Quantum transport simulations in hierarchical nanostructured materials for improved thermoelectric power factors

Neophytos Neophytou[*], Laura de Sousa Oliveira, Samuel Foster, Vassilios Vargiamidis

School of Engineering, University of Warwick, UK

* Corresponding Author: n.neophytou@warwick.ac.uk

Nanostructuring is a promising approach for the next generation thermoelectric materials yielding ultra-low thermal conductivities. More specifically, some of the lower thermal conductivities in materials have been achieved by the inclusion of hierarchically sized structures, at the atomic size, the nanoscale, and mesoscale, which can scatter phonons of various wavelengths and reduce phonon transport throughout the spectrum [1]. Less attention has been paid, however, to the power factor (PF), which in most cases is reduced.

In this work, we employ a fully quantum mechanical simulator based on the non-equilibrium Green's function (NEGF) method to compute the thermoelectric PF of hierarchically nanostructured materials that include nanocrystalline potential barrier

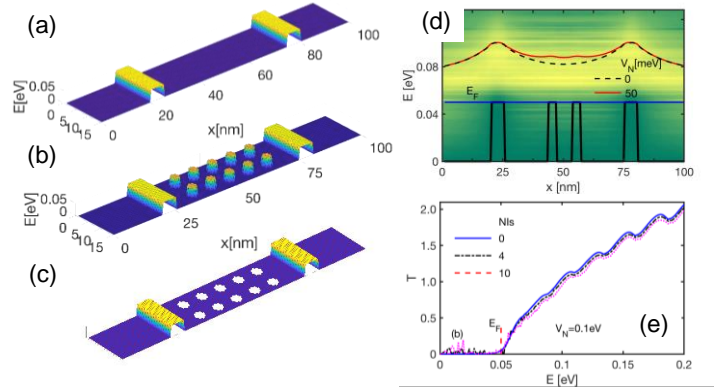
Fig. 1. (a-c) The nanostructures considered: a) nanocrystalline boundaries, b) boundaries and nanoinclusions, c) boundaries and voids. (d) The energy of the current flow, which determines the Seebeck coefficient. (d) Quantum mechanical transmission in structure (b) for different densities of nanoinclusions, indicating immunity to the nanoinclusion density.

boundaries (Fig. 1a), boundaries plus nanoinclusions (Fig. 1b), and boundaries plus voids (Fig. 1c). These are structures that have already experimentally shown promising PFs [2-4]. The simulator we present, includes all important nanoscale transport effects, and treats scattering from complex geometrical features naturally, without the need for any approximations. Importantly, we include the computationally expensive electron-phonon interaction treatment, which allows robust treatment of nanoscale transport physics from ballistic/coherent all the way to the diffusive/incoherent regime, both included within the same formalism. Our formalism can naturally also provide the specifics of the current flow resolved both in energy and space (Fig. 1d), from which the Seebeck coefficient can be resolved, as well as the effect of nanostructuring on the Seebeck and the conductivity. Carrier clearly absorb optical phonons to gain energy and overpass the barriers, then emit optical phonons and relax in the quantum wells, with the nanoinclusions (NIs) pushing their energy a bit higher, resulting in a slight increase in the Seebeck coefficient.

We discuss the conditions under which the PF is less affected by the presence of nanostructuring, and conditions under which the PF is even enhanced. More specifically, we illustrate a design methodology which dictates the position of the Fermi level, the height of the potential of the nanocrystalline barriers, the sizes of the grains and grain boundaries, which makes the design by large immune to the density of nanoinclusions and their barrier height, i.e. the quantum mechanical transmission becomes insensitive to disorder (Fig. 1e) [5-6]. Disorder, on the other hand, has a drastic effect on the thermal conductivity. We couple our results with Molecular Dynamics simulations to compute the reduction in thermal conductivity in our hierarchically disordered structures. As expected, the large reductions in the thermal conductivity, and the immunity of the power factor to disorder in our designs, allows for large increases in the ZT. Our findings can complement the large volume of work on reducing thermal conductivity, and point to new design directions that further increase the ZT figure of merit, this time through allowing power factor improvements for materials that are already known to have ultra-low thermal conductivities.

References
1. K. Biswas, J. He, I. D. Blum, C.-I. Wu, T. P. Hogan, D. N. Seidman, V. P. Dravid, and M. G. Kanatzidis, Nature 489, 414 (2012).
2. N. Neophytou, X. Zianni, H. Kosina, S. Frabboni, B. Lorenzi, D. Narducci, Nanotechnology 24, 205402 (2013).
3. B. Lorenzi , D. Narducci, R. Tonini, S. Frabboni, G.C. Gazzadi, G. Ottaviani, N. Neophytou, X. Zianni, Journal of Electronic Materials, 43, 10, 3812-3816 (2014).
4. L.-D. Zhao, V. P. Dravid, and M. G. Kanatzidis, Energy Environ. Sci. 7, 251 (2014).
5. S. Foster, M. Thesberg, and N. Neophytou, Phys. Rev. B, 96, 195425, (2017).
6. V. Vargiamidis and N. Neophytou, Phys. Rev. B, 99, 045405, (2019).