Thermoelectric properties of complex band and nanostructured materials

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Thermoelectric (TE) materials convert heat from temperature gradients into useful electrical power. Over the last two decades, an enormous progress has been achieved, owning to the synthesis of complex bandstructure materials and their alloys (Fig. 1), and nanostructured materials (Fig. 2) [1]. The figure of merit, $ZT=\sigma S^2T/\kappa$ where σS^2 is the power factor (PF) and κ the thermal conductivity, quantifies the efficiency. It has increased by 2-fold, to values above $ZT > 2$ across materials and operating conditions, with a maximum of $ZT=2.6$.

In this work, we describe a computational framework to compute the electronic and thermoelectric transport in materials with multi-band electronic structures of an arbitrary shape (Fig. 3) by coupling density function theory (DFT) bandstructures to the Boltzmann Transport Equation (BTE). We explain the computational complexities and the approximations made. We then show how 'real-space' methods, such as the non-equilibrium Green's function (Fig. 4) and the Monte Carlo methods (Fig. 5) can be used to design nanostructured materials with very high TE PFs. We elaborate on the computational framework needed to advance the field of TEs.

Here, we present four related studies. First: due to the complexity of the electronic structure, it is customary to use a constant value for the relaxation times within the BTE. We show that the full energy and momentum dependence of the relaxation times is essential in capturing the correct transport features [2]. Second: in order to identify high performance materials within the myriad of possibilities, we present the development of a set of descriptors that can be used in materials screening studies [3]. A combination of the number of valleys, dielectric constant, conductivity effective mass, deformation potential, and bandgap, forms a useful descriptor. Third: we present a study in which we show a possible route to achieve over an order of magnitude increase in the TE power factor, by utilizing narrow gap materials and bipolar transport, a regime which is usually avoided [4]. Four: We demonstrate a practical nanostructuring design which can also allow for 10-fold improvement in the PF [6, 7].

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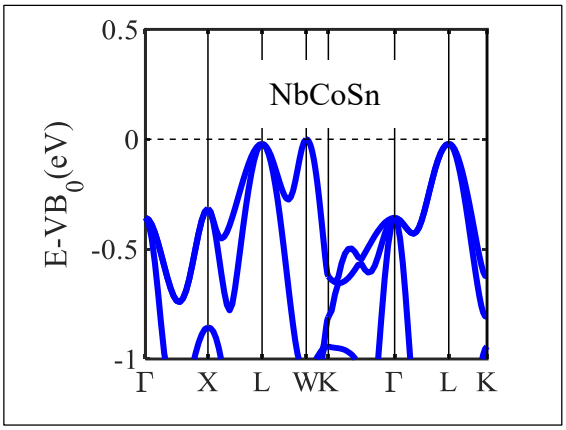

Fig.1: A typical example of a multi-band TE material, in this case the half-Heuselr NbCoSn. Multiple pockets are available for electronic transport.

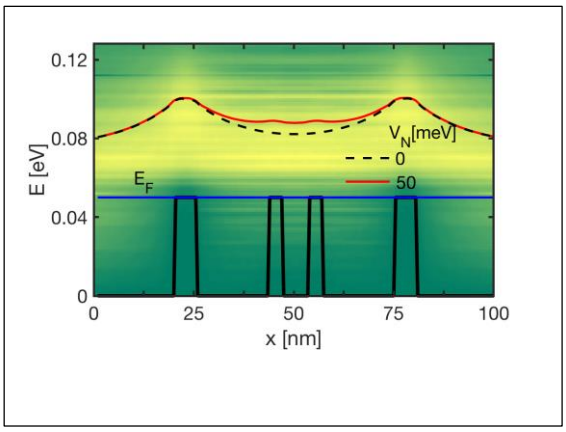

Fig.4: A typical NEGF simulation for the energy of the current flow in nanostructures, a quantity that determines the Seebeck coefficient. Phonon scattering (acoustic and optical) is included.

