Heat conduction in nanostructured materials

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Abstract
An overview of the direct and ab initio simulation as well as experimental tools addressing heat conduction in dielectric nanostructures and nanostructured materials is proposed. Recent advancements based on density functional theory to compute phonon properties and thermal conductivity are exposed and the state of the art in metrology is detailed. Finally, new progresses in the experimental demonstration of coherence effects in heat conduction are illustrated, emphasizing the open questions in the still poorly understood field of phononic heat transport.

Key words: Phonons, Nanostructures, Superlattices, Density functional theory, Molecular dynamics, 3 omega, TDTR, MEMS, Phononic crystals, Thermocrystals, Phonon coherence

1. Introduction

1.1 Context

The Laws of Heat Transfer have been under consideration since more than two centuries and they have been guiding considerable discoveries in the field of physics, be it the start of conceptualization of quantum mechanics through Planck’s Law, the difficult birth of phonons to explain heat diffusion in solids or the still not resolved issue of turbulence. In the second half of the twentieth century, sciences of thermal engineering were summarized and established based on key Laws –Fourier Law for conduction, Stefan-Boltzmann Law for radiation, and Newton’s Law for convection to say a few-, those principles proving to be robust and accurate in a sufficient extent and in a vast range of technical problems.

With the formidable and accelerated integration of transistor devices started in the sixties –about 9 billions CPUs are in function today- and the mid-heighties apparition of nanosciences, which finally benefited mostly to the materials community, the question of the thermal management of small systems and of the micro-to-nanoscale thermal design of materials led to the necessity of revisiting heat transfer principles.

While convection at small scales reduces to well-known conduction, radiation and heat conduction undergo fundamental modifications. In this article, we have chosen to only consider the alteration of heat conduction and more specifically phonon heat conduction at short space scales. Heat is carried by free electrons and phonons in solids. Phonons are quanta of the atomic vibrational energy and they significantly contribute in a broad range of materials, including those of interest in nanotechnology, while electrons are predominant in metals only. The electronic thermal conductivity did however not draw considerable attention since it is directly yielded from Lorentz’s Law and the electrical conductivity, the physics of which has been mastered in the broadest set of situations for several decades. Phonons are more difficult to apprehend as they are non-local quasi-particles like photons, but unlike those, phonons are strongly interacting together through inelastic or anharmonic three- or four-body interactions and there typical
wavelength at room temperature remains lower than 10nm (whereas photon Wien’s wavelength is of ten microns). Phonon thermal conductivity has nevertheless been defined rather successfully based on a quasi-particle flux yielding a sum over the mode spectrum –typically from zero to several TeraHertz- of the mode heat capacity times the mode group velocity times the mode mean free path similarly to what is done in the classical kinetic theory of gases. When sizes shrink, heat conduction in crystals is altered by two main physical effects, i.e. phonon confinement (Montroll, 1950; Baltès et al., 1973) and ballistic regime of transport (Cahill et al., 2003). This latter effect appears when the system size ranges in the same order of magnitude as the phonon mean free path, which covers a wide interval from a few nanometers to a few microns. In this regime, the surface or interface scattering becomes predominant compared to the internal scattering, (Hochbaum et al., 2008; Chen et al., 2008). Thermal wavelengths comparable to the system size lead to confinement, which in turn yields to a reduced phonon transport due to group velocity decrease but also to relaxation time modification (Kazan et al., 2010). Alteration of heat conduction is significant at room temperature in micro-to-nanoscale devices but also at low temperatures in macroscopic systems because mean free paths and thermal wavelengths are reversely proportional to temperature.

1.2 Status and challenges
The above-mentioned mechanisms remain to be completely proven theoretically and experimentally. While the impact of surface and interface scattering has been shown (Volz, 2009), the detailed descriptions of the phonon wave mechanisms occurring at the boundary remain to be established. Confinement has been observed in nanowires and films (Kazan et al., 2010) and even stronger proofs have been provided regarding heat conduction in superlattices (Luckyanova et al., 2012; Ravichandran et al., 2014). If modeling of realistic phonon properties such as relaxation times, have recently underwent progresses owing to density functional theory approaches (Ward et al., 2009), their estimations however remain limited to periodic or few atoms systems. Their implementation is then restricted to Boltzmann based thermal conductivity formula where a long list of approximations have to be accepted, i.e. (i) single mean relaxation time (ii) local near-to-equilibrium approximations (iii) local occupation number, (iv) ad hoc or fitted boundary scattering rates and (v) neglecting the cross mode correlations, which have been proven to play a key role in low dimensional systems. A less approximated approach of thermal conductivity calculation is based on classical Molecular Dynamics (Ladd et al., 1986; Volz et al., 1996) that directly provides the atom trajectories and consequent heat fluxes and temperatures. It is however limited to a few millions atoms –i.e. a few nanometers three-dimensional objects- over a few nanoseconds and more importantly, it depends on the validity of the interatomic pseudopotential. This method is of course not representing quantum populations and is discarding the contribution of free electrons. Consequently, a remaining fundamental unknown mechanism is the one of the metal/dielectric interfacial resistance, which involves both electrons and phonons, but also their coupling.

