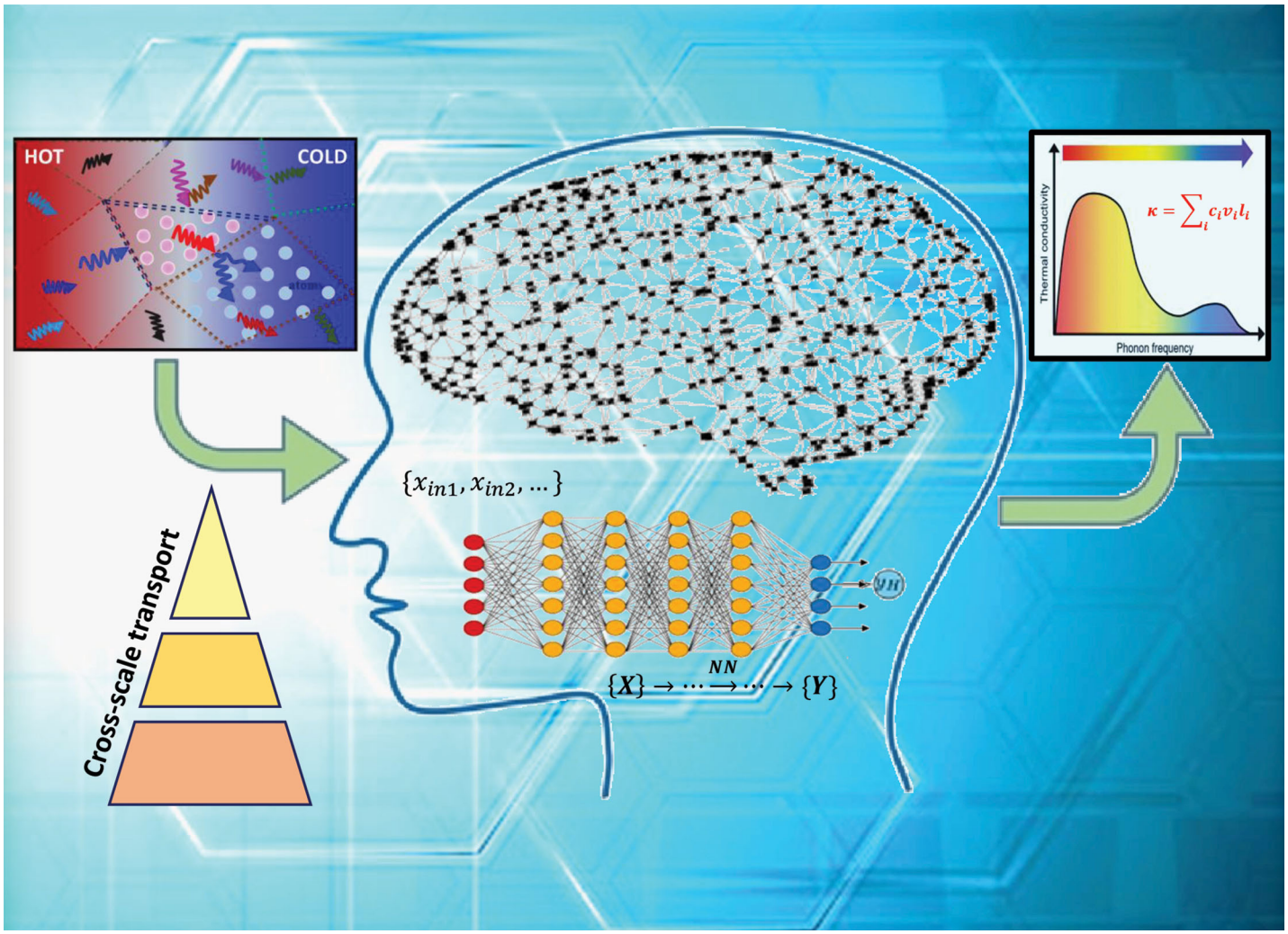

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Perspective on multi-scale simulation of thermal transport in solids and interfaces

Phonon-mediated thermal transport is inherently multi-scale. The nature of multi-scale thermal transport resides in that there are different heat transfer physics across different length scales with strong entanglement and interaction with each other. This perspective highlights the great potential of quantitative deep learning for multi-scale modelling of thermal transport in solids and interfaces, by leveraging its strong feature learning and ability to unlock the strongly nonlinear relationship between complex microscopic structures and macroscopic cross-scale transport behaviours.

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Perspective on multi-scale simulation of thermal transport in solids and interfaces

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Phonon-mediated thermal transport is inherently multi-scale. The wave-length of phonons (considering phonons as waves) is typically at the nanometer scale; the typical size of a phonon wave energy packet is tens of nanometers, while the phonon mean free path (MFP) can be as long as microns. At different length scales, the phonons will interact with structures of different feature sizes, which can be as small as 0D defects (point defects), short to medium range linear defects (dislocations), medium to large range 2D planar defects (stacking faults and twin boundaries), and large scale 3D defects (voids, inclusions, and various microstructures). The nature of multi-scale thermal transport is that there are different heat transfer physics across different length scales and in the meantime the physics crossing the different scales is interdependent and coupled. Since phonon behavior is usually mode dependent, thermal transport in materials with a combined micro-/nano-structure complexity becomes complicated, making modeling this kind of transport process very challenging. In this perspective, we first summarize the advantages and disadvantages of computational methods for mono-scale heat transfer and the state-of-the-art multi-scale thermal transport modeling. We then discuss a few important aspects of the future development of multi-scale modeling, in particular with the aid of modern machine learning and uncertainty quantification techniques. As more sophisticated theoretical and computational methods continue to advance thermal transport predictions, novel heat transfer physics and thermally functional materials will be discovered for the pertaining energy systems and technologies.

