

Engineering thermophysical properties of micro- and nanostructures [☆]

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Abstract—Engineered micro- and nanostructures may possess superb properties unattainable in nature. Examples are the photonic crystals for electromagnetic waves and quantum wells for electronic waves. In this article, we focus on engineering thermophysical properties of micro- and nanostructures. A brief historical perspective is provided on the motivations to engineer the properties of matters, followed by a discussion of some necessary conditions for the engineering of properties. Examples are given on the energy spectrum and thermal conductivity of low-dimensional structures such as superlattices. Future opportunities in thermally engineered structures are also elaborated. © 2001 Éditions scientifiques et médicales Elsevier SAS

thermophysical properties / nanoscale / energy spectrum / wave regime / thermal fluctuation / phonon

Nomenclature

C	specific heat	$\text{J}\cdot\text{m}^{-3}\cdot\text{K}^{-1}$
D	density of states	$\text{m}^{-3}\cdot\text{Hz}^{-1}$
d	quantum well width	m
E	energy level	J
f	distribution function	
K	thermal conductivity	$\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$
k	wavevector	m^{-1}
k_B	Boltzmann constant	$\text{J}\cdot\text{K}^{-1}$
m	electron mass	kg
T	temperature	K
U	internal energy	$\text{J}\cdot\text{m}^{-3}$
v	group velocity	$\text{m}\cdot\text{s}^{-1}$
\hbar	Planck constant divided by 2π	$\text{J}\cdot\text{s}^{-1}$
τ	relaxation time	s
τ_{12}	transmissivity between 1 and 2 angular frequency	Hz

x	x -direction
ω	spectral quantity

1. A BRIEF HISTORICAL PERSPECTIVE OF ENGINEERED MATERIALS

Mankind has been constantly improving existing and searching for new materials with superior properties. There were two distinct directions in the pursuit of better materials. One is through the use of composites, for example, to reduce weight while increase the mechanical strength. Another is the synthesis of novel materials with new physical properties by mixing elements under different conditions. This latter route has led to promising materials as represented by the discovery of high-temperature superconductors. In between the two methodologies is the control of microstructures, as in the heat treatment of steels. The degree of control in this later approach is often limited.

Yet another field, the optical thin-film coatings, which is not traditionally viewed from the materials perspective, has been developed and perfected over the last century [1]. Thin-film effects on light propagation were reported as early as in Newton's times, long before the

Subscript

1	point 1
2	point 2
n	index for energy level

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modern theory of light was established. With the advancement of thin-film deposition technologies, one can design various multilayer structures to achieve desired reflection and transmission in selected wavelength range, using successive reflection at each interface and superposition of electromagnetic waves. For example, using a periodic structure of thin films, one can design Bragg reflectors with high reflectivity in the desired wavelength range (stop band) and zero reflectivity in other wavelength range (pass band). Over most of the last century, however, the control of optical waves stayed at the level of coatings, although one might argue that diffractive gratings go beyond the concept of interference effects dominant in coatings. All of them can be considered as one-dimensional (or quasi one-dimensional) structures.

In contrast, naturally existing crystals are three-dimensional periodic structures. Electrons and phonons inside the crystals experience the periodic potential variation in all directions. The potential surrounding each atom can be thought as a reflector for the electron or phonon waves. The diffraction of the electron and lattice waves leads to familiar energy band structures for electrons and phonons. The electrons, for example, experience a periodic potential variation in every crystallographic direction, which creates stop bands similar to that existing for photons in a Bragg reflector in every direction. If a common stop band exist among all the directions, an absolute bandgap is formed. The existence of bands and bandgaps distinguishes familiar material classes such as metals, semiconductors or insulators.

This comparison between optical coatings, particularly periodic Bragg reflectors in optical coatings, with naturally existing crystals is very intriguing. First, it shows that the periodicity in one direction (as in Bragg reflectors) and three-dimension (as in crystals) leads to similar attributes (such as stop bands). Second, absolute bandgaps only exist in three-dimensional crystals. Third, one can design thin films to obtain the desired optical properties of the structures. The analogy between photons and material waves such as electrons and phonons led to several profound ideas developed over the last three decades of the 20th century.

