Sound Speed Differentiates Thermal Transport in Lead Halide Perovskites

G. Elbaz[1], W.L. Ong[1,2], E. A. Doud[1], P. Kim[3], D. W. Paley[1], X. Roy[1], J. A. Malen[2]

[1] Department of Chemistry, Columbia University, New York, NY, 10027
[2] Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, PA 15217
[3] Department of Physics, Harvard University, Cambridge, MA 02138

Lead halide perovskites ($APbX_3$) have received widespread attention in the last few years due to their remarkable properties as active materials for solar cells. A lot of effort has been put into understanding their carrier dynamics and optoelectronic properties. By comparison, little attention has been given to understanding thermal transport in lead halide perovskites, despite its importance in solar applications and other emerging functions such as optoelectronics and thermoelectrics. As with carrier dynamics, current theories implicate the A-cation motions as the cause of the ultra-low thermal conductivity.

Here, we provide direct experimental evidence that challenges this theory by performing a comprehensive study on phonon thermal transport in a series of single crystal lead halide perovskites. Using frequency domain thermal reflectance, differential scanning calorimetry and nanoindentation, we measure the thermal conductivity, heat capacity and sound speed of a series of five lead halide perovskites: $CH_3NH_3PbI_3$, $CH_3NH_3PbBr_3$, $CH_3NH_3PbCl_3$, $CH(NH_2)_2PbBr_3$ and $CsPbBr_3$. This work represents the first report of thermal conductivity for all but $CH_3NH_3PbI_3$. We find that, like previous work on $CH_3NH_3PbI_3$, the thermal conductivity of all these perovskites is low. However, we show that the differences in thermal conductivity are primarily due to variations in the sound speed of the materials, and not due to differences in the phonon mean free paths. Moreover, with improved modeling, we show that the A-cation has little effect on the phonon scattering process contrary to previous assumptions. To this end, the thermal conductivity accumulation functions for lead halide perovskites show that at least 75% of thermal conductivity results from phonons having a mean free path shorter than 100 nm, regardless of whether resonant scattering is invoked. From a technological perspective, these results indicate that to improve the thermoelectric figure of merit of the perovskites, any reduction in the thermal conductivity would require creating sub-100 nm nanostructures.