1. Introduction

In all fields of modern science and technology, phenomena across space and time scales are the most fascinating issues. The micro-scale components of materials and structures are atoms and molecules, and their interaction in principle determines all macro-scale behavior of the materials, which is the most interesting scale for practical applications. Therefore, understanding the characteristics and principles of materials at different spatial and time scales has a great appeal for technological innovation. In the material modeling and simulation, this can be roughly divided into the following four feature space scales (Fig. 1):

(1) Nanoscale (10^{-9} m): the electrons are the dominant players, and quantum mechanics determines the interaction between them.

(2) Microscale (10^{-8}–10^{-6} m): atoms play a major role, and their interactions can be described using classical atomic potentials, with or without fixed formula, including the effects of chemical bonds between them. Some thermodynamic quantities such as temperature cannot be well defined in some extreme cases.

(3) Mesoscopic scale (10^{-5}–10^{-4} m): microstructures play an important role in determining the material properties, such as grain boundaries, voids, and other microstructure elements. Empirical models are usually effective in handling the phenomena in this scale.

(4) Macro-scale ($>10^{-3}$ m): materials are regarded as continuous medium and constitutive laws govern the behavior of the physical systems. At the macro-scale, continuous fields, such as density, velocity, temperature, displacement, and stress fields, can be well defined. Constitutive law can be also used to analyze some effects from defects.

In contrast to spatial scales, the time scale spans from femtoseconds (10^{-15} s) to seconds, ~20 orders of magnitude change which is much larger than that for length scale. For instance, during femtosecond laser fabrication,[1] photons are mainly absorbed by electrons, which is governed by femtosecond photon–electron interactions, and the subsequent energy transfer from electrons to ions (usually phonons) is of femtosecond to picosecond order. Therefore, the femtosecond laser fabrication process can be improved by controlling localized transient electron dynamics, which poses a challenge for measuring and controlling at the electron level during the fabrication processes. Another example

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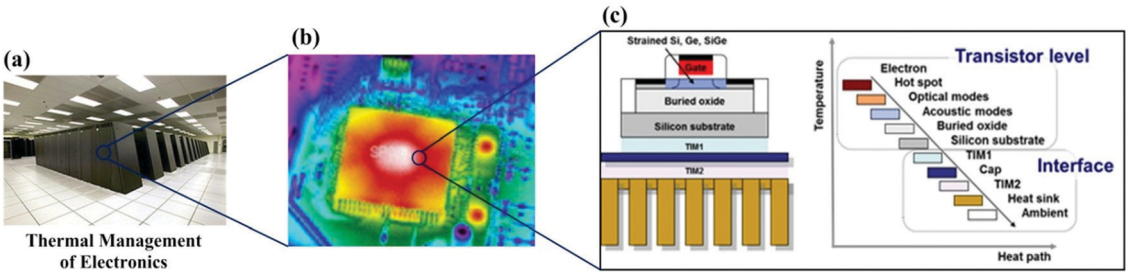

Fig. 1 Schematic of the dominant role of interfacial heat transfer in thermal management of high power electronics. (a) Overview of Blue Gene supercomputer stacks. (b) Temperature contour indicating the microprocessor as the hottest part of electronics. (c) Heat path from the source in the drain region of individual transistors to the heat sink shows that the overall thermal resistance is dominated by a series of interfaces at both the transistor level and external package level.

is the additive manufacturing with short pulse width and high energy of a femtosecond laser,[2] where current limitations in understanding the effects of the laser-metal or laser-alloy interactions, particularly the dynamics associated with highly non-equilibrium melt pool environments, present a significant barrier to the rapid optimization, simulation, and rational design of the process conditions. In short, effectively treating cross-time-scale problems is much more challenging than dealing with cross-length-scales.

In recent years, with the intersection of materials science–physics–computational modeling and simulation, several important and dominant methods have been developed including first-principles calculations (nanoscale), molecular dynamics (microscale), and Monte-Carlo and phase-field (mesoscale), and continuum mechanics (macroscale). Overall, these methods work very well in their respective length- and time-scales (herewith we call them ''mono-scale methods''), while expanding them to the neighboring length- and time-scales usually requires careful treatment and most of the time the solution or methodology is problem based and/or system specific, *i.e.*, a general or universal framework was rarely reported.

From the thermal transport point of view, the microelectronics field is the earliest engineering field where the macro-, micro- and nano-scale heat transfer solutions need to merge. Developments in the hard surface and interface science have given us ever faster computers and communication technologies. With the advancement in nanotechnologies, nowadays, electronic devices are getting smaller and smaller, while their power requirements are increasing. The size scaling of transistors and increase of clock rates, according to Moore's law, led to an explosion in the power-density for logic circuits, communication devices, and memories. Although the energy per operation is still decreasing, cramming more and more transistors in the same area increases the density of dissipated power to an unacceptable level that threatens the current fast rate of progress in the industry. Therefore, a proper design of thermal management within the devices is becoming increasingly critical to sustain performance, reliability, and disruptive development of electronics. In most modern electronic systems (computers, radios, and radar modules, *etc.*), the electronic device is the warmest element in the system, and waste heat is removed by conduction, spreading, and convection to an appropriate working fluid (*e.g.* air, water, or a refrigerant) with gradual reductions in the temperature as heat travels from the source to the fluid. Considering the high-speed Blue Gene supercomputer as an example [Fig. 1(a)], along the heat path from the source in the drain region of individual transistors to the heat sink, whether in air or in liquid cooler, the heat flux crosses a multitude of interfaces [Fig. 1(c)]. To date, thermal interfaces are responsible for around 1/3 to 1/2 of the total thermal resistance in power single inline packages or microprocessor systems. Multiscale strategies are therefore very important to ensure efficient heat removal, such as package-scale thermoelectric coolers, thermal interface materials, and transistor level approaches, all of which need to work synergistically to accomplish the mission. These approaches all include thermal (mainly phonons) transport issues at multi-interfaces that still need to be addressed.

