Monte Carlo Simulations of Thermal Conductivity in Nanoporous Si Membranes

Stefanie Wolf, Neophytos Neophytou, Zlatan Stanojevic, and Hans Kosina

Institute for Microelectronics, Technical University of Vienna, Austria
{wolf | neophytou | stanojevic | kosina}@iue.tuwien.ac.at

Abstract

We present a Monte Carlo (MC) study of heat transport in Si nanomeshes. Phonons are treated semiclassically as particles of specific energy and velocity that undergo Umklapp scattering and boundary scattering on the surfaces of the nanomesh pores. We investigate the influence of: i) geometric parameters such as the pore arrangement/randomness and porosity and, ii) the roughness strength of the pore surfaces on the thermal conductivity of the nanomeshes. We show that the nanomesh porosity has a strong detrimental influence on the thermal conductivity. Boundary roughness still degrades the thermal conductivity, but its influence is smaller.

Introduction

Nanoporous membranes made of single-crystalline Si (referred to as "holey" Si) are promising candidates for thermoelectric materials as they can provide extremely low thermal conductivity κ, relatively high thermoelectric power factors, and the structure stability that other low-dimensional systems are lacking. Since Si is an abundant, non-toxic material with well-established manufacturing processes, such structures, once optimized, could provide the feasibility of large-scale applications. Recently, room temperature ZT values up to 0.4 were demonstrated in nanomeshes, a large increase compared to bulk $ZT_{bulk} \sim 0.01$ [1]. To understand the parameters which determine the thermal conductivity in nanomeshes, involved simulation work is required. In this work we calculate the thermal conductivity of nanoporous single crystalline Si membranes by solving the Boltzmann transport equation (BTE) for phonons using the MC method [2, 3]. We describe the theoretical methodology, and examine the influence of geometry including pore arrangement (rectangular, hexagonal, random), and material porosity, as well as the influence of boundary scattering on the thermal conductivity. We show that the material porosity strongly affects the thermal conductivity. Boundary scattering affects the thermal conductivity as well, but its effect is weaker.

Theory

The geometry of the devices is defined and a tetrahedral simulation grid is created using the gtsFramework [4]. The devices consist of a simulation domain (channel) and two thermal contacts, i.e. a heat source and a heat sink which act as black bodies. The MC algorithm accounts for longitudinal and transversal acoustic polarizations and nonlinear dispersion relations. As shown in **Fig 1**, following the work of Ref. [5], we use $\omega(k) = v_s k + ck^2$ as a fit for the bulk dispersion relation under the isotropic Brillouin zone approximation, where k is the wave vector norm and v_s and c are fitting parameters taken from Ref. [5] to match the thermal conductivity of bulk Si in the [100] direction.

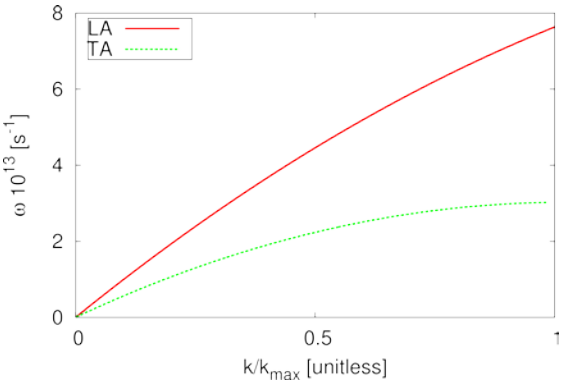

Fig. 1 Dispersion relations for longitudinal acoustic (LA) phonons in red, and transversal acoustic (TA) phonons in green.

The simulation steps are as follows: A phonon is initialized at a random position in the device. Initialization involves sampling of the phonon frequency, wave-vector, polarization and group velocity. The group velocity is defined as the slope of the dispersion relation and is co-directional with the wave-vector. The phonon alternates between free flight and scattering events. During the free flight phase, phonons move linearly in time. Boundary scattering does not change the phonon frequency, but solely its direction. In order to implement surface roughness we use a constant specularity parameter *p* to treat specular or diffusive boundary scattering. Three-phonon scattering is treated using phonon lifetimes as functions of frequency and temperature *T* for both polarization branches [6].

In case of boundary scattering the direction is reset. In case of three-phonon scattering the energy of the phonon is reset. This violates energy conservation, and to account for the energy difference the cell energy and temperature are modified accordingly. If the phonon reaches an outward boundary of the thermal contacts it is absorbed and the simulation continues with a newly created phonon. This sequence is repeated until the cell temperature difference between iterations is below the error tolerance. After reaching steady-state we calculate the heat flux by sequentially injecting a prescribed number of phonons (*N*) first from the source, into the device. We sum the phonons' incident energy to E_{in}^{H}. As the phonons travel through the device, they engage in three-phonon scattering which can alter their frequency and direction. The energy of all back-scattered phonons (leaving the device through the source contact) is summed up to E_{out}^{H}. Additionally, we calculate the average time that it takes for a phonon to travel the distance through the device (time-of-flight, TOF). Then we repeat the procedure by initializing phonons from the heat sink and calculate E_{in}^{C} and E_{out}^{C}. The phonon flux is then given by $F = N_{tot}\dfrac{\left(E_{in}^{H} - E_{out}^{H}\right) - \left(E_{in}^{C} - E_{out}^{C}\right)}{N < TOF >}$,

where N_{tot} is the total number of phonons inside the device.

The thermal conductivity is calculated using the heat flux through the medium for a given thermal gradient ∇T by applying Fourier's law as $\kappa = F / \nabla T$.

Results and Discussion

We investigate the thermal conductivity in nanomeshes of three different hole arrangements: rectangular, hexagonal, and random. **Fig. 2** shows examples of the simulated device geometries, as well as phonon trajectories in each structure.

