Progress in the widespread adoption of all solid heat-to-electricity technologies has largely been hindered by the absence of suitable thermoelectric materials. In pursuit for new thermoelectrics recent advances in large-scale deployment of first-principles calculations could be useful in identifying new promising material systems. However, the need to predict electron and phonon transport properties with sufficient accuracy renders direct assessment of the thermoelectric figure of merit (zT) for large numbers of systems unfeasible. This is true even in the case of relatively simple material systems, which could be described by the computationally inexpensive single particle theories such as DFT. While the state-of-the-art DFT based approaches to charge carrier and heat transport of semiconductors can deliver desired accuracy, they are still limited to relatively simple chemistries and/or case-by-case studies. In this talk I will discuss integrated theory-computation-experiment efforts in developing a robust set of material descriptors that: (1) are rooted in the Boltzmann transport theory, but do not rely on classic and largely inapplicable constant relaxation time or constant mean free path approximations, (2) are computationally tractable allowing material searches across large chemical spaces, and (3) are sufficiently accurate to provide reliable predictions. Our approach is demonstrated to correctly identify known thermoelectric materials\textsuperscript{1} and reliably suggest new and promising candidate semiconductors.\textsuperscript{2} At the end, I will review successes and failures in our quest for new thermoelectrics, and discuss dopability of semiconductors as the critical outstanding challenge in achieving high zT materials.