Another example is thermoelectric energy conversion. Thermoelectrics offer an attractive pathway for addressing an important niche in the globally growing landscape of energy demand, since they can convert waste heat into electricity, the highest form of energy in terms of thermodynamic quality. In general, development schemes to improve thermoelectric conversion efficiency in the past few decades were guided by the concept of ''phonon glass–electron crystal'',[3,4] *i.e.*, reducing the lattice contribution to the thermal conductivity as closely as possible to an amorphous state, while keeping a relatively high electrical conductivity and Seebeck coefficient by optimizing the doping level.[5] In this framework, exhaustive scientific efforts have been dedicated to reducing the lattice thermal conductivity.[6] The ''phonon glass–electron crystal'' approach has stimulated a significant amount of new research and has led to a significant increase of ZT for several compounds, such as skutterudites,[7,8] clathrates,[9,10] and half-Heusler intermetallic compounds[11,12] (Fig. 2). Recently, due to the improving capability to synthesize nanostructured materials, nanostructuring of existing TE materials of interest has emerged as a promising pathway to greatly reduce the lattice thermal conductivity to lower values at the theoretical limit and, as a result, to improve thermoelectric performance[13] (Fig. 2). Typical examples in this route include low-dimensional nanostructures such as quantum dots, nanowires along with subsequent structure modulation,[14–16] nanocomposites,[17] superlattices,[18–20] and bulk nanostructured materials.[21] By exploiting nano-scale effects, such as strong boundary or interfacial phonon scattering, and by

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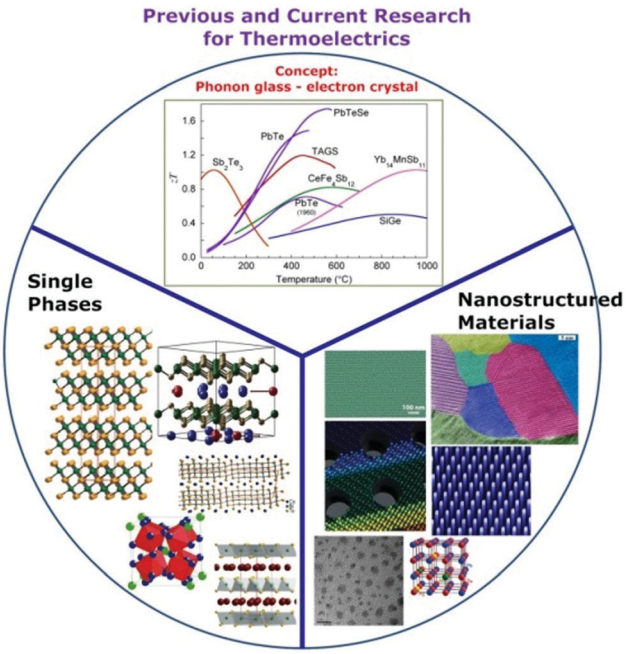

Fig. 2 Representative progress in thermoelectrics research with the main concept of "phonon glass–electron crystal".

taking advantage of the quantum confinement effect, nanostructured materials can achieve decent *ZT* values at room temperature and record-high *ZT* values of 1.5–2.0 at medium and high temperatures. However, there is little room to further improve the *ZT* coefficient of nanostructured materials. Although nanostructures in bulk thermoelectrics allow effective phonon scattering of a significant portion of the phonon spectrum, leading to unprecedentedly low lattice thermal conductivity, phonons with long mean free paths (MFPs) still remain largely unaffected. To this end, recently researchers have proposed a concept of all-scale hierarchical architectures to achieve the maximum reduction in lattice thermal conductivity by considering sources of scattering on all relevant length scales in a hierarchical fashion, from atomic-scale lattice disorder and nanoscale endotaxial precipitates to mesoscale grain boundaries.[22] The experimental tests on PbTe were very promising, which call for direct modelling of phonon transport across different length scales for further structure tailoring and system optimization. The key to achieving breakthrough *ZT* coefficient of nanomaterials for the next generation of thermoelectrics is to precisely predict the detailed collective phonon transport in the "hierarchical" materials that has different interfaces, spanning from atomic to mesoscopic and even to the macroscopic level (Fig. 3).

2. Review of major mono-scale computational methods for heat transport

As heat carriers for semiconductors and insulators, phonons in a material typically have wavelengths and mean free paths that span several orders of magnitude and are usually mode dependent. This complexity leads to the coexistence of ballistic and diffusive transport and the simultaneous action of scattering sources at multiple scales. Furthermore, phonon distributions depend on wavevector and polarization and are also inhomogeneous in space and time. The classical Fourier's law, which considers that heat is conducted diffusively with the thermal conductivity of bulk materials, is found to be only valid in the continuum scale. When the characteristic length of the structure reduces to micro-meter and even down to nano-meter, a growing number of experimental measurements have observed the reduction of the thermal conductivity compared to that of the bulk value.[23,24] The performance of micro–nano materials depends on the results of nonlinear coupling evolution of different physical processes on multiple spatial and time scales from micro and meso to macro, and the development of corresponding multi-scale heat conduction simulation methods has become a hotspot of research in the past few decades.

