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# Thermal conductivity of deca-nanometric patterned Si membranes by multiscale simulations

Hayat Zaoui [a], Pier Luca Palla [a], Stefano Giordano [a], Fabrizio Cleri [a], Maxime Verdier [b], David Lacroix [b], Jean-François Robillard [a], Konstantinos Termentzidis [b,c], Evelyne Martin [a,*]

[a] Univ. Lille, CNRS, Centrale Lille, ISEN, Univ. Valenciennes, UMR 8520 – IEMN, F-59000 Lille, France
[b] Université de Lorraine, CNRS, LEMTA-UMR7563, Vandœuvre-lès-Nancy F-54500, France
[c] Université de Lyon, CNRS, INSA-Lyon, Université Claude Bernard Lyon 1, CETHIL UMR 5008, F-69621 Villeurbanne, France

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ABSTRACT

The hollowing of silicon membranes to form a lattice of cylindrical holes, also called phononic crystal, has been used by several experimental groups willing to fabricate efficient thermoelectric modules. The idea is to reduce the thermal conductivity without impacting the electronic conductivity. For several a priori identical materials, i.e. thin films containing periodic cylindrical holes, drastically different levels of thermal conductivity reduction have been reported in the literature: from 1–2 W $K^{-1}$ $m^{-1}$ to 15–40 W $K^{-1}$ $m^{-1}$, i. e. half the thermal conductivity of the plain membrane. These differences may be due to variations in the geometrical patterns, or to the technological processes specific to each group. It is therefore highly desirable to understand which level of reduction can be expected from the basic concept. In this work, we address the question by applying a fully atomistic framework, the approach-to-equilibrium molecular dynamics (AEMD), to study two deca-nanometric patterns used in the literature and reported respectively with a high and low level of thermal conductivity reduction. For both patterns, the thermal conductivity roughly decreases by a factor 2 only compared to the plain membrane. Thanks to Monte Carlo simulations, in agreement with AEMD for the two patterns, we propose that the origin of stronger reductions could be an increase of the surface roughness during the step of hole fabrication.



## 1. Introduction

Nanostructuration opens perspectives in the field of thermoelectricity to materials that are not a priori good candidates. This is the case for crystalline silicon, a safe and abundant material whose technology has been largely developed and that one would like to use on the same chip to process the information and handle the energy conversion. Several options of nanostructuration have been studied, such as the 1D structuration in nanowires combined with an optimisation of the surface roughness [1–3]. In the same idea of limiting the phonon free paths to decrease the thermal conductivity, the fabrication of nanostructured membranes including nano-holes also raised a great interest [4–8]. In silicon at room temperature, 80% of the total thermal conductivity is provided by phonons that experience mean free paths larger than 100 nm [9,10]. The pattern dimensions (hole diameter and pitch) should ideally be choosen in the range 10–100 nm to act as efficient cutoffs on the phonon MFP distribution and strongly reduce the thermal conductivity with a limited impact on the electronic conductivity (MFPs of a few nanometers). Patterns in this range are however challenging to obtain even by the most advanced nanofabrication techniques, but a cutoff at 500 nm can still suppress half the total thermal conductivity [9].

However, there is a controversy on the level of thermal conductivity reduction resulting from nanostructuration. In Refs. [4,5], values of 1–2 W $K^{-1}$ $m^{-1}$, close to the amorphous limit are reported, while in Refs. [6–8], the thermal conductivity only reduces to 15–40 W $K^{-1}$ $m^{-1}$, i. e. half the thermal conductivity of the plain membrane. The discrepancy can not be attributed to the range of dimensions and volume fraction, since it is similar in Refs. [4,5,7] (between 10 and 100 nm and 10–30%) and in Refs. [4,6,8] (several hundreds of nm and ≈35%). It is therefore highly desirable to be able to distinguish between the effect of nanostructuration and of specific nano-fabrication methods by simulating holey silicon membranes free of any defects but with real dimensions.

In the present work, we focus on two membranes falling in the deca-nanometric range: the membranes of Refs. [5] (hereafter called "Y") and [7] (hereafter called "H"). The characteristic lengths

* Corresponding author.
*E-mail address:* evelyne.lampin@univ-lille.fr (E. Martin).

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of the plain and nanostructured membranes are defined in the schematical representation of Fig. 1. Their values are reported in Table 1. The thermal conductivities measured in Ref. [5] are equal to 17 W $K^{-1}$ $m^{-1}$ for the plain membrane and 1–2 W $K^{-1}$ $m^{-1}$ for the nanostructured membrane, while in Ref. [7], the thermal conductivity are equal to 59 ± 10 W $K^{-1}$ $m^{-1}$ for the plain membrane and 34.5 ± 7.5 W $K^{-1}$ $m^{-1}$ for the nanostructured membrane. The higher absolute values obtained in Ref. [7] can be explained by the greater thickness of their membranes. However, the reduction of thermal conductivity is far more pronounced in Ref. [5] (≈10) than in Ref. [7] (<2).