The first development is to make multilayer structures for electron waves similar to optical coatings for photons. In a landmark work, Esaki and Tsu proposed the concept of superlattices, which are made of periodic alternating thin films [2]. The period thickness is designed for electron waves and is thus very short compared to optical structures, but the concept shares similarities with Bragg reflectors for optical waves. The electron waves

propagating in such a periodic structure resemble in many ways the propagation of photons in a Bragg reflector. Rather than using the optical terminology of reflection and transmission, the electronic properties of superlattices are described using the solid-state terminology such as band structures. In a superlattice, the electrons acquire new characteristics nonexistent in their constituent bulk materials, such as the mini bandgaps, which are equivalent to the stop bands for optical waves. Novel transport properties can be obtained from the new band structures, such as negative electrical conductivity, etc. The analogy of phonon propagation in superlattices to optical waves in Bragg reflectors was explored by Narayanamurti and coworkers [3]. This again was quickly converted into the solid-state terminology [4]. Since the superlattice concept was proposed, many other quantum structures that explore size and interface effects on material waves have been explored and used to make better devices. Examples are quantum wells (very thin films), quantum wires, and quantum dots, as well as their periodic structures such as quantum-wire superlattices and quantum-dot superlattices.

While the engineering of electrons has followed a path similar to the control of optical waves in thin films, i.e., from three-dimensional periodic potentials in natural crystals to one-dimensional artificial potentials in superlattices, the engineering of photons gained a significant boost from making analogy to three-dimensionality in natural crystals [5]. In this case, the idea is to examine the propagation of optical waves in all directions in a three-dimensional periodic structure called a photonic crystal. If a common intersect of stop bands exists in all directions, one get an absolute bandgap for photons, in which no radiation can exist inside the structure at all. Such an analogy to naturally existing crystals leads to many new exciting developments. Yet it is not the end of the story. More recently, strange characteristics of light propagation in artificial structures have been intensely discussed, such as the negative refractive index materials in which light refraction has unconventional behavior [6].

This very brief history serves to illustrate the tremendous potential of exploring the structural effects on the physical properties as manifested by electrons, phonons, and photons. Will such exciting progresses have any impacts on thermal science and technology? I believe the answer is yes. In this article, I will explore some opportunities to engineer the thermophysical properties using micro- and nanostructures.

2. NECESSARY CONDITIONS FOR ENGINEERING

What are the necessary conditions required to fundamentally alter the characteristics of a material through structural effects? We should compare the characteristic length of energy carriers (electrons, phonons, photons, and molecules) with the characteristic lengths of the structures [7, 8]. Major characteristic lengths of carriers are their wavelength and their mean free path. A structure can also have characteristic lengths at different scales. Taking a crystal as an example, the characteristic lengths involved are the macroscopic size of the crystal, the size of the unit cell, and the basis (atoms or molecules) attached to each lattice point. The properties of a crystal depend on these characteristic lengths, as well as the characteristics of the atoms or molecules attached to each lattice point.

2.1. Engineering in the wave regime

The brief historic perspective provided above is based primarily on the wave characteristics of matters. This is because the superposition of waves leads to many variations and thus provides the best ground for engineering properties. When the wavelength is comparable or larger than the characteristic lengths of a structure, such as the thickness of a thin film, the waves are constrained by the interfaces and boundaries. The new boundary conditions reshape the waves, thus changing their frequency and energy states. To utilize the wave effects, the following conditions are necessary:

(1) *Long mean free path to preserve the phase information carried by the waves.* This imposes a limit on the maximum length scale of the unit cell or the crystal size. The mean free paths of electrons and phonons are typically very short, of the order of 10–1000 Å at room temperature. Photons, in comparison, can have a long mean free path. The characteristic lengths of the engineered structure such as the unit cell size or the overall crystal size should have comparable or smaller dimension than the mean free path.

(2) *Hetero-interfaces to create destructive or constructive superposition of waves.* This is normally realized in composite structures, in which one component can be vacuum or air. Examples are superlattices and photonic crystals.

In addition to these two conditions, the following two factors should be considered.

