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Phonon considerations in the reduction of thermal conductivity in phononic crystals

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Abstract Periodic porous structures offer unique material solutions to thermoelectric applications. With recent interest in phonon band gap engineering, these periodic structures can result in reduction of the phonon thermal conductivity due to coherent destruction of phonon modes characteristic in phononic crystals. In this paper, we numerically study phonon transport in periodic porous silicon phononic crystal structures. We develop a model for the thermal conductivity of phononic crystal that accounts for both coherent and incoherent phonon effects, and show that the phonon thermal conductivity is reduced to less than 4% of the bulk value for Si at room temperature. This has substantial impact on thermoelectric applications, where the efficiency of thermoelectric materials is inversely proportional to the thermal conductivity.

1 Introduction

Size effects significantly impact phonon thermal transport in micro- and nanoscale systems [1]. Thorough understanding of such effects is crucial to the understand of thermal transport in micro- and nanosystems and for continued advancement of nanoscale applications, such as design and development of novel thermoelectric materials [2–5]. In particular, periodic porous structures are known to have strong tunable size effects due to increased surface area [6–8]. While electron thermal size effects have been observed on nanometer length scales in periodic porous structures [9], phonon thermal size effects have been observed on micron length scales [8]. This has substantial impact on phonon transport in phononic crystal structures, which can provide efficient material solutions for thermoelectric applications [10].

In this paper, we numerically study phonon transport in periodic porous silicon phononic crystal structures. In Sect. 2, we develop a model for the phonon thermal conductivity of bulk Si based on the measured phonon dispersion. We then modify the phonon dispersion to include phononic band gaps from 1 GHz–500 GHz, 1 GHz–1 THz and from 1 GHz–4 THz in Sect. 3, and show that a reduction in thermal conductivity due to these "coherent" band gap effects can arise solely from the modified phonon dispersion due to the phononic band gap. Considering incoherent phonon scattering mechanisms and the porosity of the phononic crystal structure, the predicted thermal conductivity of the Si phononic crystal can decrease to less than 4% of that of bulk Si, as discussed in Sect. 4. Since the efficiency of thermoelectric materials is evaluated by the Thermoelectric Figure of Merit, Z, which is inversely proportional to the thermal conductivity, κ, phononic crystals show promise as novel thermoelectric solutions.

2 Thermal conductivity model

To model the thermal conductivity of a Si phononic crystal, we must determine the various phonon scattering times that contribute to the phonon conductivity in Si. For this, we use a procedure to determine the thermal conductivity similar to that outlined by Holland [11, 12]. The thermal conductivity

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is given by

$$\kappa = \frac{1}{6\pi^2}\sum_j \int_0^{q_{\max,j}} \frac{\hbar^2\omega_j^2(q)}{k_B T^2} \frac{\exp[\frac{\hbar\omega(q)}{k_B T}]}{(\exp[\frac{\hbar\omega(q)}{k_B T}]-1)^2} \times v_j^2(q)\tau_j(q)q^2\,dq \quad (1)$$

where $\hbar$ is the reduced Planck's constant, $\omega(q)$ is the phonon dispersion, k_B is the Boltzmann constant, T is the phonon temperature, $v(q) = \partial\omega(q)/\partial q$ is the phonon group velocity, $\tau(q)$ is the scattering time of the phonons, q is the wavevector, and the thermal conductivity, κ is summed over $j = 3$ modes (one longitudinal and two transverse). To evaluate this expression, we must determine the Si dispersion and scattering times. We use measured Si dispersion data in the (100) direction [13, 14] and fit the data to a 4$^{\text{th}}$ degree polynomial for an analytical expression for $\omega(q)$ and $v(q) = \partial\omega(q)/\partial q$ [12]; we assume a doubly degenerate transverse branch. In bulk Si, phonon scattering is dominated by Umklapp scattering, impurity scattering, and boundary scattering. These scattering times are given by [15]

$$\frac{1}{\tau_{\text{Umklapp},j}} = BT\omega_j^2(q)\exp\left[\frac{C}{T}\right] \quad (2)$$

$$\frac{1}{\tau_{\text{impurity},j}} = D\omega_j^4(q) \quad (3)$$

$$\frac{1}{\tau_{\text{boundary},j}} = \frac{v_j(q)}{E} \quad (4)$$

where B, C, D, and E are constants determined by fitting (1) to the data. Equations (2)–(4) are related to the phonon scattering time in (1) via Matthiessen's Rule, so that

$$\frac{1}{\tau_j(q)} = \frac{1}{\tau_{\text{Umklapp},j}} + \frac{1}{\tau_{\text{impurity},j}} + \frac{1}{\tau_{\text{boundary},j}} \quad (5)$$