Experimental techniques –be it based on the 3 omega (Blanc et al., 2013) and the time-domain thermoreflectance techniques (Koh et al., 2007) or the scanning thermal microscopy (Lefèvre et al., 2005) - have proven robustness in estimating the thermal conductivity, thermal conductance and interfacial properties. However, the experimental estimations of TeraHertz modal phonon properties remain to be achieved and recent attempts tend to confirm the practicability of those measurements (Minnich et al., 2011), either by varying the frequency and the width of a pump beam –maximal frequencies remaining below the TeraHertz here- or by designing single modes cavities (Legrand, 2014). In the following, the most advanced studies in thermal conduction modeling based on Density Functional Theory and Molecular Dynamics leading to the direct computation of phonon mean free path, group velocity, density of states and thermal conductivity will be presented in part 2, and the to-date room to low temperature experimental measurements of thermal conductivity will be exposed in part 3 showing the impact of surface scattering and phonon confinement also termed as coherent.

2. Multiscale phonon transport calculations

2.1 Interatomic force constants from first principles
Quantitative calculation of phonon transport properties and lattice thermal conductivity had been challenging even for single crystals because it requires a rigorous treatment of anharmonic interatomic force constants (IFCs). However, with the development of the density functional theory (DFT), accurate calculation became possible, first for materials
with light elements and high symmetry such as silicon and germanium single crystals (Broido, et al., 2007; Ward, et al., 2009; Esfarjani and Stokes, 2008; Esfarjani et al., 2011), and more recently for materials with heavier elements and more complex structures (Shiomi, et al., 2011).

The anharmonic IFCs of a crystal can be defined by Taylor-expanding the force acting on the \( i \)th atom in the direction \( \alpha \) with respect to the displacements \( \nu \) of the neighboring atoms as,

\[
-F^\alpha_i = \Pi^\alpha_i + \sum_{j,k} \Phi^{\alpha j k} u_j^\beta u_k^\gamma + \frac{1}{2!} \sum_{j,k,l} \Psi^{\alpha j kl} u_j^\beta u_k^\gamma u_l^\delta + \frac{1}{3!} \sum_{j,k,l,m} \Theta^{\alpha j klm} u_j^\beta u_k^\gamma u_l^\delta u_m^\epsilon \ldots
\]

Here, \( \Phi, \Psi, \) and \( \Theta \) are harmonic, cubic, and quartic IFCs, respectively. The integers \( i, j, k, \) and \( l \) are the atom indices, and \( \alpha, \beta, \gamma, \) and \( \delta \) represent the Cartesian components. Although IFCs of any orders can be defined, it is rare to go beyond the quartic terms because lattice thermal conductivity of most of the materials can be sufficiently reproduced by IFCs up to cubic terms, and strongly anharmonic phonon-phonon scattering in some materials such as lead telluride (PbTe) (Delaire, et al., 2011) can be well described by additional quartic terms (Shiga, et al., 2012; Shiga, et al., 2014).

There are several ways to calculate anharmonic IFCs. Most rigorous method is to use the density functional perturbation theory (DFPT) (Broido, et al., 2007). This method finds IFCs of crystals through the 2n+1 theorem (Gonze and Vigneron, 1989), e.g., cubic IFCs can be calculated from the linear response of the wave function. More facile method is the real space displacement method (Esfarjani and Stokes, 2008; Esfarjani et al., 2011), where the anharmonic IFC matrix are calculated by fitting it to a set of force-displacement data obtain by DFT calculations of a supercell with various atomic displacements. In the real space method, the cut off length of IFCs (number of neighboring shells of a given atom) are chosen separately for harmonic and anharmonic IFCs to minimize the fitting residual within the computationally affordable range. Note that the appropriate cut off length depends strongly on the materials of choice and the target properties (Lee, et al., 2014). Another intuitive method is to compute IFCs by taking derivative of lower order IFCs, e.g. to compute cubic terms by taking finite difference of harmonic terms (Tian, et al., 2012). All the methods have merits and demerits, but they result in IFCs of similar accuracy when properly performed. On investigating strongly anharmonic lattices, the real space displacement method may have advantage since it takes relatively small additional effort and computational load to calculate IFCs beyond cubic terms.

