Phonon transport simulations in hierarchically disordered silicon based nanostructures

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I. Introduction

Highly disordered nanostructures are one of the most promising ways to achieve very high thermoelectric (TE) efficiencies by increasing power factor and reducing thermal conductivity (κ) leading to a high thermoelectric figure of merit (ZT). Thus, engineering such materials has recently attracted significant attention. Strong disorder, and more specifically disorder on hierarchical length scales, can scatter phonons of different wavelengths throughout the spectrum and drastically reduce κ (thus increase ZT). Using hierarchical inclusions at different scales, Biswas *et* al. reported a κ of 0.9 Wm^{-1} K^{-1} at 915 K and a ZT of 2.2. [1] More recently, using this method for the p-type $Pb_{0.98}Na_{0.02}Te$-SrTe system, Tan *et* al. reported an even lower κ of 0.5 W $K^{-1}m^{-1}$ and a higher ZT of 2.5 at 923K.[2] Drastic reductions in thermal conductivity were also reported in a recent work on SiGe nanomeshes, with ultra-low κ of 0.55 ± 0.10 Wm^{-1} K^{-1} for SiGe nanocrystalline nanoporous structures, a value well below the amorphous limit.[3] In order to develop improved materials with enhanced ZT and other desirable characteristics a strong understanding of phonon transport in such materials is needed - which can be provided through advanced theory and simulations. In this work, we solve the Boltzmann transport equation for phonons in disordered silicon nanostructures using the Monte Carlo (MC) method.

II. Method

Monte Carlo, which can capture the details of geometry in relevant accuracy, is widely employed to understand phonon transport in various nanostructures such as nanowires, thin films, nanoporous materials, polycrystalline materials, nanocomposites, corrugated structures**,** Silicon-on-Insulator devices, etc. [4,5]. The Monte Carlo (MC) approach has been adopted for a semi-classical particle based description of phonon transport. We use the 'single-phonon MC' approach which differs from the multi-phonon MC approach described in various works in the literature in terms of phonon attributes book-keeping. This has been extensively described in our published work.[4,5,6] For computational efficiency we consider a 2D simulation domain of length L_x = 1000 nm and width L_y = 500 nm. The domain is populated with nanostructured features as shown in Fig. 1.

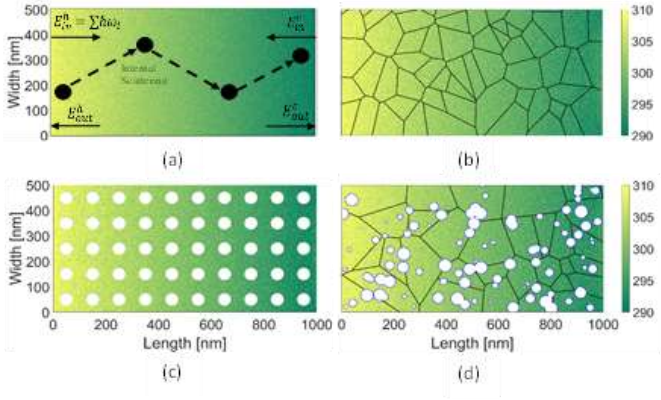

Fig. 1: Nanostructured geometries considered: (a) Pristine silicon channel. (b) Nanocrystalline (NC) channel. (c) Ordered nanopores (NP) within the channel of ~20% porosity in a rectangular arrangement. (d) Combined NC and disordered NP material. The coloring indicates the established thermal gradients when the left and right contacts are set to T_H = 310 K (yellow) and T_C = 290 K (green).

III. Results

A. Influence of grain size and boundary roughness

We begin our investigation with the effects of the grain size <d> and boundary roughness (Δ_{rms}) on κ. In the case of nanocrystalline geometries as shown in Fig. 1b, the average grain size in the simulation domain is defined as $< d >= L_x / < N_G >$, where L_x is the length of the domain in the transport direction and $< N_G >$ is the average number of grains encountered in that length. The results are shown in Fig. 2 where κ is plotted as a function of average grain size <d>. We consider average grain size from <d> = 1000 nm down to <d> = 50 nm as indicated by the geometry sub-figures above the graph in Fig. 2 (from sub-figure 1 where <d> = 50 nm to sub-figure 6 where <d> = 225 nm). Three different roughness values of Δ_{rms} = 0.25 nm, 1 nm, and 2 nm were simulated, shown in Fig. 2 by the red, blue, and black lines, respectively. Each point shown in Fig. 2 is an average of 50 simulations. Decreasing grain size causes a reduction in κ, from 97.8 $Wm^{-1}K^{-1}$ to 19.9 $Wm^{-1}K^{-1}$. This is consistent with other available theoretical and experimental results. [5,6] An important observation is that a rapid drop in κ is observed for structures in which the average grain size is below the average phonon mean-free-path (λ_{pp} = 135 nm). For these structures grain boundary scattering has a more dominant role than intrinsic

* Research supported by the European Research Council (ERC) under the European Union's Horizon 2020 Research and Innovation Programme (Grant Agreement No. 678763).