(3) *Wavelength vs. the unit cell length.* If the wavelength is comparable to the unit cell length, strong diffraction effects of waves can be observed. Such diffraction effects may lead to the formation of photonic bandgap in a photonic crystal, and the electron and phonon minigaps in superlattice structures. If the wavelength is much longer than the unit cell length, the wave samples the average properties of the unit cell. Normally an effective medium theory would suffice to describe the properties of the structure and one may be inclined to think this regime is not of much interest. There are, however, some very exciting developments in this area recently. The major idea is to make artificial “atoms”. This is particularly easy for long wavelength electromagnetic waves. For example, through the use of coil structures at each lattice site, one can create artificial materials with magnetic properties not attainable from nature [9].

(4) *Thermal fluctuation of background.* The artificial effects created in engineered structures should be strong enough to overcome the background thermal fluctuation. For example, the quantized electron energy levels in a quantum well of width d (which can be made of a thin film of thickness d) are, approximately

$$E_n = \frac{\hbar^2}{2m} \left(\frac{\pi n}{d} \right)^2 \quad (n = 1, 2, \dots) \quad (1)$$

where \hbar is the Planck constant divided by 2π and m is the electron mass. The separation of energy between two adjacent levels is of the order of

$$\Delta E_n = E_{n+1} - E_n = \frac{\hbar^2}{2m} \left(\frac{\pi}{d} \right)^2 (2n + 1) \quad (2)$$

For this energy separation to be observable, it should be larger than the thermal fluctuation of the background, which is of the order of $k_B T$. Thus, the condition for observing the energy separation is

$$\frac{\hbar^2}{2m} \left(\frac{\pi}{d} \right)^2 (2n + 1) \geq k_B T \quad \text{or} \quad d \leq \hbar \pi \sqrt{\frac{3}{2mk_B T}} \quad (3)$$

for $n = 1$. At room temperature and with an electron mass of 9.1×10^{-31} kg, the above relation leads to $d \leq 100$ Å.

2.2. Engineering in the particle regime

When the coherence properties of the materials wave are lost, either due to the scattering or the smearing of the thermal background, the energy carriers can be treated

as particles. In this regime, it is still possible to utilize the hetero-interfaces to change the directions of energy carriers and thus to control the transport properties. The necessary conditions for engineering the properties are (1) long mean free path such that the interfacial transport is dominant, and (2) existence of surfaces and interfaces to control the directions of the energy carriers.

Because the mean free path and the wavelength of electrons and phonons are normally in the order of tens to thousands angstroms, it becomes clear that engineering material properties should occur in micro and nanostructures. Electromagnetic waves, because of their wide spectrum range and long mean free path, can be engineered with relatively large structures. From examining electromagnetic waves behavior in engineered materials, one can draw analogy to possible electron and phonon waves at a much smaller scale, as well as directly benefiting from the altered radiative properties of the structures.

3. ENGINEERING THERMOPHYSICAL PROPERTIES

Because of the coherent superposition of waves in periodic structures, the energy states of electrons, phonons, and photons can be significantly changed. In *figure 1*, we compare the calculated phonon spectra in an artificial superlattice structure made of two monolayers of silicon and two monolayers of germanium with those of their parent bulk materials [10]. Only acoustic phonons in bulk materials are considered. The mismatch of phonons in bulk materials creates optical-like phonons in super-

lattices. The destructive superposition of phonons forms minigaps that do not exist in bulk materials. The energy spectrum variations also bring about corresponding changes in the density of states. *Figure 2* illustrates the density of states of electrons in low-dimensional structures and phonons in a superlattice. Due to these changes of energy states, we can envision that the thermophysical properties can be engineered towards the desired directions. In this section, we will examine the internal energy and thermal conductivity.

3.1. Internal energy and specific heat

The spectral energy density of a group of heat carriers can be expressed as

$$U(T, \omega) = \hbar \omega f(\omega, T) D(\omega) \quad (4)$$

where ω is the angular frequency of the carrier and $\hbar \omega$ its energy, f the statistical distribution of the carrier, and $D(\omega)$ the density of states. The total energy of the carrier can be obtained from integrating over all the frequencies. From this energy density, one can obtain, for example, the Planck blackbody radiation distribution. In engineered materials, based on our previous discussion, both the allowable energy levels and the density of states can be changed. This could, for example, change the thermal emission characteristics of a photonic crystal. In the bandgap region, thermal emission can be totally suppressed. In other wavelength region, it is possible to obtain a density of states much higher than obtainable in nonstructured materials [11].

