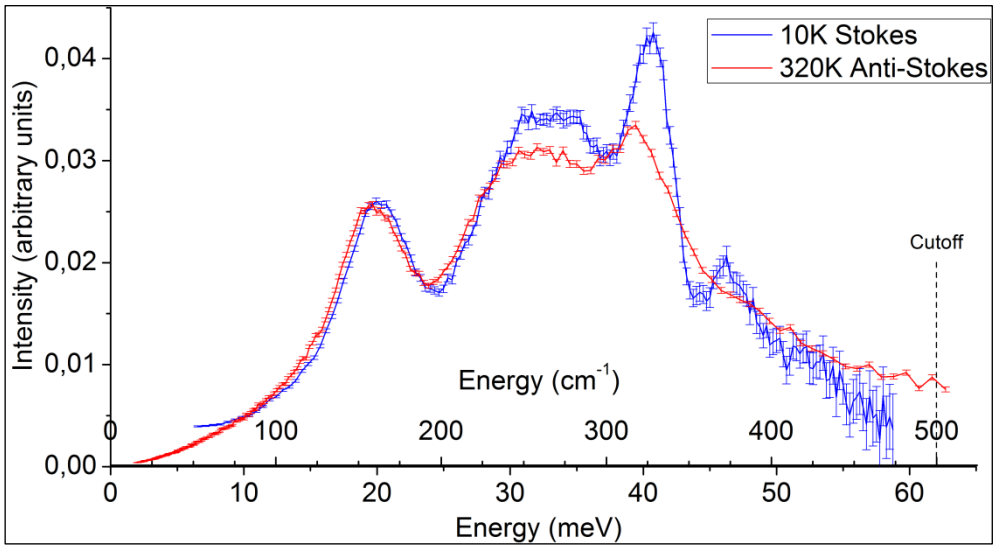

Figure 1: Phonons densities of states measured at low and room temperature for a 1.5%Yb:CaF_2 transparent ceramic.

The peaks occur at the same energies, except for the peak at 45meV on the Stokes spectrum, which is therefore considered as an artifact. However, the peaks relative intensities are not the same due to some differences in the experimental setup parameters.

The comparison of the phonons densities of states between ceramics and single crystals of similar doping rate does not show any important difference as one can see on Figure 2. This result validates the relevance of our theoretical modellings.

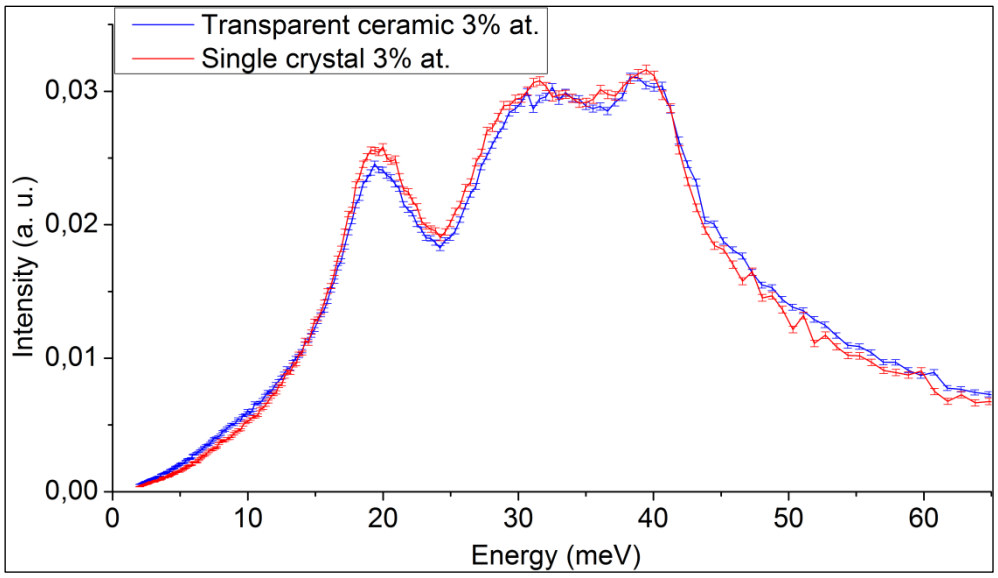

Figure 2: Phonons densities of states measured at room temperature for 3%at. Yb:CaF_2 transparent ceramic and single crystal samples.

Finally, the comparison of the phonons densities of states for transparent ceramics with different doping rates shows that there are some energy transfers going from higher energies to lower energies as the doping rate increases (see on Figure 3).