From a numerical modeling point of view, computational methods including anharmonic lattice dynamics (ALD) based on force constants from empirical force fields or first-principles

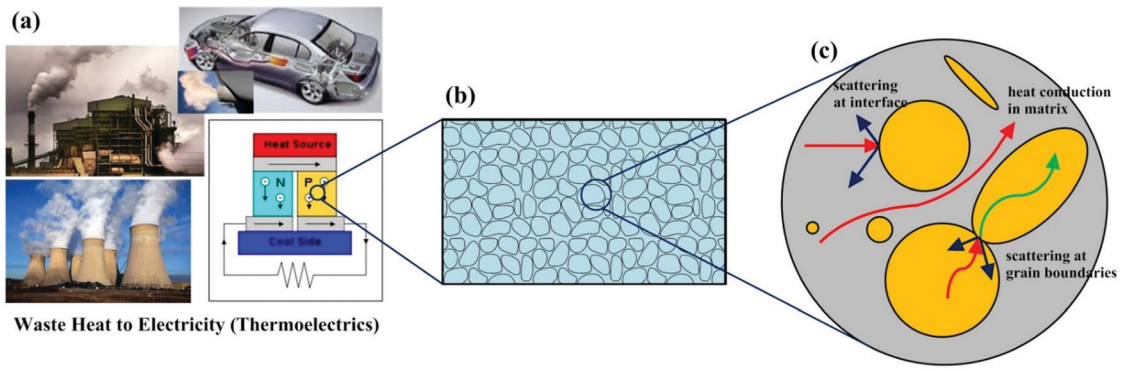

Fig. 3 Multiscale nature and critical role of interfacial heat transfer in high performance thermoelectrics. (a) Principle of thermoelectrics that converts waste heat into electricity (right bottom) and typical examples of waste heat, such as steam at the outlet of cooling towers in power plants and automobile exhaust. (b) Schematic of nanostructured materials as high performance thermoelectrics where tremendous grain boundaries hinder the propagating phonons. (c) Major mechanism of heat transfer in nanostructures, where phonon scattering at interfaces is dominant.

calculations and in combination with Boltzmann transport equation (BTE), nonequilibrium Green's function (NEGF), classical molecular dynamics (MD) simulations, and Monte Carlo (MC) simulations have been used to study nanoscale thermal transport.[25] Below is a short description of each method that is suitable and widely used for single time and spatial scale:

1. Anharmonic lattice dynamics, combined with phonon Boltzmann transport equation,[26] has been able to predict thermal conductivity with unprecedented accuracy and without the need of any empirical input, when using first-principles calculations[27,28] to evaluate force constants. The coupled ALD/BTE method has been successfully used to predict the thermal conductivity of simple perfect crystals[29–35] and some compounds.[36] However, it becomes more challenging if inhomogeneity has to be taken into account, which involves a significantly large supercell that requires unbearable computational demands. In addition, the method has difficulty in precisely characterizing the effect of free surface and boundary. Recently, lattice dynamics was also used to evaluate frequency dependent transmission coefficients at an ideal Si–Ge interface.[37] However, lattice dynamics only handles a harmonic phonon process at the interface so far.

2. Green's function: in an atomistic Green's function approach, the system is represented at the molecular level using atomistic potential models. Heat current in the system subject to a small temperature difference is related to the interatomic force constants. This heat current is expressed in terms of Green's function, and the phonon transmission as a function of phonon frequency is calculated.[38–40] This method has been widely used for studying ballistic phonon transport in nanostructures, across interfaces and molecular junctions. Recently, the Green's function method is extended to provide mode dependent transmission,[41] but it can still only handle ballistic transport so far.

3. Classical molecular dynamics simulations are based on the fully atomistic description of systems and trace the time-dependent trajectories of all atoms based on Newton's second law of motion and interatomic potentials.[42,43] Equilibrium and non-equilibrium molecular dynamics (EMD, NEMD) are the two major methods to calculate thermal conductivity with their respective advantages and disadvantages.[42] Although it is robust and "automatic" to consider the effect of surface and interface, the disadvantage of MD simulation is that it largely relies on an accurate interatomic potential. The problem of inaccurate interatomic potential can be alleviated by using *ab initio* molecular dynamics (AIMD). In recent years, due to the fast advancement of computational capability, AIMD has been largely used for quantum level simulations of a chemical process,[44,45] thermodynamics,[46,47] material physics, mechanics,[48,49] and also thermal transport,[50–53] wherein finite temperature dynamical trajectories are generated by using forces computed "on the fly" from electronic structure calculations. In addition, most of the time the MD simulations are limited by the small length scale (up to microns) and short time scale (up to tens or hundreds of nanoseconds). The limited length scale issue can be partially solved by the recent development of coarse grained molecular dynamics (CGMD) which can do meso- or even macro-level simulations. Although CGMD has been long proposed and improved for biological systems,[54–57] little progress has been made in the application and development to the thermal transport field.[58–60]

4. Monte Carlo simulations or finite volume method are often used to solve the Boltzmann equation for arbitrary structures.[61–65] However, when frequency dependent phonon mean free paths need to be considered, the computational time of the Monte Carlo simulation becomes considerably large. Moreover, for complex structures the widely used Matthiessen rule was found to be questionable especially when the structure sizes have a large variation and such a treatment can significantly overestimate thermal conductivity. A more severe problem is to feed critical input parameters such as phonon lifetime and scattering details at the interface or boundary, which cannot be straightforwardly obtained from atomistic simulations. Currently, the sophisticated method is to fit some parameters or use gray or empirical models in Monte Carlo simulations to match the experimental results or give a relatively large uncertain prediction with lower and upper bounds. Regarding length scale, recently, hybrid Monte Carlo algorithm was proposed as an excellent computational scheme that can not only significantly outperform the traditional Metropolis sampling, which was the particularly popular flavor of this technique, but also complement molecular dynamics in materials science applications, while allowing ultra-large-scale simulations of systems.[66] From this regard, the Monte Carlo method is very promising for predicting thermal transport in solids and interfaces with a length scale far beyond atomistic models such as classical MD, AIMD, and even CGMD.

