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

Patrizio Graziosi[1,], Chathurangi Kumarasinghe[1], Neophytos Neophytou[1]*

[1] School of Engineering, University of Warwick, Coventry
CV4 7AL, UK
**Patrizio.Graziosi@warwick.ac.uk*

Much effort is undertaken by the computational material science community for identifying suitable thermoelectric materials through high-throughput screening and calculation of their accurate bandstructures. However, less progress is seen in combining such calculations with accurate transport models that 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 (Fig. 1). 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.

We then investigate Co-based half-Heusler alloys that are known for their good thermoelectric performance. We 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 leading to different optimized band structures.

Half-Heuslers possess highly anisotropic bands belonging to multiple valleys having manifold degeneracies in both conduction and valence bands. Such rich bandstructure features give opportunity to achieve power factor improvements through aligning multiple bands at the same energy near the valence or conduction band edges. Next, we show that band aligning in Co-based half-Heuslers can provide substantial power factor improvements. However, the proper scattering physics needs to be taken into consideration.

(a) (b)

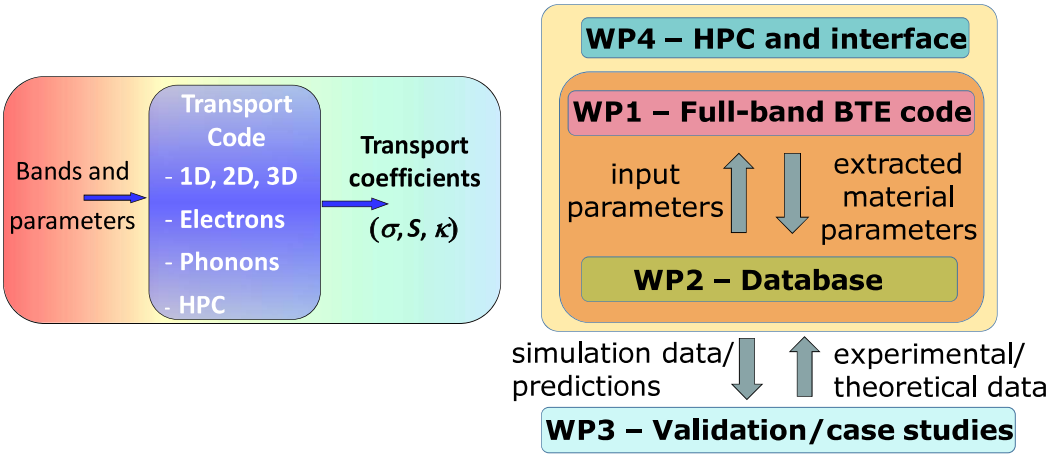

Figure 1: (a) conceptual scheme and (b) structure of the simulation code that is under development in the Marie Curie Individual Fellowship "Generic semiclassical transport simulator for new generation thermoelectric materials", project ID 788465. (a) By means a thorough use of the scattering physics the code will deliver accurate transport coefficients from initial band structures and other parameters, for different geometries and with High Performance Computing capability. (b) A semiclassical formalism based on the Boltzmann Transport Equation in mutual feedback with a complete database are the core of the code, that will be equipped with a user friendly interface and is validated in respect of experimentally well-known materials and applied to case studies.