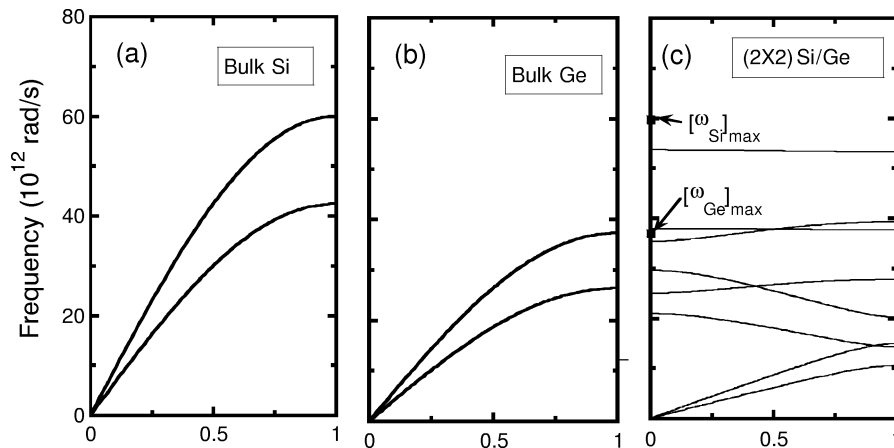


Figure 1. Comparison of acoustic phonon spectra in bulk Si, Ge and a superlattice made of two monolayers of Si and two monolayers of Ge [10].

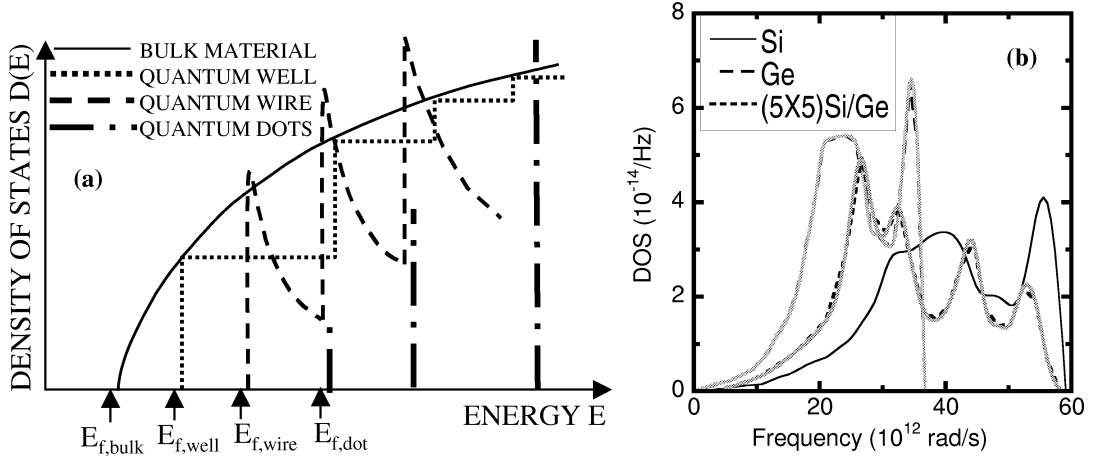


Figure 2. (a) Schematic density-of-states of electrons in bulk material, quantum wells, quantum wires, and quantum dots, and (b) density-of-states of phonons in a 5 monolayer by 5 monolayer Si/Ge superlattice [10].