Given (5) with (1), the thermal conductivity of Si is calculated and the coefficients in (2)–(4) are iterated to achieve a best fit with measured data on bulk Si [16]. The fit to the data is shown in Fig. 1.

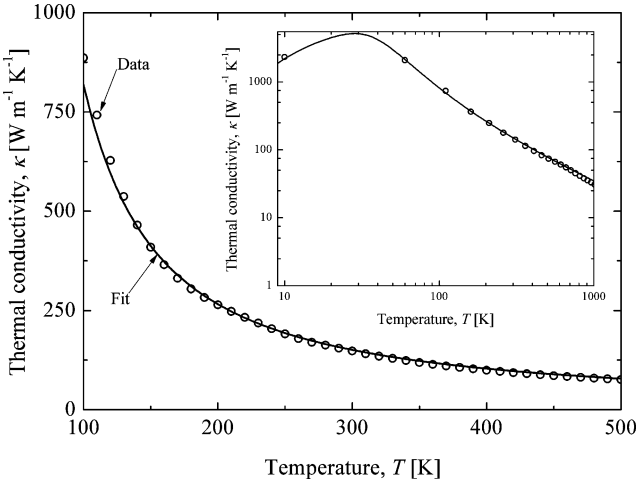

Fig. 1 Thermal conductivity model fit to experimental data on single-crystalline Si [16]. The best fit coefficients for the various scattering times ((2)–(4)) are $B = 3.73 \times 10^{-19}$ s K^{-1}, $C = 157.3$ K, $D = 9.32 \times 10^{-45}$ s^3, and $E = 2.3 \times 10^{-3}$ m

3 Coherent effects on the phononic crystal thermal conductivity

With all the parameters determined for the case of bulk Si, we now modify the phonon dispersion to imitate reflection or blocking of certain frequencies expected in the phononic crystal. We consider the three different cases of modified phonon dispersion: Case a—blocked frequencies from 1 GHz–500 GHz; Case b—blocked frequencies from 1 GHz–1 THz; and Case c—blocked frequencies from 1 GHz–4 THz. Figure 2a shows the calculations of the thermal conductivity of Si using (1) with these modified dispersion relations over a wide range of temperatures (10–1,000 K). Figure 2b shows the fraction of the "bulk" (unmodified dispersion) thermal conductivity for each of the three cases in the temperature regime of interest for thermoelectric devices in most terrestrial microelectronic applications (100–500 K). The reduction in thermal conductivity due to blocking certain phonon frequencies is apparent, especially at lower temperatures. At room temperature, significant reduction is realized by blocking longitudinal and transverse modes up to 4 THz.

A useful exercise is to analyze the spectral contribution to the thermal conductivity to evaluate why blocking certain phonon states will cause a reduction in thermal conductivity [17]. The spectral contribution to thermal conductivity is given by [12]

$$\kappa_{\omega,j} = \frac{1}{6\pi^2}\frac{\hbar^2\omega_j^2(q)}{k_B T^2}\frac{\exp[\frac{\hbar\omega_j(q)}{k_B T}]}{(\exp[\frac{\hbar\omega_j(q)}{k_B T}]-1)^2} v_j(q)\tau_j(q)q^2 \quad (6)$$

Here the transverse spectral thermal conductivity is multiplied by 2, since we assume doubly degenerate transverse branches. Figure 2c shows the fraction of the total spectral thermal conductivity at 300 K for a bulk Si dispersion and the three cases of dispersion modification (Cases a, b, and c). This fraction is defined as $\kappa_{\omega,j}/\sum_j\sum_\omega \kappa_{\omega,j}$. In bulk Si, the contribution from the transverse modes is much greater than that from the longitudinal modes below 4 THz. As the phononic bandgap increases, the dominant phonon frequencies participating in transport increase. Blocking up to 4 THz eliminates nearly all of the transverse phonon modes, which explains the large decrease in the thermal conductivity seen in Figs. 2a and b.

