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Configuring pnicogen rings in skutterudites for low phonon conductivity[1]

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During the past dozen or so years, skutterudites have attracted much interest as prospective thermoelectric materials for power-generation applications in the temperature range 500K - 850K. Primary interest was focused on filled forms of skutterudites where loosely-bonded filler species resonantly scatter normal phonon modes of the structure thus reducing the lattice thermal conductivity. Using this approach with multiple fillers and incorporating various forms of nanoinclusions, impressive figures of merit ZT = 1.5-1.7 have been reported with n-type filled skutterudites. Since the dominant heat-carrying modes in skutterudites are associated with vibrations of the pnicogen rings, disruptions of the ring structure by substitutional alloying should be a similarly effective approach of lowering the lattice thermal conductivity. In this talk I discuss our recent work exploring alloying configurations of pnicogen rings that yield particularly low values of the thermal conductivity. We found that compensated double-substitution (replacing two Sb atoms with one atom each from the column IV and column VI elements) is a very effective approach. Our ab initio calculations, in combination with a cluster expansion, have allowed us to identify stable alloy configurations on the Sb rings. Subsequent molecular and lattice dynamics simulations on low energy configurations established the range of atomic displacement parameters and values of the thermal conductivity. Theoretical results turned out to be in good agreement with our experimental thermal conductivity values. Combining both approaches of compensated double-substitution and filling of structural cages should be an effective way of further improving the thermoelectric figure of merit of skutterudites.

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