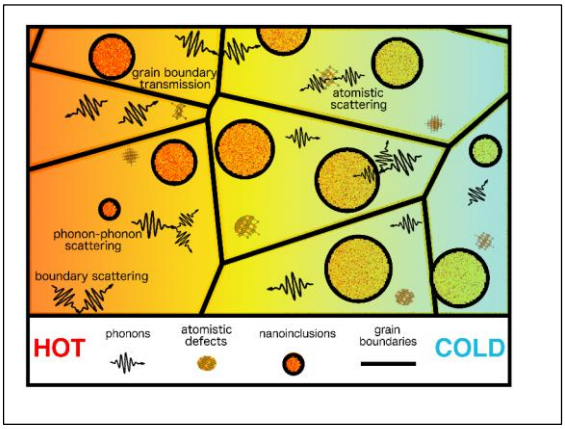

Fig.2: A typical nanostructured geometry, where grain boundaries appear at the macro-scale, nanoinclusions at the meso-scale, and atomic defects at the atomic scale. Phonons scatter effectively off these structures and the thermal conductivity is reduced. From [6].

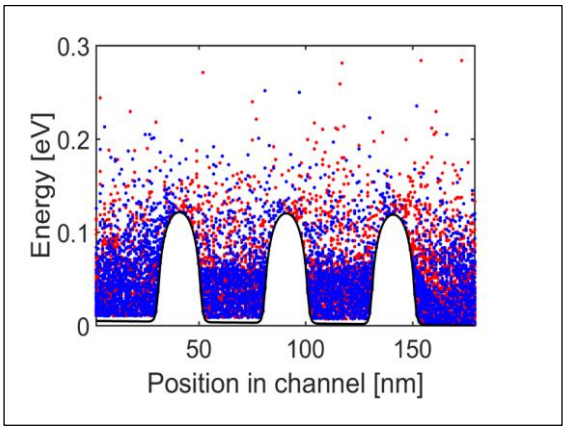

Fig.5: A snapshot from a Monte Carlo simulation in a material with alternative barriers and wells, an idealized schematic of a typical nanostructured TE material.

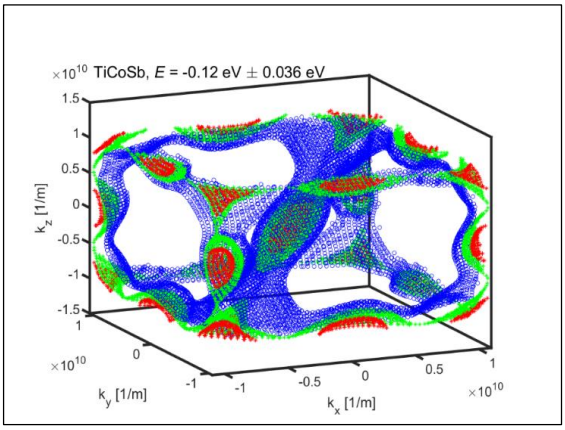

Fig.3: A typical energy surface in the 3D k-space of the valence band of TiCoSb. Points on the energy surface of different bands (blue, red, green) indicate the initial and final scattering states in the BTE.

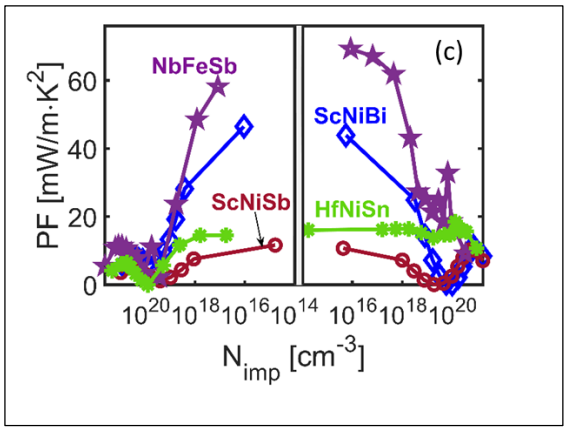

Fig.6: Low bandgap and highly anisotropic materials with regards to the properties of the conduction and valence bands can provide extremely high PRs. Adopted from [4].

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