The thermal conductivity of nanostructured membranes (hereafter called "L") has been studied by molecular dynamics (MD) simulations [11] using the Green-Kubo approach [12]. The patterns Y, H and L are compared in Fig. 2. A thermal conductivity of 45 W $K^{-1}$ $m^{-1}$ has been obtained for the plain membrane, that decreases to 2 to 12 W $K^{-1}$ $m^{-1}$ for hole diameters ranging from $D$ = 3–6.5 nm. The thickness and pitch were set to $t$ = 8.1 nm and $p$ = 8.1 nm. The reduction of thermal conductivity is significative, and ranges from 4 to 25. However the dimensions are considerably lower dimensions than in experiments, and it can not be ruled out that the thermal conductivity strong reduction is due a stronger downscaling.

In order to achieve a better understanding of the thermal conductivity of nanostructured membranes, we have studied the three patterns by using three complementary theroretical approaches, going from the atomistic level (molecular dynamics), to the mesoscopic level (Monte Carlo) and to the continuum level (effective medium theory). This work is presented as follows. Section 2 provides details on the MD method and presents the results. Section 3 is devoted to the other modelling work by Monte Carlo simulations and using the effective medium theory. Section 4 contains the comparison with experiments and discussion of our results. Concluding remarks are collected in Section 5.

## 2. Approach-to-equilibrium molecular dynamics

We have started by studying a plain membrane (hereafter called "P") to obtain a reference of thermal conductivity in absence of nanostructuration. The plain membrane is modeled by an elementary rectangular pattern presented in Fig. 1 (dashed-delimited volume). The atoms are arranged on a diamond lattice and the upper and lower surfaces are smooth. Periodic boundary conditions are applied in the two directions of the membrane

**Table 1**
Thickness $t$, hole diameter $D$ and pitch $p$ of patterns.

| Source | $t$ (nm) | $D$ (nm) | $p$ (nm) |
|---|---|---|---|
| Ref. [5] ("Y") | 22 | 16 | 34 |
| Ref. [7] ("H") | 54 | 20 | 60 |
| Ref. [11] ("L") | 8.1 | 5.4 | 8.1 |

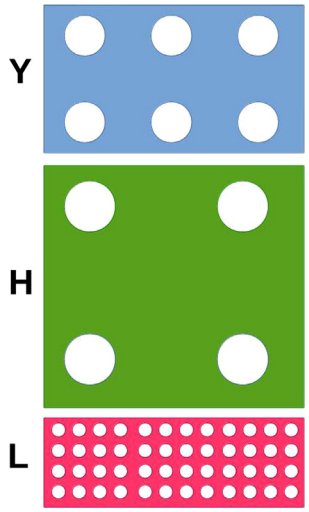


**Fig. 2.** Y, H and L patterns drawn on the same scale.

plane. In the present study, the thickness has been fixed to $t$ = 8.1 nm (15 lattice units) for all the membranes. The square section of the rectangular supercell that models the plain membrane has a side equal to $a = t$ = 8.1 nm. It has been shown [19] that the dependence of the thermal conductivity on the cross section of the simulation cell is weak with the MD methodology presented below. On the other hand, the thermal conductivity exhibits a pronounced dependence on length, both in bulk materials [19] and nanowires [20], and the length $L$ must therefore be increased until the thermal conductivity becomes constant in order to guarantee that the thermal conductivity does not present any size effects inherent to the method, and can be compared to other theoretical or experimental values. In the present case and for the plain membranes, this has required to increase the length $L$ from 80 up to 800 nm.

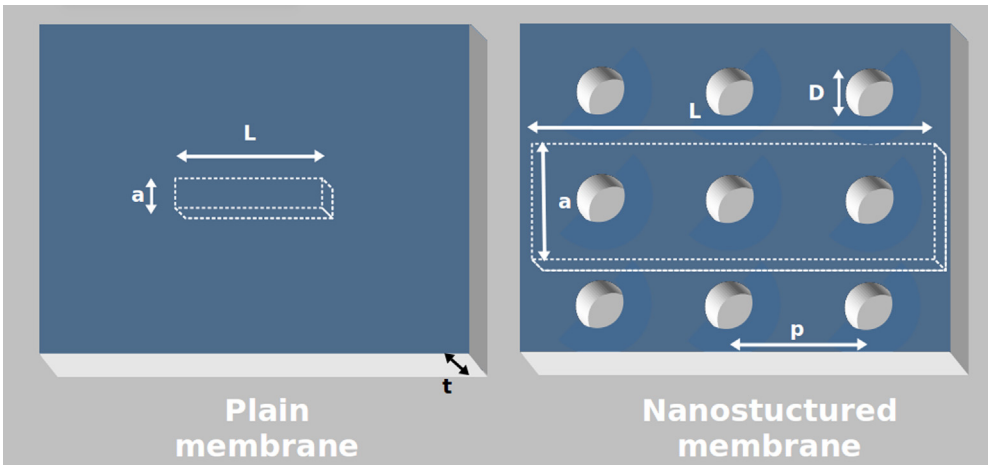


**Fig. 1.** Schematic representation of the plain and nanostructured membranes studied in the present work. $t$ is the thickness of the membrane, $p$ is the pitch of the pattern and $D$ the diameter of the cylindrical holes. The dashed volumes represent the elementary pattern of the simulation boxes (cross section $t \times a$, length $L$, equal to $3p$ in the present case).