5. Continuum level modeling: this type of modeling includes finite element method (FEM) and computational fluid dynamics (CFD), which are usually used for simulating conductive and convective heat transfer, respectively. The governing equations, *e.g.*, Fourier's law of heat conduction, are established on the general heat transfer principles, such as energy conservation laws and thermodynamics laws. Although mature for continuum scales, sometimes the thermal energy equations were pushed down to the limit of micro- or even nano-scale solid particles.[67] Care must be taken for the important properties in the continuum methods, *e.g.*, the effective thermal conductivity and interfacial/volumetric heat transfer coefficient at the solid–solid or solid–liquid interfaces. Moreover, the continuum model of Fourier's law of heat conduction cannot be used for instantaneous thermal response under highly out-of-equilibrium thermodynamic conditions.

A special note is addressed to the historically basic approaches to simulate the thermal conductivity of continuum media, in particular composite materials. A Mori–Tanaka[68] scheme was presented for modeling the overall thermal conduction behavior of composites containing reinforcements with interfacial resistances and prescribed size distributions. The approach was used for studying composites reinforced by spherical particles with monomodal and bimodal log-normal volume fraction distributions. But the Mori–Tanaka predictions were partially corroborated by two-dimensional numerical simulations confirming the experimentally observed considerable sensitivity of macroscopic conductivities to the shape of particles. Halpin–Tsai[69] derived a theoretical model for the transverse thermal conductivity using

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the analogy between in-plane field equations and boundary conditions to the transverse transport coefficients.

$$\kappa_{yy} = \kappa_{xx} = \kappa_m \left[\frac{1 + \xi v_f \eta}{1 - \eta v_f}\right], \quad (1)$$

$$\eta = \frac{\kappa_f/\kappa_m - 1}{\kappa_f/\kappa_m + \xi}, \quad (2)$$

where κ_{xx} and κ_{yy} are the transverse thermal conductivities in the x- and y-directions respectively, κ_f is the thermal conductivity of the fiber, v_f is the fiber volume fraction, and κ_m is the thermal conductivity of the matrix. ξ is the empirical geometric shape parameter used to account for discontinuous lamellar arrangement fibers. The factor ξ is usually determined experimentally using curve fitting methods. Numerical solutions consistent with the governing equations of elasticity have been developed for the effect of filament shape on transverse moduli. In 1904, Maxwell Garnett developed a simple but immensely successful homogenization theory,[70] which approximates a complex electromagnetic medium. The Maxwell Garnett mixing formula gave the permittivity of this effective medium in terms of the permittivity and volume fractions of the individual constituents of the complex medium. It was the basis relation of many recent models for effective thermal conductivity of nanofluids. The model was based on the solution of heat conduction equation through a stationary random suspension of spheres. The effective thermal conductivity (κ_{eff}) depends on the thermal conductivity of the spherical particle (κ_p), base fluid (κ_f) and particle volume fraction of the suspension (ϕ). The Maxwell's formula[71] is expressed as

$$\kappa_{eff} = \frac{\kappa_p + 2\kappa_f + 2(\kappa_p - \kappa_f)\phi}{\kappa_p + 2\kappa_f - (\kappa_p - \kappa_f)\phi}\kappa_f \quad (3)$$

The Maxwell's formula gave a good result for well-dispersed non-interacting spherical-shaped particles with low particle volume concentrations and with negligible thermal resistance at the particle/fluid interface. The model failed to predict a good match with the experimental results for high solid concentration as well as the effect of different parameters involved, especially the particle size of nanoparticles, even in low particle volume concentrations (Table 1).

3. State-of-the-art multiscale heat transfer modeling

In recent decades, revolutionary progress has been made in nanotechnology and nanoengineering, in particular in the synthesis and processing of materials with representative structures or functional parts down to the nano-meter scales. There is an urgent demand for a deeper understanding of thermal transport in nano-scale devices, nanostructures, and heterostructures due to the development of such modern science and technology. Lots of progress in relevant experiments and modeling with individual computational methods has been achieved in the past two decades. To acquire this knowledge, we suggest readers should read the recent comprehensive review papers.[72,73] Over the past few decades, interest in the simulation of micro- and nano-scale heat transfer has sparked the development of a variety of multi-scale models and numerical methods for phonon transport in semiconductors and dielectrics. There are also a few comprehensive literature review papers in this area.[74–76] Here, we highlight some representative developments. Hybrid models are defined herein as concurrent multiscale modeling techniques in which discretized continuum methods (*e.g.*, finite element method) are coupled with various types of atomistic methods (*e.g.*, molecular statics (MS) or molecular dynamics (MD)). Boundary conditions have been shown to strongly affect the lattice temperature inside the device.[77–79] An accurate treatment of boundary conditions requires linking subcontinuum thermal transport by phonons inside the device to the continuum heat diffusion outside the device. An essential feature of multiscale modeling approaches is the way in which communication (energy and momentum exchange) is handled in the interface between the continuum and atomistic regions. This is also the common challenge for all present multiscale modeling of other physical properties. Generally, the interface region is divided into two subregions: a "handshake" zone and a "padding" zone. The size and nature of these zones depend on the specific type of the multiscale modeling method.[80] The coupling boundary conditions between the continuum and atomistic regions can be subdivided into "strong compatibility" and "weak compatibility", both being applied within the so-called padding region. The handshake region, wherein the transition can be abrupt with no handshake region, can exist and provide a gradual transition from the atomistic to the continuum model treatment of the continuum region itself, which can be based on FEM. Currently, there are two ways to achieve the hybrid method for heat conduction: (a) decoupled scheme, according to which the information is transferred by some parameters and different methods for different length scale are conducted individually; and (b) coupled scheme, where the solutions of different methods will be coupled during their solving process, and the final full solution correspond to the converged results of different methods.[76]