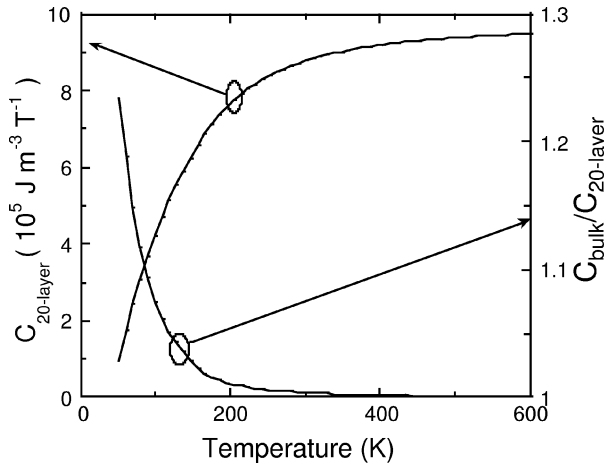


Figure 3. Specific heat of a 20 manolayer of Si quantum well [16].

The specific heat of a solid can be obtained from integrating the spectral energy density and its temperature derivative,

$$C_t = \frac{\partial U_t}{\partial t} = \int \hbar\omega \frac{\partial f}{\partial T} D(\omega) d\omega = \int C(\omega) d\omega \quad (5)$$

For phonons, the effect of integration is generally to smear out the difference in spectral properties, leaving only a small change in the specific heat compared to bulk materials [12–14]. *Figure 3* shows an example of the computed specific heat of a Si quantum well [14]. The differences become larger at low temperatures where long wavelength phonons become more important. For

extremely thin layers such as carbon nanotubes, the observed specific heat is higher at low-temperatures from their parent material graphite [15].

Compared to phonons, the specific heat of electrons can have stronger size dependence due to the following factors: (1) The energy quantization of electrons is more dramatic than phonons because of the small electron mass and thus longer wavelength. (2) The specific heat also depends on the Fermi level, particularly the rate of change of the density of states at the Fermi level, which can be controlled through the size of nanostructures. Indeed, existing studies show that the specific heat is a strong function of the size [16].

3.2. Thermal conductivity and heat transfer

Compared to thermodynamic properties, the transport properties can be altered more significantly by nanostructures. We can examine the possibility of engineering the energy transport from two different formalisms. One is the kinetic formulation and the other is the Landauer formalism. In a kinetic formulation, the heat flux in x -direction can be expressed as

$$q_x = \int v_x \cdot E \cdot f d^3\mathbf{k} \quad (6)$$

where v_x is the carrier group velocity along x -direction and f is the local carrier distribution function. The integration is carried out over the wavevector space.

For bulk materials, this leads to the following kinetic expression of its thermal conductivity,

$$K = \frac{1}{3} \int v^2 \cdot C(E) \cdot \tau(E) dE \quad (7)$$

where τ is the relaxation time of carriers with energy E . Compared to specific heat $C(E)$, group velocity and relaxation time are two additional quantities that determine the transport. In low-dimensional structures, these quantities can be altered. In bulk materials, the group velocity does not change much for a given material. Scattering is thus left as the major approach to control the thermal conductivity. In applications where low-thermal conductivity solids are desired, for example, the thermoelectric energy conversion, methods such as alloying and institutional addition of atoms into open cages (the phonon rattlers) have been developed to reduce the phonon mean free path [17, 18]. In low-dimensional structures, the group velocity can be significantly reduced, in addition to established methods to increase the scattering. In *figure 4* we show an example of the averaged phonon group velocity in Si/Ge superlattices along the film plane (in-plane) and perpendicular (cross-plane) directions. Such group velocity change may cause a variation in the scattering rate and thus further affects the thermal conductivity.

The Landauer formalism [19] can also be used to examine what are potential approaches in engineering the transport properties. The Landauer formalism treats transport as a transmission process. The heat flux between any two points 1 and 2 can be expressed as

$$q_{12} = \int v_x \cdot E \cdot (f_1 - f_2) \cdot \tau_{12} d^3\mathbf{k} \quad (8)$$