2.2 Anharmonic lattice dynamics

Using the harmonic IFCs, the phonon dispersion relations can be calculated through the dynamical matrix. This then gives the phonon group velocities \( v^s_\k \) for arbitral wavevector \( \k \) and branch \( s \). Furthermore, phonon relaxation time \( \tau^s_\k \) of the 3 phonon scattering process (including both normal and Umklapp processes) can be calculated from the cubic IFCs. For a single crystal, phonon relaxation times are typically calculated by the (anharmonic) lattice dynamics method. Here, relaxation time is calculated through generation and annihilation probability given by the Fermi’s Golden rule. Again, it is often enough to consider only the first-order 3-phonon process, which is the leading source of intrinsic thermal resistance in many single crystals, although this could depend on materials and the temperature range. As for the validation of the calculations, while it is a common practice to compare the calculated dispersion relations to that measured by inelastic neutron scattering experiments, it has been further demonstrated for some materials such as PbTe that the calculated phonon scattering rate is in reasonable agreement with the line width obtained by the inelastic neutron scattering experiments for representative phonon modes (Shiga, et al., 2012; Shiga et al., 2014).

Figure 1(a) shows the calculated frequency-dependent phonon relaxation time \( \tau \) of various materials. The overall profiles of the phonon relaxation time can be characterized by three different frequency regimes: (1) the low frequency regime, where the relaxation time is inversely proportional to squared frequency for acoustic phonons with linear dispersion relations as modeled by Klemens (Klemens, et al., 1951), (2) the intermediate frequency regime, where the dispersion relations are no longer linear and the phonon relaxation time exhibits a maximum due to increase in the joint density of states of 3-phonon process, and (3) the high frequency regime, where the phonon relaxation time decreases steeply with increasing frequency for optical phonons with small dispersion. Of course, more detail features depend on the materials. For instance the difference between the LA and TA phonons is much more significant in PbTe than the others due to a selective phonon scattering of the small-wavevector LA phonons due to strongly anharmonic transverse optical phonons in PbTe (Shiga, et al., 2012).

The phonon mean free path can be calculated by multiplying \( \tau \) with the absolute value of group velocity, \( \Lambda^{s_\k} = |v^s_\k| \tau^{s_\k} \). Figure 1(b) shows that the phonon mean free path spans over many orders of magnitude depending on the phonon
frequency, indicating a need for multiscale approach to calculate lattice thermal conductivity in nanostructures.

With the phonon transport properties, lattice thermal conductivity can be obtained based on the kinetic theory of phonon gas. The linearized Boltzmann equation with the relaxation time approximation gives the thermal conductivity as,

$$\kappa_{\text{ph}} = \frac{1}{3V} \sum_{k\nu} C_{k\nu} |v_{k\nu}|^2 \tau_{k\nu}$$  \hspace{1cm} (2)

where $C$ and $V$ are heat capacity per mode ($k\nu$) based on Bose-Einstein statistics, and volume of the material, respectively. The approximation has been confirmed to successfully reproduce the measured temperature dependence of lattice thermal conductivity for many materials (Broido, et al., 2007; Ward, et al., 2009; Esfarjani and Stokes, 2008; Esfarjani, et al., 2011; Shiomi, et al., 2011).

The knowledge of mode-dependent phonon transport properties allow us to discuss the influence of nanostructures on thermal conductivity. To this end, it is useful to plot the cumulative thermal conductivity (Dames and Chen, 2005) to illustrate the contribution of phonons with specific mean free path to thermal conductivity,

$$\kappa_c(\Lambda_0) = \frac{1}{3V} \int_{\Lambda_{\text{min}}}^{\Lambda_0} C_v \Lambda d\Lambda.$$  \hspace{1cm} (3)

Here, the contribution from phonons with different phonon mean free paths is accumulated from $\Lambda=\Lambda_{\text{min}}$ to $\Lambda=\Lambda_0$. Figure 1(c) shows the cumulative thermal conductivity normalized by the bulk thermal conductivity for various materials. It can be seen that the cumulative thermal conductivity continuously increases with $\Lambda_0$ and the range of phonon mean free paths with noticeable contribution to thermal conductivity is large as discussed earlier. The detail profile depends on the materials but the general features appear to be self-similar, suggesting the existence of universal scaling laws (Freedman, et al., 2013). Self-similarity is particularly pronounce in short and long mean free path regimes, and in each regime, it has been shown that cumulative thermal conductivity profiles can be collapsed on a master curve by scaling the mean free path with the characteristic length derived using phenomenological models (Aketo, et al., 2014).

