Experimental report

Higher-order phonon scattering governing thermal conductivity in AlSb

Abstract:
Recently, it was shown theoretically that lattice anharmonicity due to the four-phonon scattering process is important to explain the significantly reduced lattice thermal conductivity in binary compounds, e.g., AlSb. However, experimental evidence for revealing the microscopic origin of higher-order anharmonic scattering in AlSb and other suggested compounds is missing. Here, we propose to use inelastic x-ray scattering to investigate the lattice dynamical properties of AlSb. Our focus will be on the behavior of the phonon lifetime deduced from the measured line widths. In comparison to theoretical predictions, we intend to identify the experimental evidence for the four-phonon scattering process, which could provide a novel approach to control thermal conduction in engineering materials for energy transfer, conversion, and storage application.

Generally, the three-phonon scattering process has been considered to govern thermal transport in solids, while the role of higher-order four-phonon scattering has been persistently unclear and so ignored. Recently, four-phonon scattering has been calculated to be strong in zincblende binary BAs reducing thermal conductivity from ~ 2200 to ~ 1400 W m^{-1} K^{-1} even at room temperature (RT) in agreement with experimental observations [1-3]. A similarly strong impact of four-phonon scattering has been reported to explain a ~50% reduction of thermal conductivity in binary zinc blended AlSb from the first principle prediction [4]. With the inclusive of the four-phonon scattering, the calculated κ values (c.f. Fig.1(c) from [4]) for AlSb are in reasonable agreement with experiment results, suggesting that four-phonon scattering is responsible for the discrepancies between calculations and experiments.

However, the experimental evidence of high-order anharmonicity of AlSb remains unclear. Herein, we used the inelastic X-ray scattering experiments on ID 28 beamline at European Synchrotron Radiation Facility (ESRF) to map out the phonon dispersion of AlSb and study the phonon linewidth of the acoustic and optical phonon modes at 100, 300, and 500 K. The measurements were performed using Si (9,9,9) monochromator and multi-analyzers, with a constant incident energy 17.794 keV, ~2.8 meV energy resolution and ~2.43 nm^{-1} monument resolution. As shown in Fig. 1(a), our measured single-crystal AlSb was 150 μm hor. × 10 μm ver. and had the in-plane high-symmetry directions, e.g., the [001], [110] directions.

Based on our preliminary ab-initio lattice dynamical calculations on the AlSb sample, we identified some specific Brillouin zones with high structural factors, e.g., (002) and (222) Brillouin zones. Firstly, we measured the sample at 300 K from -25 to 50 meV for covering the whole acoustic and optical phonon modes. As plotted in Fig. 1(b-c), we mapped out a significant part of the phonon dispersion in [001] and [110] high-symmetry directions. Our measured phonon dispersions are in reasonable agreement with our own calculated data as well as the published data [4], highlighting the good quality of our measured sample. Afterward, we used the Nitrogen cryo-stream system to access the desired temperature range, i.e. 100 and 500 K. We continued to scan the acoustic and optical phonon modes and repeated the scans at 100 K

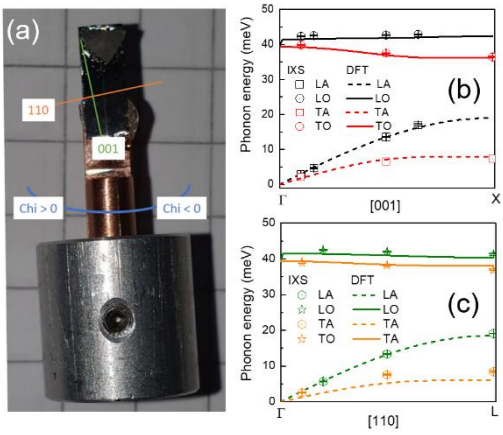

Fig.1(a) image of AlSb single crystal ([001] direction marked as a green line, [110] marked as an orange line); (b-c) the measured and calculated phonon dispersion along the [001] and [110] directions.

for obtaining enough statistical intensity. Moreover, we heated the sample up to 500 K and scanned the acoustic and optical phonon modes along with the high-symmetry directions.

We collected the temperature-dependent data of AlSb and analyzed the phonon linewidth of the respective acoustic and optical phonon modes. The raw data can be fitted with a damped harmonic oscillator (DHO) function convoluted with ~ 2.8 meV energy resolution, in combination with an elastic line and a flat background. As summarized in Fig. 2(a-b), the acoustic phonon modes hardened with evaluated temperatures, while the optical phonon modes softened due to anharmonicity. It is worth noting that the "abnormal" hardening signature might be from the diffusive signal of Al_2O_3 since AlSb was extremely sensitive to moisture. In addition, as shown in Fig. 2(c-d), analysis of phonon linewidth suggested the decreasing phonon linewidth of acoustic phonon modes and almost maintained phonon linewidth phonon modes at high temperatures. The inclusion of the acoustic modes and diffusive signal made it difficult to accurately extract the phonon energy and line width. Thus, we can only make an experimental conclusion on optical phonon modes. In summary, our IXS results identified a less anharmonicity effect than that of the calculated results and a less pronounced broadening phonon linewidth of optical phonon modes within our current energy resolution of ID 28. The rather high-energy resolution instrument, e.g., Raman scattering, may serve as a possible platform to examine the phonon linewidth from this high-order anharmonicity effect.

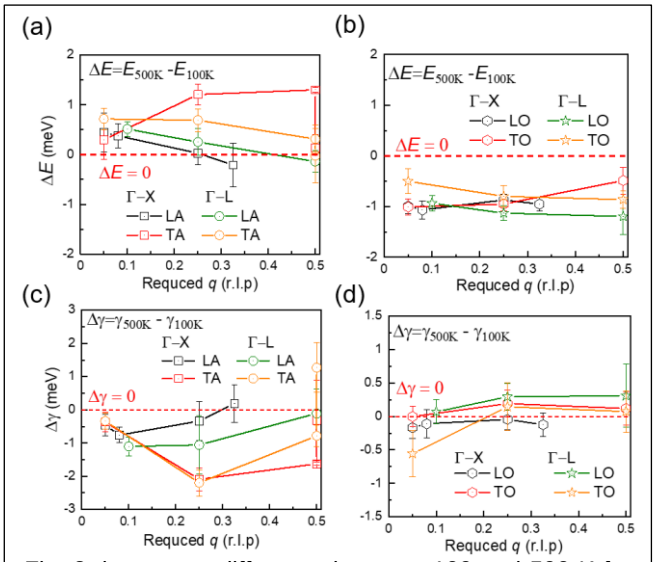

Fig. 2 the energy difference between 100 and 500 K for (a) acoustic and (b) optical phonon modes. (c) the phonon linewidth difference between 100 and 500 K for (c) acoustic and (d) optical phonon modes an orange line. Γ-X ([001]) and Γ-L ([110]).

Reference

[1] Li et al., *Science* 361, 579-581 (2018);
[2] Tian et al., *Science* 361, 582 (2018);
[3] Kang et al., *Science* 361, 575 (2018);
[4] Yang et al., *PRB* 100, 245203 (2019).