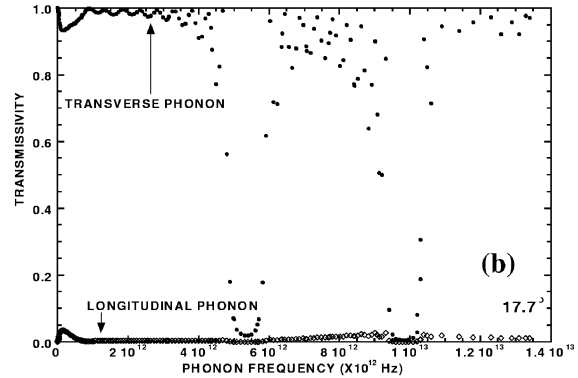
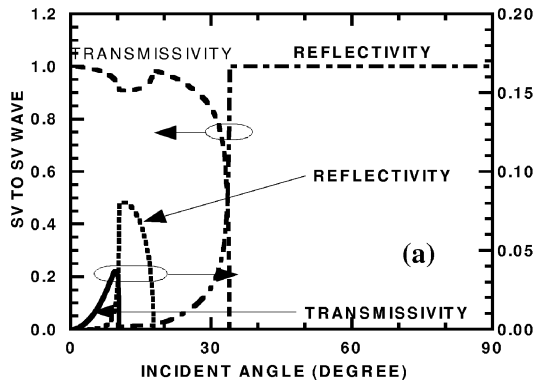


Figure 5. (a) Phonon reflectivity and transmissivity at one interface similar to Si/Ge and (b) phonon transmissivity through a $5 \text{ \AA} \times 5 \text{ \AA}$ Si/Ge superlattice [20].

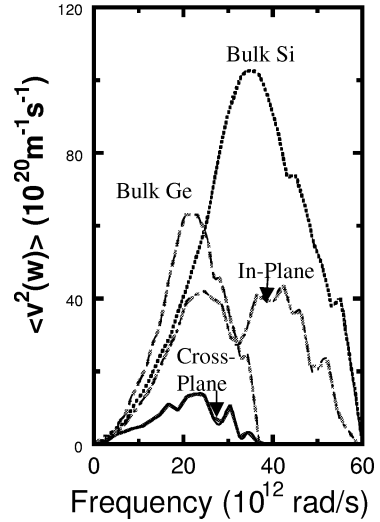


Figure 4. Square of group velocity averaged over wavevectors [10].

where τ_{12} is the transmissivity of the carrier between points 1 and 2. In nanostructures, we can use interfaces to control the transmissivity. *Figure 5* shows the simulated phonon transmissivity at an interface similar to that of silicon and germanium and through a Si/Ge multilayer structure [20].

The wave effects on phonon thermal transport are strongest in extremely small nanostructures where the superposition of thermally excited phonons still demonstrate strong phase coherence. Because of the broadband nature of thermal phonons, their coherence lengths are typically very short [21]. For thermal transport, we can often neglect the wave nature and treat them as particles. In this later regime, however, we can still use in-