Table 1 Advantages and disadvantages of typical numerical methods for heat transfer

	Advantages	Disadvantages
ALD/BTE	Very accurate when combined with first-principles calculations	High computational demand for large supercells; cannot simulate interfaces
NEGF	Can obtain frequency dependent transmission coefficients	To date, applies to ballistic phonon transport only
Classical MD	Fast; high computing efficiency; can simulate large-scale inhomogeneity	Empirical; largely relies on accurate potential; lack of quantum effects
MC/BTE	Fast; can simulate large length scales (even beyond microns)	Requires input parameters from atomistic level simulations
Continuum	Fast; can simulate continuum length scales and long time scales	Cannot simulate non-Fourier heat conduction

3.1 Decoupled scheme

The decoupled scheme is mainly applicable for the coupling of microscopic and mesoscopic methods, since both methods deal with the detailed information of heat carriers and it is easy to achieve information exchange just by several parameters. For example, in previous studies, molecular dynamics simulations and finite element methods were used to evaluate the effective thermal conductivity of graphene epoxy nanocomposites,[81–83] SiC-reinforced aluminum metal matrix composite,[84] nanometer-scale integrated circuits,[85] and interfacial phonon transport through a Si/Ge multilayer film.[86] Thermal boundary conductance between crosslinked epoxy and the graphene sheet is obtained by performing classical molecular dynamics simulation, which can take into account various atomic-level structure topology and detailed interface conditions, and is input later to the finite element based representative volume elements to evaluate the local thermal conductivity constants of the nanocomposites (Fig. 4).

Another example is the multiscale modeling of thermal conductivity of polycrystalline graphene sheets.[87] First, Green–Kubo equilibrium molecular dynamics (GK-EMD) simulations were performed for the evaluation of the thermal conductivity of ultra-fine grained graphene sheets with grain sizes ranging from 1 nm to 5 nm consisting of 25 to 400 grains (Fig. 5). Then, the macroscopic polycrystalline graphene models were constructed using the finite element approach, where all the grain boundaries were assumed to exhibit an effective contact conductance which was acquired by fitting the finite element results to the GK-EMD results for ultra-fine grained structures. By performing the finite element calculations for the systems with larger grain sizes, close agreement between the finite element results and the GK-EMD extrapolated curve was observed. With this model, the effect of grain size distributions on the effective thermal conductivity of polycrystalline graphene sheets was further investigated. Obviously, the accuracy and effectiveness of the large scale finite element results depend on the strong assumption of uniform contact conductance across the grain boundaries, which is questionable in the cases of highly inhomogeneous grain orientations and large extended defects occurring at the boundaries.

3.2 Coupled scheme

Instead of using some parameters to transfer information across different length scales, the coupled scheme runs different methods simultaneously and the information exchange is achieved during every iteration step. In the nano- and micro-scale, phonon scattering by nano-scale structural features and anharmonicity can be calculated atomistically using first-principles, whereas ballistic and diffusive transport are automatically accounted for *via* the space-dependent distribution functions.[88] However, the solution of the BTE is a challenging task, especially in complex geometries. The Monte Carlo method is especially flexible for use with complex geometric configurations and can readily include different scattering mechanisms. To explicitly study the non-Fourier heat conduction, phonon MC simulation is an approach worth recommending. Li *et al.*[89] presented a new hybrid phonon Monte Carlo-diffusion method for ballistic-diffusive heat conduction. They used an alternating method, similar to the Schwarz technique proposed for the coupled Stokes/DSMC problem in the fluidic simulation, to couple the phonon tracing MC and Fourier's law.[90] It is found that the hybrid method can accurately predict the distributions of temperature and heat flux in the system with nearly the same precision as the phonon tracing MC while the computation time can reduce up to 90%, validating its potential use for larger and more complex structures. An efficient method has been developed to solve the space-dependent Peierls–Boltzmann equation *via* variance reduced Monte Carlo (VRMC).[91–93] The calculations presented in this pioneering work were not *ab initio* and resorted to the simplifying assumption of spherically symmetric phonon dispersions with scattering rates only dependent on energy. Recently, VRMC has been generalized and implemented with full dispersions and scattering amplitudes calculated *ab initio* for real materials and released as the program almaBTE.[94] Regarding the critical parameters of the interfacial phonon scattering process, currently almaBTE adopts the assumption that particles (phonons) reaching the interfaces between dissimilar materials will be transmitted/reflected according to the previously widely used diffuse mismatch model that allows for elastic mode conversions. It is worth pointing out that all

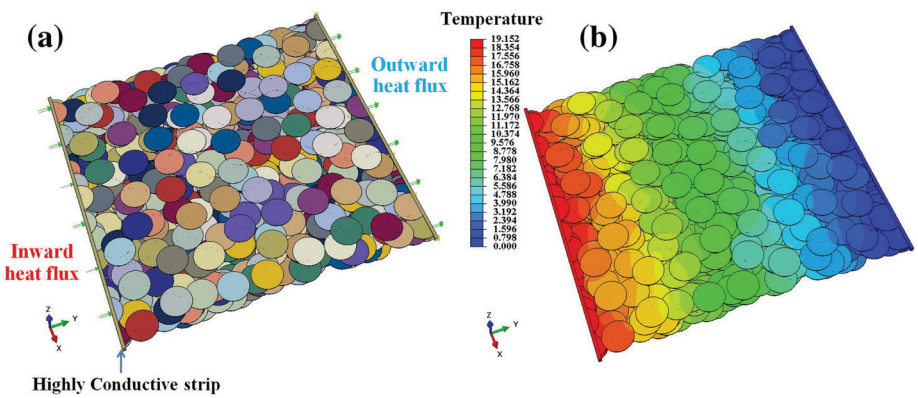

Fig. 4 A typical 3D representative volume element of graphene laminate constructed in Abaqus/Standard. This figure was taken from ref. 184 with permission from Elsevier.