The cumulative thermal conductivity gives insight into the potential ability of the nanostructures to reduce thermal conductivity through the size effect. Such means to reducing thermal conductivity without altering electrical conductivity is important to improve the material thermoelectric conversion efficiency. If we base on the picture of nanostructure sketched in Fig. 1 (c), the diffusive phonons ($\Lambda<L$) are scattered by other phonons to the same extent as in the single crystal even with nanostructures, but the ballistic phonons ($\Lambda>L$) are scattered at the interface much more frequently than by other phonons. For instance, in case of nanostructured ZrCoSb half Heusler compounds with $L=40$ nm, the phonons that would contribute to 50% of lattice thermal conductivity in case of single crystal becomes ballistic and can be strongly influenced by the interface (size effect) (Shiomi, et al., 2011). On the other hand, in case of PbTe, to inhibit phonons with 50% contribution to lattice thermal conductivity by the size effect, $L$ needs to be less than 10
nm. Synthesizing such nanograins is challenging, and even if we could, that would also terminate transport path of electrons, and thus, this calculation result suggests that the appropriate class of nanostructure for PbTe is not the nanograins but a matrix with nanoinclusions with paths left for electrons to transport in between the nanoinclusions. Recently such structures have been synthesized by precipitating SrTe nanocrystals inside the PbTe matrix, and $ZT=1.7@500^\circ \text{C}$ has been achieved (Biswas, et al., 2011).

### 2.3 Molecular dynamics

While lattice dynamics describes dynamics of phonons in the wavevector-space representation, it is sometimes more convenient to use the real space representation. Alloy crystals, an often used materials for thermoelectrics, is a material of that kind. A common strategy there is to randomly substitute a specific element with a homologous element to reduce lattice thermal conductivity primarily using mass difference scattering without severely altering the electronic states. The effect of mass difference can be treated by first order perturbation of Hamiltonian of lattice dynamics (Tamura, 1983), and it has been shown to successfully reproduce the measured thermal conductivity reduction in silicon germanium (Garg, et al., 2011). On the other hand, when the mixing ratio is large, as it is typically the case in many experiments, the validity of the perturbation theory in not evident. In that case, classical molecular dynamics simulations with the force fields of Eq. (1) can be useful as it allows us to handle higher order terms relatively easily. In addition, being a method solving atomistic trajectories in real space, molecular dynamics method allows us to account for the local change in the forces fields (Murakami, et al., 2013). Although the drawback of using classical molecular dynamics is that the statistics is classical, most of the thermoelectric materials for energy generation is expected to be used in the temperature range with little quantum effect. The MD calculations have been performed for various alloys such as half-Heusler compounds (Shiomi, et al., 2011; Yan, et al., 2012) and PbTe$_{1-x}$Se$_x$ lead chalcogenide (Murakami, et al., 2013) and good agreement with the experiments was confirmed.

![Fig. 2 Multiscale phonon transport calculations](image)

### 2.4 Monte Carlo

While the cumulative thermal conductivity is useful to gain knowledge in potential reduction of lattice thermal conductivity, there is certainly limitation in providing qualitative understanding and predictions since phonon scattering processes in the actual nanostructured materials can be more complicated with phonons reflecting or transmitting at the interface depending on the phonon states and interface characteristics. One way to handle the complication is to directly solve the Boltzmann transport equation using Monte Carlo method (Mazumdar and Majumdar, 2001; Lacroix, et al., 2006; Hao, et al., 2009), which is capable of accounting for the complex interface geometries. With this, we can formulate multiscale phonon transport calculation as illustrated in Fig. 2.

For phonon transport, the following linearized Boltzmann transport equation with relaxation time approximation is commonly used.
Here, \( f(\omega) \) is phonon distribution function, \( f_0 \) is the Bose Einstein distribution function, \( \mathbf{e} \) is the unit vector in the direction of phonon transport. By plugging in the phonon group velocities and relaxation times calculated by the above lattice dynamics method, simulations with non-empirical bulk properties can be realized. There are also ways to solve the collision terms more rigorously by directly combining the equation with lattice dynamics. However, in case of phonon transport, the relaxation time approximation is known to be reasonable expect for limited materials with high thermal conductivity (Lindsay, et al., 2010). In the Monte Carlo simulations, phonons are transported according to their group velocity, and scattered by phonon-phonon scattering inside the grain with probability of 

\[
\Delta f_s(\omega, \mathbf{r}) = \frac{f_s(\omega, \mathbf{r}) - f_0(\omega, T)}{\tau_s(\omega, T)}
\]

within the duration \( \Delta t \). When a phonon is scattered, the phonon states (frequency, polarization, group velocity, direction) is reset to the equilibrium distribution of the local temperature. When the phonon reaches the interface, the phonon is transmitted or reflected following the given transmission probability in the direction stochastically determined by the scattering characteristics (e.g. specular or diffusive). The simulation has been used for instance to evaluate lattice thermal conductivity of PbTe matrix with embedded nanoinclusions (Hori, et al., 2014).

