

Bandstructure engineering in Co-based Half-Heuslers for high thermoelectric powerfactors.

Chathurangi Kumarasinghe*, and Neophytos Neophytou

School of Engineering, University of Warwick, Coventry, CV4 7AL, UK

* Chathu.Kumarasinghe@warwick.ac.uk

Half-Heuslers (HHs) have impressive thermoelectric power factors as a result band and orbital degeneracy combined with weak electron-phonon scattering. Investigations into their band structures, reveal that they have multiple bands that can be aligned through different band engineering approaches, giving opportunity to further improve their power factor. In this work we explore the power factor optimization of Co-based p-type HHs TiCoSb, NbCoSn, ZrCoSb and ZrCoBi using ab-initio Density Functional Theory (DFT) calculations and semi-classical Boltzmann transport. For this, we first extract the ab-initio power factor of these materials (using BoltzTrap). We then develop simplified bandstructure models based on the non-parabolic effective mass approximation, considering all relevant local and global band maxima found in Γ , L and W points in the Brillouin zone, that match the ab-initio power factors for Fermi levels up to 0.25eV into the valence band. This allows the exploration of the influence of band alignment in a trivial manner. We then calibrate our simple models to experimental data and more involved electron-phonon coupling calculations using Electron Phonon Wannier (EPW) to identify the correct form of the scattering rates, and perform a comprehensive study on the optimal band alignment for maximizing their thermoelectric power factor. Finally, using DFT again we explore alloying and second phasing possibilities (with full-heuslers), that would achieve the identified optimal alignment in the bands of the HHs under consideration.