Experimental report

14/02/2020

Proposal: 7-01-480 **Council:** 10/2018

Title: Phonon scattering and kagome modesin the thermal barrier material La2Zr2O7

Research area: Physics

This proposal is a new proposal

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Samples: La2Zr2O7

Instrument	Requested days	Allocated days	From	To
IN8	5	5	14/09/2019	19/09/2019
IN3	1	1	13/09/2019	14/09/2019

Abstract:

From next generation gas turbines to scavenging waste heat from car exhausts, finding new materials with ultra-low thermal conductivity has the potential to lead to large gains in device efficiency. Crystal structures with inherently low thermal conductivity are consequently desirable, but candidate materials are rare and often difficult to make. Using first principles calculations and inelastic neutron scattering we have studied the pyrochlore La2Zr2O7 which has been proposed as a next generation thermal barrier. We find that there is a highly anharmonic, approximately flat, vibrational mode associated with the kagome planes. Our results suggest that this mode is responsible for the low thermal conductivity observed in the pyrochlores. We now wish to test this with detailed measurements of phonon scattering rates to compare to beyond the harmonic approximation calculations.

Experimental report 7-01-480: Phonon scattering and kagome modes in the thermal barrier $La_2Zr_2O_7$

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The aim of this experiment was to measure the phonon lifetimes of a number of modes in the potential thermal barrier material $La_2Zr_2O_7$ (LZO). These experimental lifetimes would be used to test first principles calculations of the phonon lifetimes and test the hypothesis that low energy modes associated with the kagome planes of the pyrochlore lattice suppress the thermal conductivity.

Previous measurements on MERLIN at ISIS have shown that these modes exist and that the harmonic approximation predicts the overall energies and intensities of the modes well. These existing calculations were used to select promising locations to make energy scans at.

IN8 was set up with the Cu monochromator and PG analyser with no collimation. As optic modes were the primary focus q-resolution was not felt to be as important and the Cu-PG setup still gives a good energy resolution of 0.89 meV. A limited number of scans were also run with the Si monochromator to test for spurious scattering. The LZO crystal was aligned in the *hhl* scattering plane on an Al mount and a standard orange cryofurnace was used to control the temperature between 1.8 and 500 K. In total 18 energy scans encompassing approximately 20 optic phonons were measured at 1.8, 150, 300 and 500 K. Unfortunately, data at 500 K is extremely limited as the sample became loose on the stick and the a3 angle was no longer reproducible. Although this was discovered in time, the extreme activity of the sample (8.6 mSv) prevented us from reattaching it before the end of the experiment. The optic modes ranged in energy from 7 to 37 meV.

Initial fitting has focussed on modes measured at the Γ point. The kagome mode previously identified on MERLIN is shown in figure 1a while two other optic modes are shown in figure 1b. It has been assumed that the 2 K data is resolution limited. The fit to this 2 K data has then been convolved with a Lorentzian to extract the phonon-phonon scattering at other temperatures.

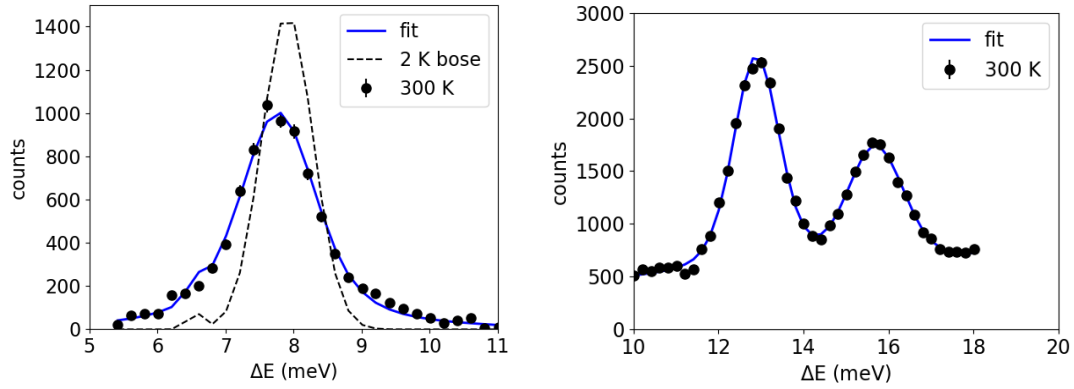

Figure 1: (a) The kagome mode previously identified on MERLIN measured at 660 and 300 K. The 2 K data has been scaled by the bose factor. The small peak at 6.5 meV is a spurion as confirmed by a measurement with the Si monochromator. (b) two additional phonons measured at the 118. The peak at 13 meV remains resolution limited while the 16 meV has some evidence of broadening.

hkl	E (meV)	Temperature (K)	Lorentzian HWHM (meV)	Phonon lifetime (ps)
6 6 0	7.8	150	0.23(2)	2.9(3)
6 6 0	7.8	300	0.38(2)	1.7(1)
1 1 8	13.1	150	0.001(7)	Unable to determine
1 1 8	13.1	300	0.01(4)	Unable to determine
1 1 8	15.8	150	0.06(2)	11(3)
1 1 8	15.8	150	0.21(1)	3.2(2)

Table 1: the widths and lifetimes extracted from the phonons shown in figure 1 so far.

Reassuringly the different modes analysed so far have different widths. One of the modes appears to remain resolution limited at both higher temperatures while the kagome mode broadens significantly faster than the others. These widths also agree with the limited fitting we were able to perform on the MERLIN data. The full list extracted so far is shown in table 1.

Thus far the calculations have proved extremely challenging although good progress has been made and we are optimistic that in the near future we will be able to compare the calculations directly.