Phonon dynamics on thermoelectric material of Mg3Sb2

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Thermoelectric power generation is expected to contribute to energy conservation through the generation of electricity from waste heat. To realize this technique, intense efforts have been devoted to improving the performance. The difficulty in improving material performance comes from the conflicting requirements to exhibit high electrical conductivity while keeping the thermal conductivity low.

Mg3Sb2 is one of promising candidate exhibiting high thermoelectric performance. It shows high value of the dimensionless figure of merit ZT ~ 1.65 with quite low lattice thermal conductivity of kL ~ 0.7 W/mK at T = 725 K [1,2]. Great attention has been attracted in this compound for the origin of its low kL. One assumption is based on the existence of lone pairs around Sb atoms. It has been argued that interatomic potential could be anharmonic due to lone pairs, thus, heat carrying phonons can be heavily scattered. But there is no clear evidence about the claim. Therefore, phonon must be studied using inelastic neutron scattering.

In this experiment, we aim to clarify the origin of low kL of Mg3Sb2 by examining phonons at FRM2 using PUMA. The weight of Mg3Sb2 single crystal was 1.21 g. Measurements were done on scattering planes of (hk0) and (hhl) to cover wide range of Brillouin zone. Phonon calculations were performed using the ABINIT program.

Mg3Sb2 has CaAl2Si2-type crystal structure with the P-3m1 space group. Mg atoms occupy both Ca and Al sites. Thus, it is basically a 122 Zintl phase compound. Charge density calculations on SrZn2Sb2 that has the same crystal structure as Mg3Sb2 reveal the existence of lone pairs around Sb atoms [3].

Figure 1 shows phonon dispersion at T = 298 K. Solid lines are results of calculations. Overall, observed peaks agree well with the calculations, ensuring the correctness of the mode assignment. Observed modes are demonstrating by colored lines. Gray lines are remaining modes that we must measure next time. To find anharmonic phonons that can be the origin of low lattice thermal conductivity in Mg3Sb2, we will measure temperature dependences in the near future. Further experiments are required to obtain overall picture of phonon dynamics and to clarify the role of lone pairs that are believed to be responsible for the anharmonicity.

[1] H. Tamaki et al., Adv. Mater. 28, 10182 (2016). [2] J. Zhang et al., Nature Commun. 8, 13901 (2017). [3] E. S. Toberer et al., Dalton Trans. 39, 1046 (2010)

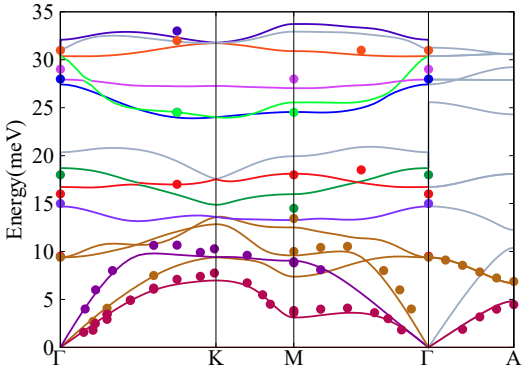

Fig. 1. Phonon dispersions of Mg3Sb2 at T = 298K. Solid lines are results of calculations. Color and gray lines depict observed and unobserved modes, respectively.