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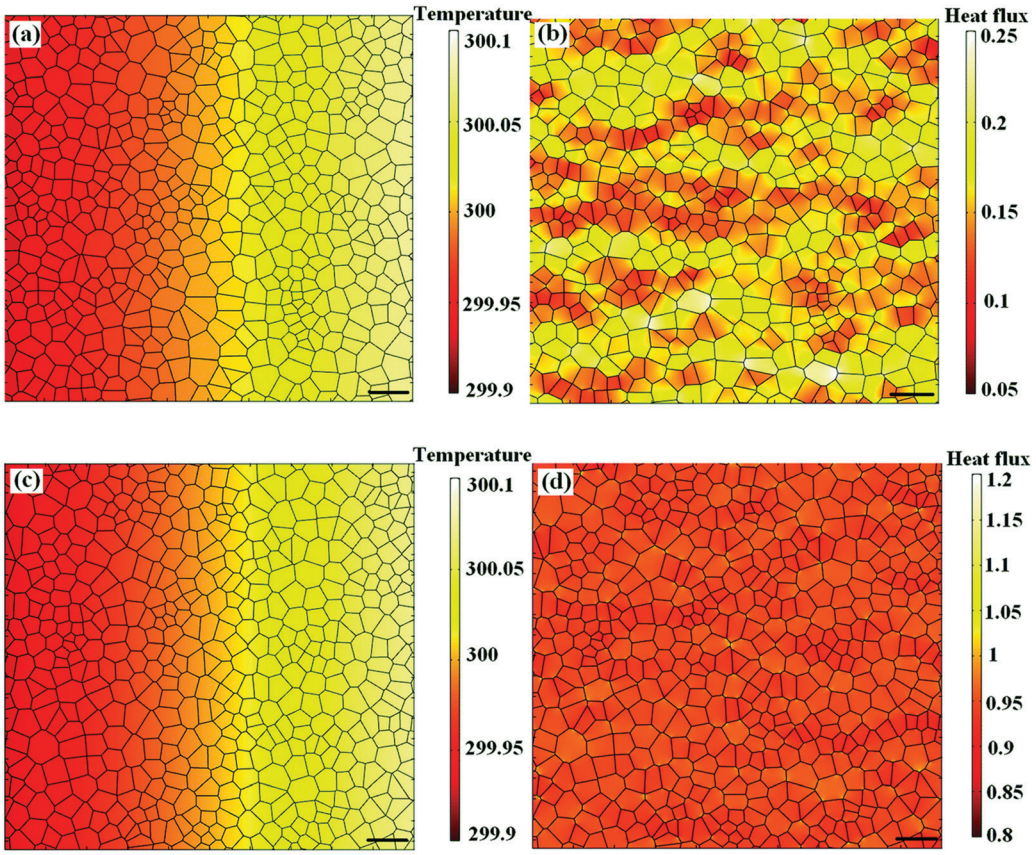

Fig. 5 Samples of constructed finite element models for the evaluation of thermal conductivity of polycrystalline graphene structures. (a) Temperature and (b) heat flux distribution of a polycrystalline graphene sheet with an average grain size of 5 nm. (c) Temperature and (d) heat flux distribution of a polycrystalline graphene sheet with an average grain size of 500 nm. Figure was taken from ref. 87 with permission from the Royal Society of Chemistry.

empirical models, such as the diffuse mismatch model, may have some problem or yield an inaccurate phonon scattering process, in particular when the interfacial structures become complicated and/or there are multiple interfaces/interlayers with thickness well below the characteristic length of phonon mean free paths in the respective bulk materials (the particle picture of phonons will fail in this case, *i.e.*, the wave nature of phonons would become significant).

4. Perspective on future multi-scale heat transfer modeling

4.1 Promising role of artificial intelligence in bridging mono-scale methods

4.1.1 Urgent need of accurate but robust interfacial heat transfer model. As discussed above, the most critical element for future multi-scale heat transfer modeling is the development of a robust, as accurate as possible, "interfacial" heat transfer model to deal with the phonon scattering process across boundaries and interfaces, near voids and surfaces, and a sophisticated model connecting the simulations across different length scales. An ideal multi-scale modeling framework should be equipped with the feature that there are different heat transfer physics across different length scales and in the meantime the physics crossing different scales is interdependent and coupled. To this end, the above coupled scheme that integrate different mono-scale methods into a single framework is very promising. Certainly there is an urgent need to smoothly bridge these mono-scale computational methods. A single, seamless, and concurrent multiscale thermal transport framework is illustrated in Fig. 6. A similar approach has been implemented in solid mechanics, such as a finite-temperature quasicontinuum (QC) method.[95–99] Such a method would provide a complete description of thermal transport in complex structures from scattering at atomistic defects to the thermal transport behavior at very large length scales. To ensure the accuracy of predictions, machine learning interatomic potentials tuned to first-principles calculations can be used at the MD level to correctly capture defect effects, as machine learning has shown a promising role in MD simulations toward calculating intrinsic lattice thermal conductivity and interfacial thermal resistance[100–102] *via ab initio* trained neural network interatomic potentials.[103–105] As such, this approach may be an alternative to direct *ab initio* calculations of forces, and may enable examination of larger and more complex material systems with the highest possible accuracy.