By using the energy-based variance-reduced Monte Carlo formulation (Peraud and Hadjiconstantinou, 2011), the simulations can be greatly speeded up, enabling direct calculations of mesoscale systems. Note that there are also simpler and quicker methods to sample the mean free path without solving the distribution function (McGaughey and Jain, 2012; Hori et al., 2015) for systems where the distribution can be assumed to be at equilibrium, and mode conversion due to scattering is not important.

3. Experimental techniques and outcomes

3.1 Measurement methods

Effective thermal conductivities of nanostructured materials have been well measured by differential 3ω method (Cahill, et al., 1994), periodical-laser heating method (Hatta, 1990), thermoreflectance method (Taketoshi, et al., 1997) and so on. The MEMS based suspended heater-thermometer method (Shi, et al., 2003) is a useful tool for the thermal conductivity measurements of single nanostructures. The thermal conductivities of various nanostructures such as, nanotube, nanowire, Si based films thermal conductivities were measured. The nanostructured materials are supported between two pads with resistive heater-thermometer metal strip wires as shown in Fig. 3. One pad is joule heated using a metal strip wire, and thermal energy is transferred to another pad through the nanostructures. Temperatures of both pads are measured via the electrical resistance of metal strip wires, and the applied thermal energy is directly calculated by joule heating. The effective thermal conductivity of the nanostructured material is measured by temperature difference, the size (length, cross sectional area) of the nanostructures, and thermal energy. For the sake of simplicity, the Fourier Law is assumed even if the thermal energy transport is not diffusive to yield an effective, i.e. non intrinsic size dependent, thermal conductivity. A similar MEMS based thermal conductivity measurement follows the T-type nanosensor method proposed for a carbon nanotube thermal conductivity measurement (Fujii, et al., 2005).

The differential 3ω method is often used for the cross-plane thermal conductivity measurement of thin films, such as superlattice thin films (Borca-Tasciuc, et al., 2000), nano-composites (Putnam, et al., 2003), thermoelectric thin films (Takashiri, et al., 2007) and so on. The configuration of the 3ω method is illustrated in Fig.4. The metal strip wire (~20μm width, ~5mm length) is deposited on the thin film with an electrical insulator. The AC current is applied to the metal strip wire for heating the sample, and the temperature response of the heated sample is measured by the electrical resistance of the same metal strip wire. The temperature rise of the sample with the thin film is measured, and the measured temperature rise is compared with that of the reference sample (without the thin film). The thermal resistance of the thin film is measured by the 3ω method, and the effective thermal conductivity is calculated from the thermal resistance. Thermal boundary resistance between the nanostructured materials and the substrate may affect the above thermal conductivity measurements, however those effects on the thermal transport is included in the total thermal resistance. The effective thermal conductivity is therefore underestimated.

In the past decade, Time-domain ThermoReflectance (TDTR) method has been significantly developing, and is applied to the effective thermal conductivity measurements of nanostructured thin films (Chiritescu, et al., 2007). The sample is coated by a metal film, and the metal coated material is heated by the pump laser. The reflectivity of the
metal depends on temperature. Therefore the surface temperature of the material can be measured by the probe laser. The effective thermal conductivity is determined by the time response of the reflected signal of the probe laser. The heat conduction equation (diffusion equation) is assumed in the TDTR method, and the thermal boundary resistance and the effective thermal conductivity are unknown parameters. The measure time dependent temperature curve is best fit by the above mentioned parameters. The thermal boundary resistance can be determined by TDTR, and a wide range of thermal conductivity can be measured. For example, it is difficult to measure the high thermal conductivity of a film by 3ω method due to its small thermal resistance. TDTR can instead be applied to a small sample in various conditions such as high pressure in a diamond anvil cell (Yagi et al., 2011) or as in a closed chamber with low/high temperature because the measurement is a purely non-contact method. TDTR is known as a powerful tool eventually yielding the spectral phonon mean free path.