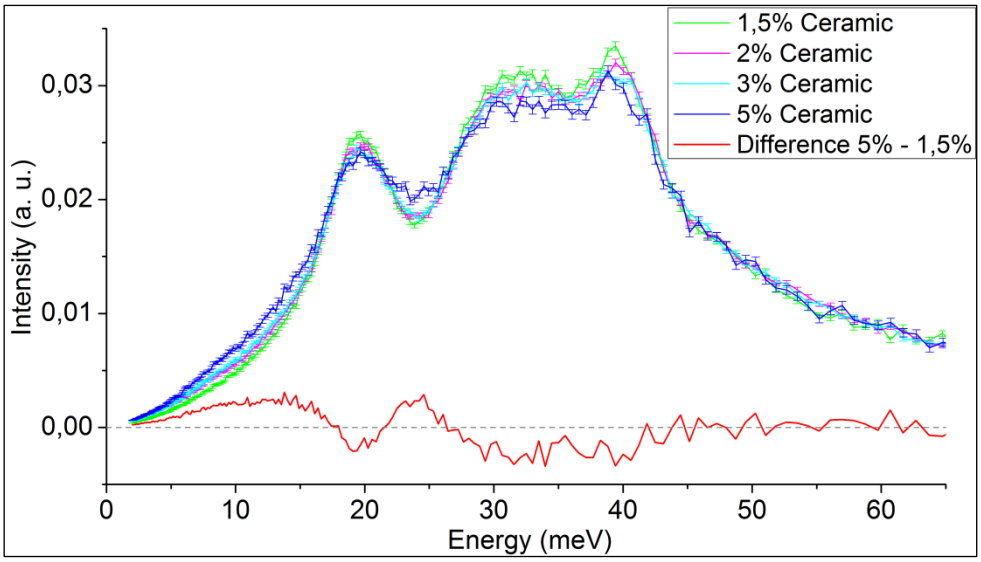

Figure 3: Compared phonons densities of states measured at room temperature for transparent ceramics with various doping rates.

This trend was confirmed with simulations also performed at ILL. Calculations are currently still running, but preliminary results tend to reproduce the experimentally observed effect of the introduction of ytterbium in fluoride calcium matrix (see Figure 4).

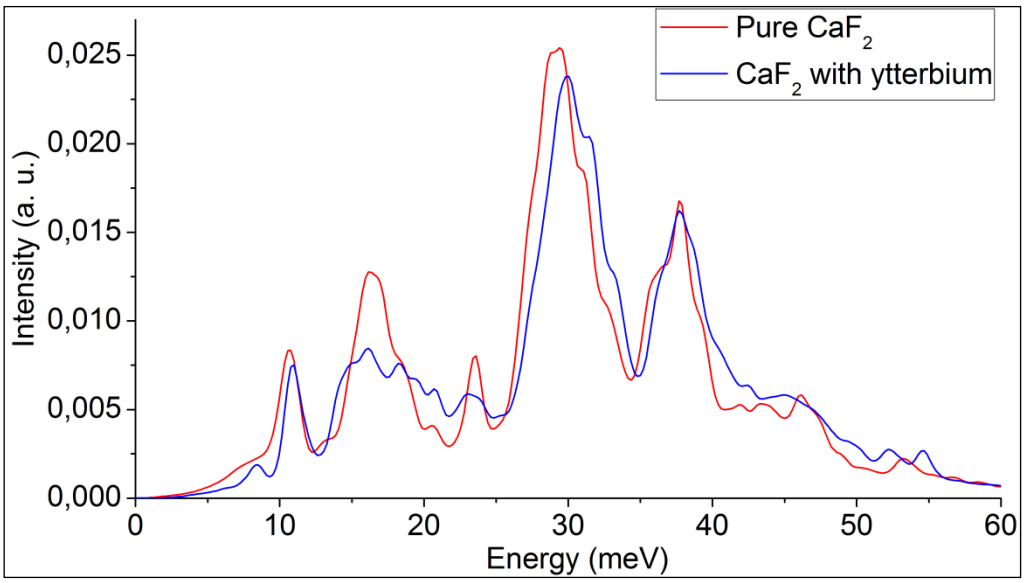

Figure 4: Phonons densities of states of pure and ytterbium doped CaF_2 calculated with molecular dynamics method.

Calculations were performed with direct method for non-doped material and obtained the same results as described in previous works from literature [1]. Molecular dynamics method was then used for doped material.

Some simulation works are still under study in order to identify a potential specific effect of the ytterbium clusters from the Yb:CaF_2 structure on the phonon density of states.

References

[1] K. Schmalzl, D. Strauch, and H. Schober, “Lattice-dynamical and ground-state properties of CaF2 studied by inelastic neutron scattering and density-functional methods,” *Phys. Rev. B*, vol. 68, no. 14, p. 144301, 2003.