MAR12-2011-003147

Abstract for an Invited Paper
for the MAR12 Meeting of
the American Physical Society

Computational approaches to finding earth-abundant thermoelectric materials[1]

VIDVUDS OZOLINS[2], Dept of Materials Science and Engineering, University of California, Los Angeles

Good thermoelectric materials should possess a combination of seemingly incompatible properties, such as high electronic mobility and low lattice thermal conductivity. Therefore, search for crystalline materials with glass-like thermal conductivity has been an active field of research. Several cubic I-V-VI_2 semiconductors, the paradigm for which is $AgSbTe_2$, have been shown to exhibit minimal values of lattice thermal conductivity at ambient temperatures when the phonon mean free path equals the interatomic distance. These modes are due to the existence of highly polarizable lone s^2 electron pairs on the group V cations. Electrostatic repulsion between the lone s^2 pairs and the valence charge on group VI anions tends to favor locally distorted bonding configurations and may lead to unstable phonons. We present the results of first-principles density functional theory (DFT) calculations of phonon dispersion and electron-phonon interactions in cubic I-V-VI_2 semiconductors, where the group I elements are Cu, Ag, Au or alkali metals, the group V elements are P, As, or Bi, and the group VI elements are S, Se, or Te. Compounds that have only marginally stable phonons have extremely large Grüneisen parameters that result in a thermal conductivity limited by Umklapp processes to values at the amorphous limit above 200 K. Following the *ab initio* calculation, we synthesized $AgSbTe_2$, $AgSbSe_2$, $AgBiTe_2$, $NaSbTe_2$, $NaSbSe_2$, and $NaBiTe_2$ and report their thermal conductivity and specific heat: in all cases, the experiments confirm the theory.

[1]This work was supported as part of the "Revolutionary Materials for Solid State Energy Conversion," an Energy Frontier Research Center funded by the U.S. Dept of Energy, Office of Science, Office of Basic Energy Sciences under Award Number DE-SC0001054.

[2]In collaboration with Michele Nielsen and Joseph Heremans, Department of Physics, Ohio State University