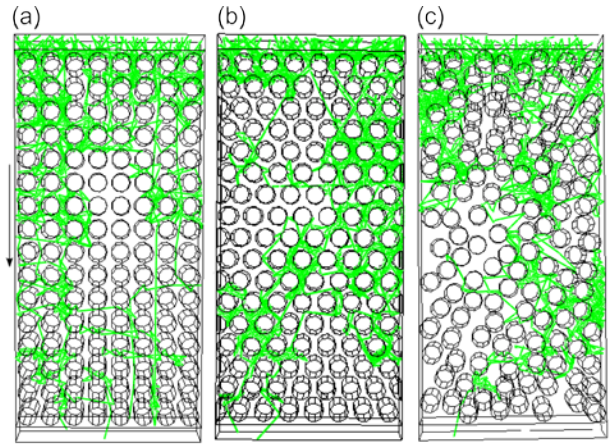

Fig. 2: Nanoporous devices with (a) rectangular, (b) hexagonal and (c) random arrangement of holes. The green lines indicate trajectories of phonons, which were initialized at the top contact (source, *H*). Phonons undergo three-phonon and boundary scattering events before that reach the bottom contact (heat sink, *C*).

The data in **Fig. 3** present simulation results for the thermal conductivity in the nanomeshes examined. The thermal conductivity is plotted as a function of porosity Φ and results for structures with different boundary specularity parameter are shown (p=1 denotes fully specular boundaries, whereas p=0.1 almost fully diffusive boundaries). In **Fig. 3a** the red lines represent the results for the nanomeshes with rectangular arrangement of the dots and the blue lines the results for the nanomeshes with hexagonal arrangement of the dots. **Fig. 3b** shows the results for nanomeshes in which the dots are arranged randomly. In the latter case, the standard deviation is denoted by the error bars. In all cases the diameter of the pores is fixed at 50 nm.

Two main conclusions can be deduced from this figure: i) The hexagonal and the randomized arrangements yield somewhat lower thermal conductivities compared to the rectangular ones. This is attributed to the fact that the rectangular arrangement provides straight paths for the phonons to travel, in contrast to the other two arrangements. ii) As expected, the thermal conductivity decreases with increasing porosity but, interestingly, most of the reduction is observed up to a porosity of 35%. Increasing the porosity from 0% to 35% results in a factor of ~4x reduction in the thermal conductivity. Further increase in the porosity results in smaller relative thermal conductivity reduction.

Specifically for the geometries consisting of randomized pores, after extracting the statistics from 100 different sample devices with random hole arrangements for each porosity value we find that: i) Under the same porosity conditions, increasing the roughness strength by an order of magnitude (from p=1 down to p=0.1), reduces the thermal conductivity by only ~40% (see the difference between the upper and lower line in **Fig. 3b**). ii) Under the same roughness conditions, however, it takes only 10-20% increase in the porosity to reduce the thermal conductivity by the same amount (~40%) (see **Fig. 3b**). In addition, we find that with increasing porosity the roughness strength becomes less effective in determining the thermal conductivity, indicating the relative

importance of the porosity over the roughness. For example, at 50% porosity, increasing the roughness of the pores from 50% diffusive (p=0.5) to 90% diffusive (p=0.1), changes the thermal conductivity only marginally.

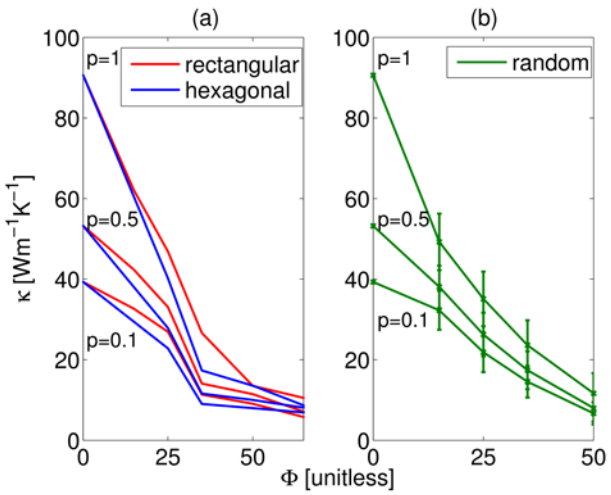

Fig. 3 The thermal conductivity as a function of porosity. Cases for pore surface roughness specularity parameter p=1 (specular), p=0.5 (50% specular), and p=0.1 (90% diffusive) are shown. In (a) blue lines denote hexagonal pore arrangements, whereas red lines denote rectangular pore arrangements. (b) Ramdomized pore arrangements. 100 samples are simulated for each data point.

Conclusions

In this work we present a Monte Carlo simulator for the calculation of heat transport in Si nanomeshes and a consequent study of the thermal conductivity in these structures under different geometrical constraints. We show that hexagonal arranged pores in nanomeshes could achieve lower thermal conductivities compared to rectangular arranged pores. We also show that the influence of porosity on the thermal conductivity is much stronger compared to the influence of the boundary roughness. Strong thermal conductivity reduction can be achieved in nanomeshes as the porosity increases to ~35%, whereas for larger porosities the reduction is slower.

Acknowledgements

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References

[1] J. Tang et al, Nano Lett., 10, 4279 (2010); [2] S. Mazumder, A. Majumdar, J. Heat Transfer 123, 749 (2001); [3] D. Lacroix et al Phys. Rev. B 72, 064305 (2005); [4] http://www.globaltcad.com; [5] M.G. Holland, Phys. Rev. 132, 2461 (1963); [6] E. Pop et al, J. Appl. Phys 96, 4998 (2004)