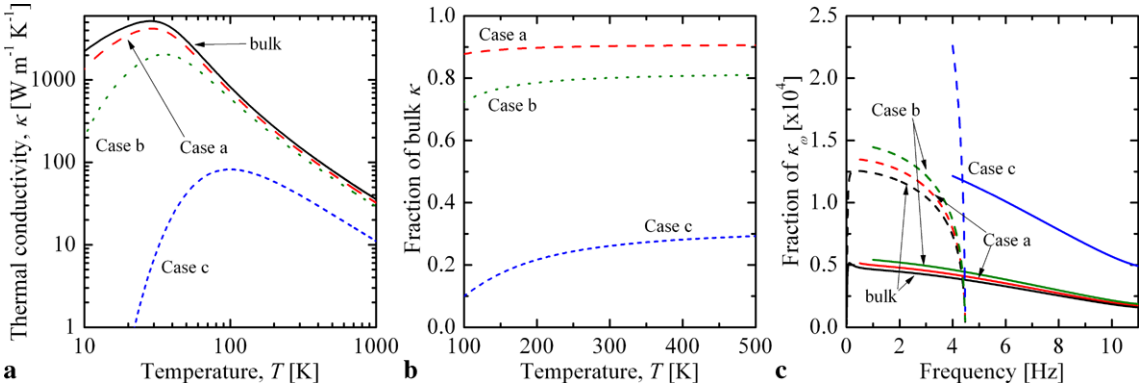

Fig. 2 (**a**) Thermal conductivity of Si assuming an unmodified dispersion relation (bulk), blocked frequencies from 1 GHz–500 GHz (Case a), blocked frequencies from 1 GHz–1 THz (Case b), and blocked frequencies from 1 GHz–4 THz (Case c). (**b**) Fraction of the "bulk" (unmodified dispersion) thermal conductivity for each of the three cases in the temperature regime of interest for thermoelectric devices in most terrestrial microelectronic applications (100–500 K). (**c**) Fraction of the total spectral thermal conductivity at 300 K for a bulk Si dispersion and the three cases of dispersion modification (Cases a, b, and c). The longitudinal modes are represented by the *solid lines* and the transverse modes by the *dashed lines*

4 Incoherent effects on the phononic crystal thermal conductivity

We now turn our attention to the effects of incoherent, classical scattering from the geometry of the phononic crystal. Our crystal will have periodic "holes" or large vacancies that are arranged to block the various phonon frequencies. Therefore, further reduction in thermal conductivity will occur from classical boundary scattering along with reducing the amount of bulk material in the crystal (i.e., the porous nature of the phononic crystal). To simulate this, we consider the approach to analyzing a porous nanowire matrix discussed by Hopkins et al. [9]. In this approach, we introduce an addition boundary scattering mechanism in (5), given by

$$\frac{1}{\tau_{\text{porescattering},\,j}} = \frac{v_j(q)}{L} \tag{7}$$

where L is linear distance between scattering sites. Note that this differs from (4) since L is not a fitting parameter. The thermal conductivity is then calculated by adding (7) into (5), and calculating the thermal conductivity via (1).

Now, let's consider the effects of boundary scattering for the phononic crystals discussed in this work. The scattering sites are the voids in the Si crystals that occur at periodic distances, so mathematically we treat them similar to grain boundaries via (7) [18, 19]. We estimate the center-to-center spacing of the vacancies in the phononic crystal as $a = \pi v/f$, where v is the average speed of sound in Si, which we estimate as $v = 6{,}545$ m s $^{-1}$ by averaging the zone center phonon velocities of the longitudinal and transverse branches [20], and f is the upper limit of the phononic band gap. Assuming $r/a = 0.4$, which is a reasonable ratio of the vacancy radius, r, to the center-to-center spacing for a Si phononic crystal with evacuated periodic inclusions [10], the linear distance between the scattering sites (edges of the periodic vacancies) is estimated as $L = a - 2r$. Therefore, the scattering distances for Cases a, b, and c are $L = 8.2$, 4.1, and 1.0 nm, respectively.