Scanning thermal microscopy is another direction for thermal properties measurements of nanostructured materials (Nakabeppu et al., 1995). The temperature sensor (thermocouple, thermistor) is fabricated on the probe of an Atomic Force Microscopy (AFM). The temperature spatial resolution is up to ~10nm. Non-contact AFM based on near-field optical probes have also have been developed in this decade (Taguchi et al., 2004, Wilde et al., 2006). The critical issue of the SThM is the temperature calibration. To solve the artifact due to the thermal resistance between the SThM probe and the sample, the temperature of the SThM probe can be controlled by a micro-machined heater on the tip (Nakabeppu, 2002). Otherwise the temperature distribution of the measured surface with SThM probe with the adsorption layer should be analyzed for accurate temperature measurements (Shi and Majumdar, 2002). Some researchers have carried out the SThM measurement in a vacuum chamber to remove the adsorption layer on the surface for the measurement simplification (Hinz et al., 2007).

3.2 Measured thermal conductivity of nano-structured materials

The measured thermal conductivities of nano-porous Si thin films are summarized in Fig. 5. The thermal conductivities of porous Si thin film are much lower than that of bulk Si (~140W/(m·K)). Even the thermal conductivity of Si thin film without any holes is lower than the bulk value, which is partially explained by the classical phonon transport model (Sondheimer, 2001). The mean free path of phonons is the fitting parameter in the equations, and it reaches the micron to fit the experimental results (Hagino, 2014). In its mode averaged version, the thermal conductivity \( k \) can be expressed as the product of heat capacity \( C \), group velocity of phonons \( v \), and phonon mean free path \( l \) as follows:

\[
k = \frac{1}{3} C v l \tag{1}
\]

Thermal conductivity \( k \), heat capacity \( C \), and group velocity \( v \) (sound velocity) can be measured experimentally. The mean free path \( l \) is simply calculated by Eq.(1), but it is too short to explain the measured effective thermal conductivity of the nanostructured materials (JSTP ed., 2014). Molecular dynamics and lattice dynamics are currently carried out to understand the effective thermal conductivity. Their calculated results are well consistent with each other, and the mean free path of phonons is much longer than the expected mean free path estimated by Eq.(1). The spectral model for mean free path is useful to reach a simple understanding (Dames and Chen, 2004).

\[
k = \int_{0}^{\infty} \frac{1}{3} C(\omega)v(\omega)(\omega) d\omega \tag{2}
\]

Phonon is the quantum of the lattice vibration in solids. Therefore phonon heat capacity, phonon group velocity and

| Material      | Diameter[nm] | \( k \) [W/(m·K)] | Method  | Reference         |
|---------------|--------------|-------------------|---------|-------------------|
| Si            | 52           | 1.6               | MEMS    | Hochbaum et al.   |
| Si            | 10×20(square) | 0.76              | MEMS    | Boukai et al.     |
| Si            | 78           | 31                | Raman   | Doerk et al.      |
| Ge doped Si   | 100          | 6.9               | -       | Pan et al.        |
| Si/SiGe SLs  | 58           | 6                 | MEMS    | Li et al.         |
| GaAs          | 165          | 19                | Raman   | Soini et al.      |
| InAs          | 100          | 6.4               | -       | Ren et al.        |
| ZnO           | 70           | 7.5               | MEMS    | Bui et al.        |
Mean free path of phonons are the function of frequency of vibration. The Debye model can be applied for heat capacity and group velocity. The function of mean free path $l(\omega)$ had been well investigated (Tritt ed., 2004). Several coefficients are included in the mean free path $l(\omega)$, and those are determined by the experimental results. The mean free path of phonons calculated by the spectrum model with experimental results is much longer than the value from the simple calculation, and the long mean free path is consistent with the low effective thermal conductivity of the nanostructured materials. Recently, the long mean free path has been measured by TDTR (Siemens et al., 2010, Hu et al., 2015).

The measured thermal conductivities of nanowires are summarized in Table 1. The MEMS based measurement method is used mostly, but the experimental results of micro-Raman spectroscopy are also included. The measured values are basically much lower than that of the bulk, and the extremely low thermal conductivity cannot be explained by conventional simple mean free path of phonons calculated by Eq. (1). The order of magnitude of the thermal conductivity of nanostructured materials can be roughly evaluated by the spectral model of Eq. (2) although the spectral model is not enough to yield a deep understanding of the physics of heat conduction of periodic nanostructures such as superlattices. For example, the thermal conductivity of a nanowire with rough surfaces is lower than that of a nanowire with smooth surfaces. The understanding of the reflection of the phonons at the boundary is also very important (Blanc et al., 2013). The details of phonon transport at the interface have been investigated by numerical simulations such as molecular dynamics, however the thermal resistance have also been measured directly under the electron microscope (Liu et al., 2014, Kawamoto et al., 2015). Temperature rise was measured across the crystal grain boundary using a Focused Ion Beam or Focused Electron Beam as a tiny heater. Detailed phonon transport mechanisms have been intensively investigated experimentally.

