Anomalous Change of Heat Transfer in Sr(Ti,Nb)O_3 solid solution

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Strontium titanate ($SrTiO_3$) is one of the most promising oxide based thermoelectric (TE) materials for replacing heavy metal based TE materials. It shows a very high power factor (PF) of ~2.5 mW m^{-1} K^{-2} at room temperature, which is comparable to that of the commercial Bi_2Te_3.

Recently, great efforts have been poured into further improving PF of $SrTiO_3$ based TE materials. In 2017, we have clarified the TE phase diagram of $SrTi_{1-x}Nb_xO_3$ solid-solution (STNOss, $0.05 \leq x \leq 1$) films with an emphasis on PF, and a conduction band transition from Ti 3d to Nb 4d was confirmed at x~0.3. Furthermore, their electronic transport properties showed a phase boundary at x~0.5[1]. In addition, using the longer de Broglie wavelength and high electron transport in heavily Nb-substituted $SrTiO_3$ superlattices, we further increased the effective PF to 5 mW m^{-1} K^{-2}, which is double the optimal bulk value[2].

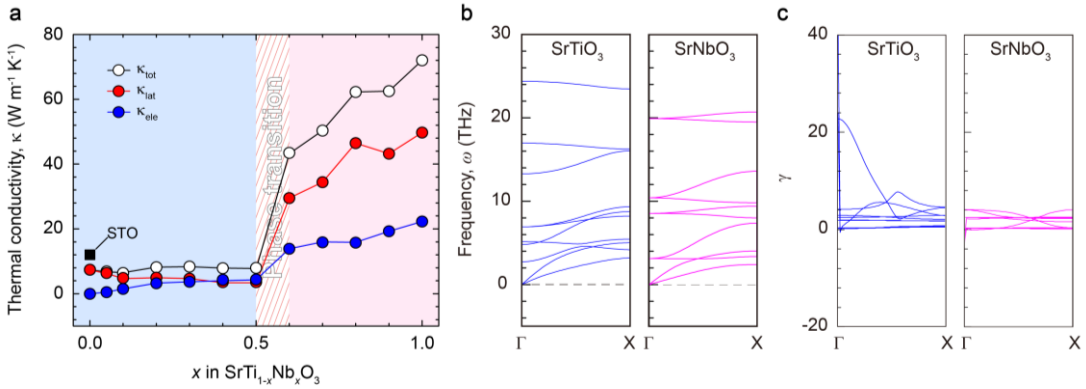

Fig.| (a) x-dependent thermal conductivities of $SrTi_{1-x}Nb_xO_3$ at room temperature; (b) Phonon dispersion and (c) Grüneisen parameter of $SrTiO_3$ and $SrNbO_3$ along [001] direction.

In order to fully understand the overall TE performance of $SrTiO_3$ based materials, systematic investigations should also be carried out on their thermal transports. Here, using STNOss epitaxial films, we investigated heat transfer properties and observed anomalous behavior in the thermal conductivity. Figure (a) shows the x-dependent thermal conductivities of STNOss. The lattice thermal conductivity (κ_{lat}) was extracted by $\kappa_{lat} = \kappa_{tot} - \kappa_{ele}$, where κ_{tot} is the total thermal conductivity measured by TDTR, and κ_{ele} is electron thermal conductivity calculated with the Wiedmann-Franz law. Steep increases in both κ_{tot} and κ_{lat} were observed around x~0.5, suggesting a phase transition from $SrTiO_3$-dominated phase to $SrNbO_3$-dominated phase. This phase transition is also supported by polaron effect, which only exits in $SrTiO_3$-dominated phase. Unlike the traditional alloy systems, $SrNbO_3$, which has larger average atomic mass, shows much higher κ_{lat} than $SrTiO_3$. According to the phonon dispersion calculations, $SrTiO_3$ shows much more intense scattering between optical and acoustic branches than $SrNbO_3$, which suppresses phonon mean free path for $SrTiO_3$ [Fig. (b)]. The Grüneisen parameters of $SrTiO_3$ were much higher than those of $SrNbO_3$, which will reduce the phonon transport properties [Fig. (c)].

Our findings of heat transports in STNOss will be a fundamental contribution to the current knowledge in heat transportation and may be utilized to develop new TE materials.

Reference

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[2] Y. Zhang, H. Ohta *et al.* ***Nature Commun.*** 9, 2224 (2018).