|	**Experiment title:** Phonon lifetime and lattice thermal conductivity of MgO	**Experiment number**: ES-148
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Report:

The goal of this research project is the study of the acoustic phonon modes of MgO as a function of temperature. In particular, we are interested in the determination of the phonon energy and phonon width (the inverse of the life-time) and how they vary by varying the temperature.

We carried out inelastic x-ray scattering (IXS) measurements on MgO single crystals at ambient temperature and at T=1200 K. Specifically, we measured the longitudinal acoustic (LA) phonon dispersions along the (1,0,0) and the (1,1,0) direction and the transversal acoustic (TA) phonon dispersion along the (1,1,1) direction at ambient condition, while at 1200 K we measured both the LA and TA phonon dispersions along the (1,0,0), (1,1,0) and (1,1,1) directions. Spectra have been collected in transmission geometry, using the Si(12,12,12) instrument configuration. Beam spot at sample position was 280 x 90 μm. Specimens were cut with the (1,0,0) orientation perpendicular to the sample surface. The direction and size of the momentum transfer q were selected by an appropriate choice of the scattering angle and the sample orientation in the horizontal scattering plane. Orientation of the crystals on the spectrometer was accomplished using an in-line CCD camera. This allowed us to place the appropriate Bragg reflection in the horizontal plane of the spectrometer as well as defining the orientation matrix. From the collected diffraction patterns we also obtained the sample volume and were able to monitor the crystal quality via rocking curve widths.
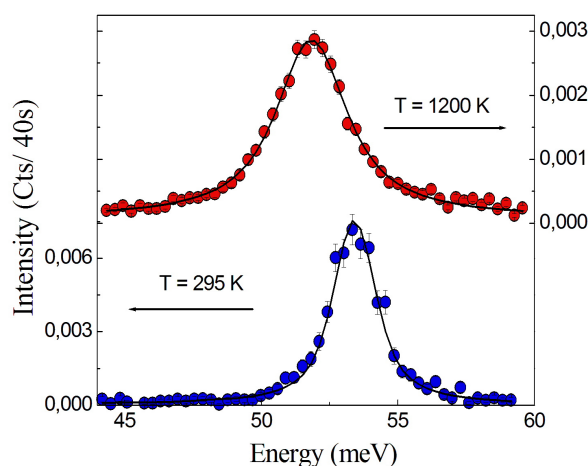
Figure 1: Longitudinal acoustic phonons in MgO along (1,1,0) direction at reduced q=0.6 measured at ambient temperature (blue points) and 1200 K (red points). Lines are fits to the experimental points.

The inelastic excitations have been fit by a Lorentzian function convoluted with the experimental determined resolution function, utilizing a standard χ^2 minimization routine. Data analysis is still in progress, but from the accurate line-shape analysis we have been able to determine both phonon energy and line-width. Examples of the data obtained at ambient conditions and at high temperature are illustrated in Figure 1. Measured line-shapes can be well fit by Lorentzian functions convoluted with instrumental resolution. The temperature-induced anharmonic broadening and shift are sizable.

In parallel to data analysis, we are performing calculations of the anharmonic phonon-phonon scattering coefficients of MgO within the density functional perturbation theory approach as implemented in the Quantum-ESPRESSO package [1]. The anharmonic phonon-phonon scattering determines the temperature dependence of the phonon width and of the phonon frequency. These calculations can be done fully ab-initio by using the so-called 2n+1 theorem [2,3]. Such a kind of calculations is very complex and has been done in literature very few times (see [2,3] and references therein), but the theoretician involved in the present proposal (M. Lazzeri) has a proven experience in this field. Preliminary calculations at ambient conditions of the phonon width are shown in Figure 2. The obtained phonon energies compare favourably with our experiments as well as previous theoretical work [4]. The detailed comparison of the calculated phonon width dispersions with measurements will allow validating theoretical treatment of anharmonic phonon-phonon scattering coefficients. Once validated, calculations will be used to i) extrapolate the results to extreme pressure and temperature conditions which are not experimentally accessible; ii) predict quantities which are not directly measured as the thermal conductivity.
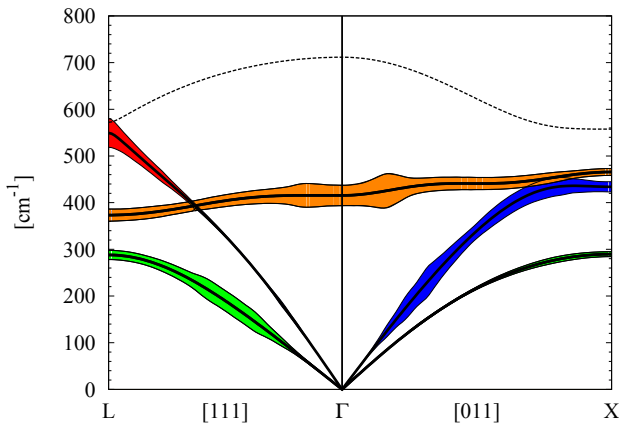
Figure 2: Calculated MgO phonon dispersion. The low energies phonon branches are represented with a variable-width filled band: the graphical width is equal to the respective anharmonic width expressed in cm^{-1} and multiplied by a factor 10. The anharmonic width is calculated ab initio with the approach described in [3].

References

[1] www.quantum-espresso.org

[2] Bonini et al., Phys. Rev. Lett. 99, 176802 (2007).

[3] Paulatto et al., Phys. Rev. B 87, 214303 (2013).

[4] Oganov et al., J. Chem. Phys. 118, 22 (2003); references there in.