Finally, we consider the thermal conductivity reduction due to physically removing areas of the crystal to create the phononic lattice. This can be modeled as a reduction due to porosity; to calculate the effects of the porosity, we use the effective medium approximation derived by Eucken for a solid with cylindrical pore inclusions given by [21]

$$\frac{\kappa_r}{\kappa_s} = \frac{1-p}{1+\frac{2}{3}p} \tag{8}$$

where κ_s is the thermal conductivity of the corresponding solid assuming no porosity (i.e., (1) with (5) and (7)), κ_r is the resultant thermal conductivity of the porous structures, and p is the porosity. For a square phononic crystalline lattice with $r/a = 0.4$, we estimate the porosity from geometrical considerations as $p = 0.64$. This approach of accounting for the thermal conductivity reduction due to porosity has been validated in periodic porous Si films [12].

Using (8), with (7) and (5) in (1), we calculate the thermal conductivity of a Si phononic crystal accounting for coherent and incoherent effects for the three cases. The resulting thermal conductivity as a function of temperature is shown in Fig. 3a, and the corresponding fraction of the bulk thermal conductivity for each case is shown in Fig. 3b. We have previously studied the spectral conductivity due to incoherent effects, and so omit the analysis here, but refer the reader to our previous work for further analysis of how incoherent scattering reduces the thermal conductivity of bulk Si at different frequencies [12]. The incoherent effects reduce the thermal conductivity of the Si phononic crystals

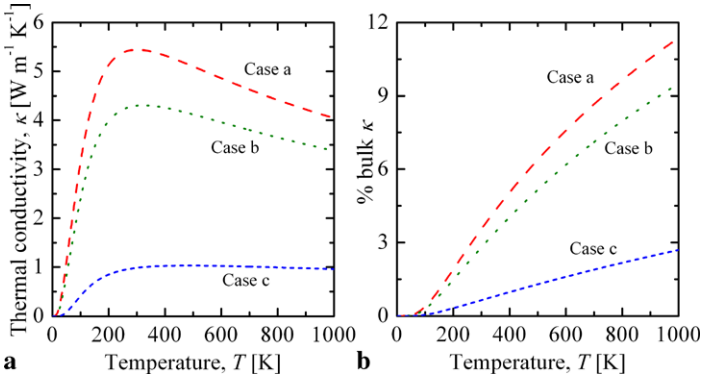

Fig. 3 Thermal conductivity of a Si phononic crystal accounting for coherent and incoherent effects for the three modified dispersion cases in Fig. 2: (**a**) thermal conductivity as a function of temperature; and (**b**) corresponding fraction of the bulk thermal conductivity

even further, and cause a greater reduction in κ than the coherent effects. However, coherent and incoherent effects together substantially reduce the thermal conductivity of the phononic crystal, leading to reductions in κ to 0.7–3.5% of the bulk value at 300 K. In practice, Case c is extremely difficult to fabricate, however, Cases a and b are more feasible for device engineers. Assuming a thermoelectric module designed from silicon phononic crystals with coherent properties of Case a, the reduction in the phonon thermal conductivity to 3.5% of the bulk value leads to an enhancement in Z by a factor of 28.5, showing that phononic crystals offer a unique material solution to enhancing thermoelectric applications. Note that, in this work, we consider a bulk Si dispersion as the basis of our thermal conductivity model. In nanostructured materials, the phonon dispersion will be different than that of bulk [22]; however, this change in dispersion will result in a reduction in the phonon group velocity which would create an even further reduction in the thermal conductivity of the phononic crystal. Therefore, the model developed in this work based on a bulk Si dispersion is considered an upper limit to the thermal conductivity of phononic crystals.

5 Conclusions

We numerically study phonon transport in silicon phononic crystals. We develop a model for the thermal conductivity of phononic crystal that accounts for both coherent and incoherent phonon effects, and show that the phonon thermal conductivity can be reduced to less than 4% of the bulk value for Si at room temperature. This has substantial impact on thermoelectric applications, as a reduction in thermal conductivity will increase the thermoelectric figure of merit.

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