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three-phonon scattering. This observation is consistent for the different values of grain boundary roughness (Δ_{rms}).

On the other hand, changes in the values of Δ_{rms} seem to play a comparatively smaller role in decreasing κ (comparing the red, blue, and black lines respectively, in Fig. 2). Phonon paths are already randomized by the numerous grains and intrinsic scattering, and thus the additional randomness from boundary roughness plays a minimal role.

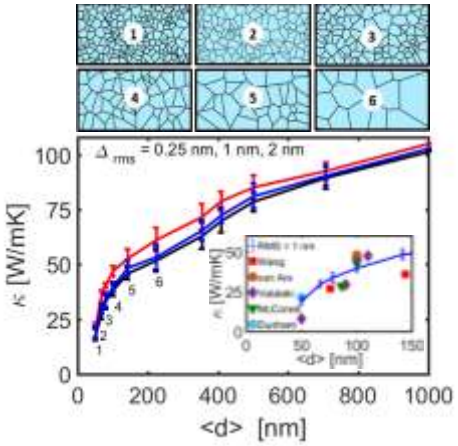

Fig. 2: The effects of grain size and grain boundary roughness (Δ_{rms}) on the κ of the silicon channel.

B. *Hierarchical disordered nanostructures*

We next combine the effects of nanocrystallinity and porosity, as in realistic nanocomposite materials. Ordered and randomized pores are considered as shown in Fig. 3a and Fig. 3b, respectively. Fig. 3c plots κ versus $\langle d \rangle$ for structures with different porosity values (φ). Roughness on the transmittable grain boundaries is fixed to Δ_{rms} = 1 nm, corresponding to rough, almost fully diffusive cases. Again, each point shown in Fig. 3c is an average of 50 simulations.

The top blue line depicts the zero porosity case, the same as the initial results for Δ_{rms} = 1 nm shown in Fig. 2. Adding pores in an ordered fashion further reduces κ. This can be seen for 5% (magenta line), 10% (light blue) and 15% (red) ordered porosity. The κ decreases as either porosity increases, or $\langle d \rangle$ decreases, with large porosity dominating at large grain sizes, whereas boundary scattering dominates at small grain sizes. The red-dashed line in Fig. 3c shows the κ versus average grain size in the case of a φ = 15% randomized porous structure. The pores are randomized in terms of diameter and position as indicated in Fig. 3b. The pore sizes are varied from D = 10 nm to 50 nm in a random fashion using a uniform distribution. In this case, at $\langle d \rangle$ = 1000 nm at the right side of Fig. 3c, there is an initial 50% drop in κ in the randomized case compared to the ordered (red-solid line), followed by a slow rate of decrease in κ as the grain size decreases. This suggests that a high degree of randomization and small average pore size, makes phonon scattering on pores much more dominant than the intrinsic three phonon scattering and grain boundary scattering. When the grain size becomes small (below λ_{pp}), then it starts to play an important role again.

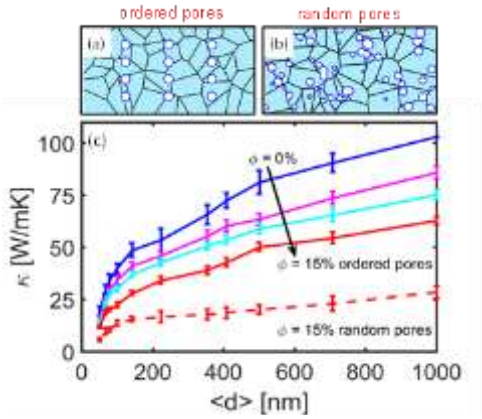

Fig. 3: Monte Carlo simulations showing the combined effects of grain size and porosity (φ) in both the ordered pores case (solid lines) and random pores case (dashed line) versus grain size <d>. The κ for porosities φ = 0%, 5%, 10% and 15% are shown by the blue, magenta, light blue, and red lines respectively.

IV. Conclusions

We investigated the presence of nanocrystalline and nanocomposite geometries, in ordered and disordered realizations. In NC the effect of grain size on κ is more pronounced at $\langle d \rangle$ smaller than the λ_{pp}. In that case, boundary scattering dominates over internal three-phonon scattering. We show that randomization in the pore features, which is often overlooked, can further reduce thermal conduction by even up to 60% compared to the ordered pore geometry. We show that the new compact models we introduce can be used within Matthiessen's rule to combine scattering from different geometrical features within ~10% accuracy. We believe our results will provide guidance in developing better understanding of thermal transport in nanostructured materials and aid the design of better thermoelectric and heat management materials.

V. Future Work

Further work on nanoinclusions, incorporation of wave effects and novel rectifier device applications are envisaged. Current efforts are focused on thermal rectification and comparing MC results to wave based methods (MD, NEGF).

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