![Fig.3 Schematic of the MEMS based suspended heater-thermometer method for thermal conductivity measurements of nanomaterials.](image1)

![Fig.4 Schematic of the 3 omega method. The metal strip wire is deposited on the thin film as a heater and a thermometer. Cross-plane thermal conductivity of the thin film is measured as a thermal resistance.](image2)

![Fig.5 Thermal conductivity of a porous Si thin film. Limiting dimension is evaluated by the distance between pores. Cumulative thermal conductivity of Si calculated by spectral phonon model is plotted as a function of the cutoff mean free path of phonon for comparison.](image3)
3.3. Coherent heat transfer

As mentioned above, a phonon in condensed matter is a quantized mechanical vibration and has the wave nature. Therefore, phonon transport can be controlled by phononic crystals (PnCs) via interference effect and its transport properties are described by the phononic band diagram. In this subsection, we focus on the thermal conduction in PnCs as a good example of coherent heat transfer. Researchers in photonics have demonstrated various kind of physics, such as lasing (Painter et al., 1999, Nomura, et al., 2010), superlens (Pendry, 2000), and slow light (Baba, 2008), using PhCs for the past three decades since the introduction of the concept (Yablonovitch, 1987). There is some analogy between the manipulation of photons and phonons transport in a PnC and PhC. However, a phononic system is more complex as the governing equation of the linear elasticity theory contains tensors and has much stronger nonlinearity. Heat transfer control is even more challenging, because heat is the transport phenomenon of phonon ensembles and phonons distribute in a broad frequency range following Bose-Einstein statistics. Therefore, control of heat transport using the wave nature of phonons still remains as a challenging topic in physics.

Several groups have been challenging this topic using phononic structures and reported reduction of thermal conductivity compared with unpatterned structures. The technological difficulty arises from the relatively short wavelength of thermal phonons (~1 nm at room temperature). Therefore, most of the experiments were performed at very low temperatures, where the thermal phonon wavelength becomes much longer and the phonon mean free path is much longer due to less incoherent scattering. The interference effect in PnC structures occurs only in a system where the phonon mean free path, more precisely coherence length (Latour et al., 2014), is sufficiently longer than the period of the PnC (Fig. 6).

Figure 7 explains how the thermal conduction is reduced by the interference effect in a PnC. The additional artificial periodicity to the crystal by the PnC folds the phononic band in the momentum space at smaller $k$ as shown in Fig. 7(a), which is the case for the PnC having the periodicity of $2a_0$ ($a_0$: lattice constant). The group velocity and density of states are changed, and a bandgap opens. These changes, called band-folding effect, results in a dramatic change of heat transport property in a PnC compared to an unpatterned structure. We show the calculation results as an example of a PnC structure with circular holes aligned in a square lattice, which is schematically shown in Fig. 7(b). The unpatterned membrane is a 145 nm-thick Si membrane, and the structural parameters of the PnC structure are: period $a = 300$ nm and radius of the hole $r = 135$ nm. We used the classical elasticity theory to compute the two-dimensional phonon dispersion in Fig. 7(c) by finite element method under Floquet condition. The thermal conductance spectrum of the PnC structure calculated for $T = 1$ K is shown in Fig. 7(e) by a red line with that of the unpatterned membrane (gray line). The PnC structure has higher thermal conductance below 7.5 GHz, but much lower above 7.5 GHz, which corresponds to only 400 mK. Therefore, thermal conductance is much lower in PnCs by the band-folding effect at realistic temperatures. We note that this is only valid for the phonons with sufficient coherence in the system. High-frequency phonons have shorter mean free path due to high incoherent scattering rate and their transport property is described by the particle picture. A longer period leads to stronger band-folding effect, but only valid for lower frequency phonons. A PnC structure with shorter period affects more phonons, but more incoherent surface scattering. This dilemma makes the realization of thermocrystals (Maldovan, 2013), defined as a phononic crystals that manipulate heat transport, very challenging.

