Molecular dynamics simulations in disordered nanoporous materials

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Abstract

The introduction of nanopores and nanovoids in Si-based materials has been amply investigated in recent years as a means of achieving drastic thermal conductivity reductions for thermoelectric applications [1, 2, 3, 4]. Researchers have shown that introducing nanoscale pores and voids in materials, such as silicon, decreases thermal conductivity (κ) to near and even below the materials' amorphous limit [2,3]. Efficient thermoelectrics also require a high electrical conductivity (σ) and optimal nanostructuring should, therefore, be able to reduce κ while ideally maintaining, or even increasing, σ. A thorough understanding of how geometry affects phonon transport is needed to design low thermal conductivity materials in such a way that the degradation of electronic conduction due to nanostructuring is minimized.

Despite several recent studies of the effects of porosity at the nanoscale, a clear understanding of the microscopic details of phonon scattering on pores and voids of various sizes and configurations, both ordered or disordered, is far from complete. In this work, we employ molecular dynamics simulations using the Green–Kubo formalism to investigate a series of geometrical factors that contribute to thermal resistance in materials with nanopores and nanovoids. An illustration of the geometries considered is depicted in Fig. 1 (a–f), where a distinction between pores and voids (a) is illustrated, and examples of geometries are shown with varying number or pores/voids (b), varying void/pore diameters (c), staggering (d), clustering (e), and randomization/disorder (f). In summary, we show that multiple mechanisms contribute to the significant decrease in thermal conductivity that disorder affords, and quantify their effect at different porosities.

We further elaborate on how a wave-packet approach can be used to perform a mode-dependent study of the influence atomistic defects have on phonon transport, to help elucidate the physics of phonon transport in geometrically complex geometries. This approach can both visually show how phonon modes interact with voids/pores as they pass through them and be used to garner quantitative insight. An example of a wave-packet propagating through silicon with and without pores is provided in Fig. 1 (d) to illustrate the method. Finally, we discuss how the knowledge acquired through a wave-packet approach can be used in tandem with large-scale Monte Carlo simulations to help bridge the gap between nano- and mesoscale transport for better predictions of thermal conductivity in highly disordered materials.

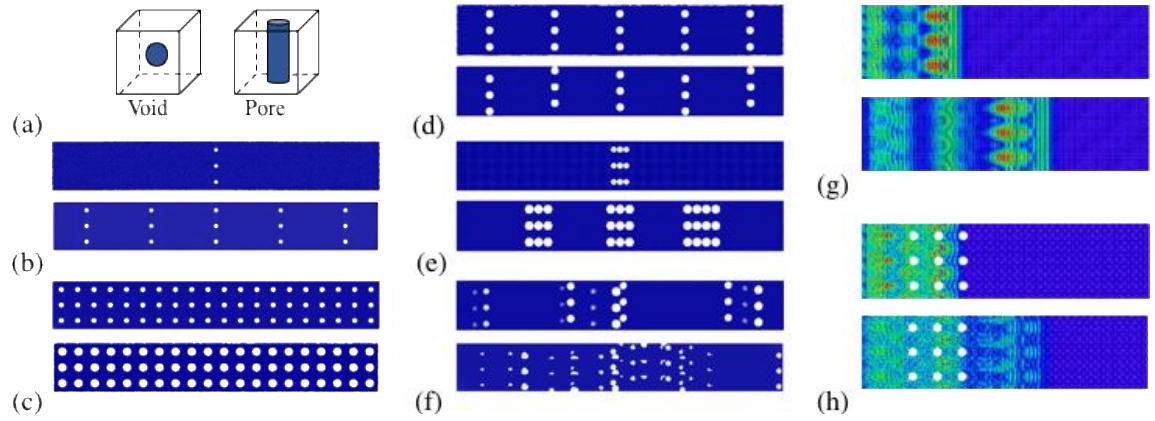

Figure 1: Illustration of geometrical variations considered: (a) voids (spherical) and pores (cylindrical), (b) varying numbers of pores/voids, (c) varying diameters of pores/voids, (d) staggering, (e) clustering, (f) randomization. Wave-packet based on the longitudinal acoustic mode propagating along the [1 0 0] direction, on silicon that is (g) pristine and (h) with uniformly distributed nanopores.

References

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