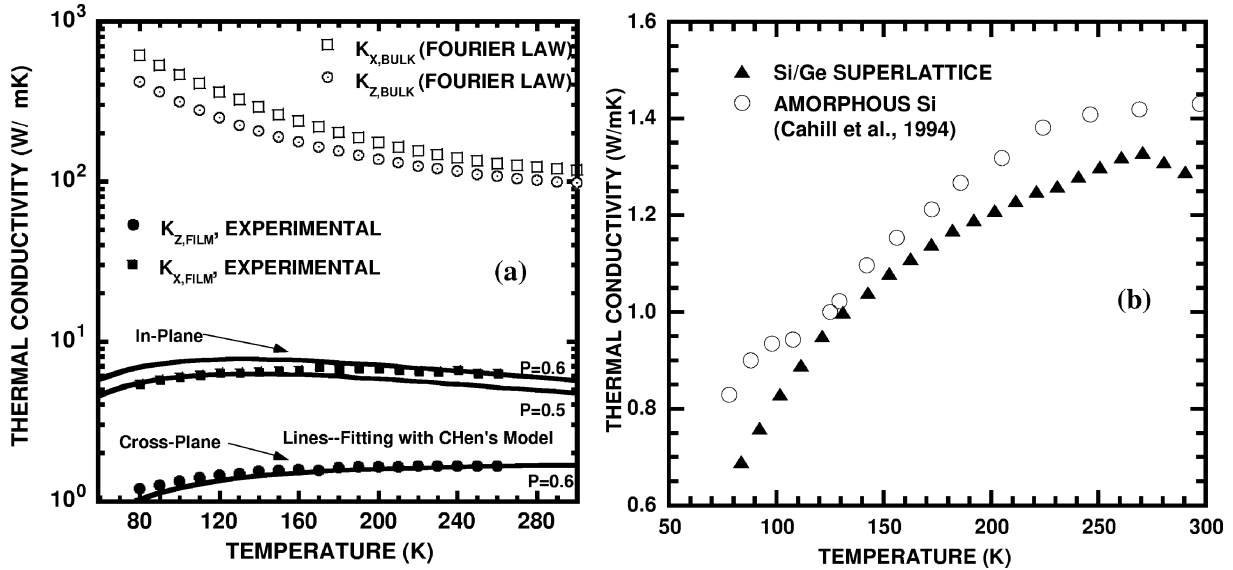


Figure 6. (a) Anisotropic thermal conductivity of a 20 Å/20 Å Si/Ge superlattice and comparison with results from the Fourier law based on the bulk thermal conductivity of Si and Ge [23], and (b) cross-plane thermal conductivity of a Si/Ge superlattice [25] and comparison with the thermal conductivity of amorphous Si.

interfaces to reduce the lattice thermal conductivity [22]. *Figure 6* shows the thermal conductivity of Si/Ge superlattices along and perpendicular to the film plane of a Si/Ge superlattice [23]. Although these structures are crystalline, the thermal conductivity is significantly reduced compared to predictions of the Fourier law based on bulk properties of silicon and germanium. Theoretical models suggest that these reductions are due to the interface reflection of phonons [21, 24]. In *figure 6(b)*, we show another example demonstrating that the thermal conductivity of such superlattices can be made comparable to amorphous materials, while maintaining their crystalline structure [25].

4. OPPORTUNITIES WITH THERMALLY ENGINEERED STRUCTURES

The new degree of freedom to control the thermophysical properties through micro- and nanostructured materials can have many potential applications. Here we will discuss a few potential areas.

Solid-state cooling and power generation based on thermoelectric and thermionic effects. In these technology areas, thermal isolation between hot and cold side is a key factor that determines the energy conver-

sion efficiency. Taking thermoelectrics as an example, the efficiency of an ideal device is determined by the thermoelectric figure of merit defined as $Z = S^2\sigma/k$, where S is the Seebeck coefficient, σ the electrical conductivity, and k the thermal conductivity. Thus a good thermoelectric material must be a good electrical conductor while being a good thermal insulator, together with a higher energy conversion factor represented by the Seebeck coefficient. Such materials have proven to be difficult to find. Modeling and experiments in recent years show that the $S^2\sigma$ can be increased using quantum size effects [26]. Such a possibility, together with the low thermal conductivity, has led to a surge of thermoelectric research in low-dimensional structures [27].

Radiatively controlled surfaces and structures. Surface emissivity control is needed in many technology areas such as IR detection, photovoltaics and thermophotovoltaics. Photonic crystals and photonic band gap effects provide new possibilities to engineering surfaces as well as three dimensional structures with desired spectral emissivity and transmissivity in three-dimensions, unlike thin-film based techniques which have stronger directional dependence. Such structures may be used to improve the energy conversion efficiency of photovoltaic and thermophotovoltaic energy conversion cells. One may also think about combining photonic structures with

combustion technology, since the concept of porous combustors has been studied before [28].

Concurrent photonic and phononic engineered structures. In thermal insulation materials, radiation can contribute significantly to the nominal thermal conductivity of the materials. By concurrently engineer the lattice thermal conductivity through phonon engineering and radiation transfer through photonic structures, one may reach much better thermal insulation materials. Such an approach may also be applied to coatings used for high temperature engines.

Thermal management of electronics and photonic devices. The poor thermal conductivity of multilayer structures used in many photonic and microelectronic devices, particularly semiconductor lasers, has significant impacts on the device performance. This is especially so when high output power from the lasers is desired. This area calls for concurrent thermal, optical, and electrical design of devices for optimum performance. One may also think about ways to increase the thermal conductivity. There is indication, for example, that the thermal conductivity of carbon nanotubes is very high [29]. This may be due to the change of scattering rules in two-dimensional structures.

Of course, the few examples we give here are by no means inclusive and only serve to illustrate the iceberg of the potentials with engineered materials.

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