Fig. 6 Key phonon scattering or transport mechanisms of the incoherent and coherent phonon transport. In general, in bulk material or high temperatures, phonon transport can be described by the particle picture. The wave nature of phonons becomes important in periodic structures if the coherence of the phonons are preserved; in nanoscale systems or low temperatures.
Figure 8 shows SEM images of a variety of phononic systems, where the reduced thermal conductivities were observed. Heron et al. demonstrated the geometry induced reduction in thermal conductance in a Si serpentine structure by 20-40% compared to a simple straight nanowire below 5 K [Fig. 8(a)] (Heron, Bera, Fournier, Mingo, & Bourgeois, 2010). Zen et al. demonstrated very low phonon emitted power below 1 K using Si PnC microstructure [Fig. 8(b)]. The reduction is about one order of magnitude and agrees with the simulation based on the band diagram calculation (Zen et al., 2014). The thermal phonon wavelength is long enough below 1 K, making for an ideal playground to study the heat transfer in the coherent regime. At the other extreme of this approach, Yu et al. studied thermal conductivity at relatively high temperature (> 80 K), but in extremely small phononic mesh structures [Fig. 8(c)] (Yu, et al., 2010). The mean free paths are shorter, but the period is also very short. The thermal conductance measurements in these three reports were taken electrically, which may cause difficulty when measuring many structures at low temperature. Nomura et al. developed an optical measurement technique for thermal conductivity in nanostructures (Nomura et al., 2015) [Fig. 8(d)] and enabled systematic studies by measuring hundreds of structures on a single chip. Jeremie et al. reported the direct proof of the thermal conductivity tuning based on the wave nature of phonons by measuring hundreds of systematically controlled disordered PnC nanostructures at 4 K (Maire, et al. 2015). They also observed that the thermal conductance change by the periodicity disappeared above 10 K in the PnC nanostructure with $a = 300$ nm. These experiments were performed at low temperatures to realize stronger interference effect by suppressing incoherent phonon scattering. Moreover, some simulations reported that the wave nature is not observable at room temperature even in sub-micrometer systems (Dechaumphai and Chen, 2012, Jain, et al., 2013). On the one hand, large reduction of thermal conductivity in Si PnC structures were reported and they concluded that it stemmed from the interference effect.
in PnC structures at room temperature (Hopkins, et al., 2011, Alaie, et al., 2015). Further study will enable to use both particle and wave nature of phonons for more effective control of heat transfer at higher temperatures.

As the wave optics provides much richer physics than the geometric optics, thermal phononics, defined as phonics in thermal regime, will provide novel physics. We introduce phononic boost effect as an example (Anufriev and Nomura, 2015). A properly designed PnC structure has a higher thermal conductivity than an unpatterned membrane under a certain condition [Fig. 9(a)]. The trick of this counter intuitive phenomenon stems from the band engineering, which changes the density of states as shown in Fig. 7(e); larger heat flux in the PnC structure below 7.5 GHz. Figure M4(b) summarizes the relative thermal conductance enhancement in triangular-lattice PnC structures (\(a = 80 \text{ nm, } h = 80 \text{ nm, and } r/a = 0.4\)) as compared to the membrane \(G_R = G_{PnC} - G_{Mem}/G_{Mem}\), plotted as a function of both the temperature and the period. The map shows that the greatest enhancement of thermal conductance (red color) is observed in the area where both the temperature and the period are relatively low.

The use of coherent heat transport is just getting started. Further advance in nanotechnology may enable us to play with thermal phononics above room temperature and even realize application to thermal management in nanodevices and thermoelectric materials. The particle-wave nature hybrid thermal conduction control may become an important technology in this century.

4. Conclusion

In contrast to photonics, plasmonics or acoustics where a single mode can be excited and analyzed, the main difficulty in phonon heat conduction consists in the treatment of a broad band Planck’s spectrum reaching tens of TeraHertz where modes are thermally excited in all directions in the form of decorrelated wave packets strongly interacting together through complex selection rules. This challenging situation makes of heat conduction a very rich science involving solid-state, statistical, quantum and transport physics.

This article has proposed an advanced though not complete overview of recent theoretical, numerical and experimental methods to predict and quantify thermal conduction in nanostructures or nanostructured media, exemplifying various types of scattering effects that arise at nanoscales and of confinement or coherent behaviours. Those latter have only been shown very recently and constitute a wealthy future field of research to be investigated. Nevertheless, a large number of open questions remain to be addressed in models describing phonon scattering, be it the validity of Boltzmann formula, the convergence of the thermal conductivity in low-dimensional structures, the measurement of the spectral mean free path, the relevance of interatomic potentials in complex atomic systems, the role of non-nearest atomic neighbours or the treatment of amorphous systems, to say a few.

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