

MRS spring meeting – Phoenix, Arizona, 22nd – 26th April 2019

Impact of scattering mechanism details on the thermoelectric power factor of complex materials

Many new thermoelectric materials including half-Heusler alloys, PbTe and SnSe based compounds, skutterudites, clathrates, to name a few, are known to possess rich and complex bandstructures, with highly anisotropic bands belonging to multiple valleys having manifold degeneracies in both conduction and valence bands. These materials often exhibit very low thermal conductivities, which can be further reduced using nanostructuring techniques to reach down to, or even below the amorphous limit. More recently, however, significant efforts are also being undertaken to take advantage of the rich bandstructure features to achieve power factor improvements as well. These include identifying the best alloying strategies, introducing resonant states in the bands, aligning of the multiple bands at the same energy, etc. Large efforts are also placed by the computational materials science community into bandstructure calculations through involved DFT codes and high-throughput material screening using data from a variety of databases.

Less progress, however, is achieved in combining such involved calculations with accurate transport models, which take into account different scattering mechanisms including their energy dependences, the details of intra/inter valley scattering, details of anisotropy in the bandstructure, electronic relaxation details, etc. Lack of these considerations leads to wrong performance estimations, inaccurate comparisons, and sub-optimal design directions. At the moment, most calculations, despite the use of accurate bandstructures, are performed using constant relaxation time approximation due to the very challenging computational complexities of accurate scattering treatment.

Here, we describe the development of an advanced simulator that uses an arbitrary material bandstructure and computes the thermoelectric coefficients beyond the typically assumed constant relaxation time approximation. Our code includes all major scattering mechanisms (e.g. phonons, impurities, alloying) encountered in the materials of interest. The code uses the Fermi's Golden Rule in the energy dependent relaxation time approximation, computed by numerical considerations of all bandstructure states.

Using this, we then report on an investigation of a few typical thermoelectric materials and explore the differences in the performance predictions in cases where the various details of the scattering mechanisms as described above are considered in the calculations, versus if they are omitted using simplified methods. We indeed show that depending on the scattering physics considered, the power factor can vary substantially and materials performance rankings can be altered. More specifically, we show that in the efforts towards improving the power factor of materials through band alignment, different optimized bandstructure conclusions are reached if one uses the constant relaxation time approximation, versus scattering rates that depend on the density of intra-valley states, versus scattering rates that depend on the total density of states. In the case of half-Heusler compounds, for example, we show that the differences in the performance estimation of band aligned structures can

vary significantly depending on the scattering mechanism. We show that this variation depends on the effective masses of the aligned bands of the material as well.

Finally, we discuss the numerical issues of these simulations and approximations that could speed up the computation significantly without compromising the accuracy.