4.1.2 Promise of machine learning interatomic potentials for studying heat transfer in heterostructures. It is clear that many traditional interatomic potentials, such as the embedded-atom

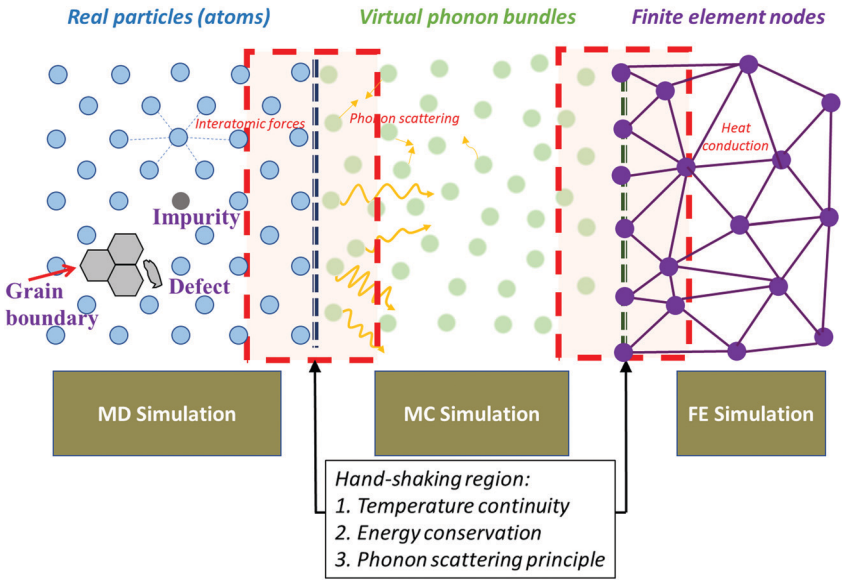

Fig. 6 Schematic of the multiscale thermal transport framework composed of atomistic (molecular dynamics) simulation, mesoscopic (Monte Carlo) simulation, and macroscopic (finite element) simulation. The red dashed boxes represent the hand-shaking regions connecting different mono-scale methods.

method (EAM), Tersoff potential, charge optimized many-body (COMB), and reactive force field (ReaxFF), have promoted the development of precisely describing the interatomic interactions. However, these potentials usually address particular classes of materials or particular types of applications and suffer from poor transferability to unknown structures. Smith *et al.*[106] demonstrated how a deep neural network trained on quantum mechanical DFT calculations can learn an accurate and transferable potential for organic molecules. Pun *et al.*[107] proposed a new approach that could drastically improve the transferability of machine learning potentials by informing them of the physical nature of interatomic bonding. With further development of machine learning based interatomic potentials, it is highly expected that such achievements will significantly change the state-of-the-art thermal transport in solids and related interfaces. Behler's group has initiated a lot of work on neural network potentials for various materials including organic molecules and surfaces.[108–111] Typical recent improvement in neural network potentials include deep potential molecular dynamics (DPMD) with a carefully crafted deep neural network that preserves all the natural symmetries,[112] neural network force fields from energy decompositions,[102] and spatial density neural network force fields (SDNNFFs).[113] Using well-trained neural network potentials, much larger scale MD simulation than traditional MD can be conducted, in particular for heterostructures or inhomogeneities that occur on a meso-scale or even larger. For example, Mortazavi *et al.* trained machine-learning interatomic potentials (MLIPs) over short AIMD trajectories that enable first-principles multiscale modeling, where DFT simulations can be hierarchically bridged to efficiently simulate macroscopic structures.[114] They performed a case study of the lattice thermal conductivity of two-dimensional graphene/borophene heterostructures and were subsequently able to extend the study of effective thermal transport along the heterostructures at a continuum level.

4.1.3 Role of machine learning in bridging thermal transport across different length scales. Machine learning may play a more critical role in bridging mono-scale methods as shown in Fig. 6 (the dashed boxes). As in all concurrent multiscale methods, the challenge is in providing rigorous energy conserving coupling between domains of different resolution. This requires that, temperature continuity, energy and momentum flux across MD/MC and MC/FE interfaces, and other phonon scattering principles should be fulfilled simultaneously (Fig. 6). To model heat energy transfer between the MD and MC regions, the frequency domain direct decomposed method (FDDDM) is a suitable approach.[115–117] FDDDM provides frequency-dependent heat flux across any virtual plane perpendicular to the heat flux direction in the system, which has already been applied to various interfaces[118–121] and even bulk systems.[122,123] For complex interfaces or grain boundaries, the phonon scattering process is too complicated to be modeled in great detail or in a deterministic way. In this case, one can use deterministic MD simulations to sample a large amount of representative interfacial or grain boundary structures and their corresponding interfacial thermal transport behavior, *e.g.*, the frequency-dependent transmission by the FDDDM,[124] and then utilize machine learning techniques to obtain high-fidelity models to quickly and accurately predict the interfacial thermal transport across new interfaces or grain boundaries. Machine learning or broader artificial intelligence algorithms can also be used for minimizing statistic errors of parameters in the empirical models such as conventional models for phonon-impurity and phonon-boundary scattering, and also for narrowing down the upper and/or lower bounds of model predictions (*i.e.* reducing prediction uncertainty).

4.2 Quantitative deep learning for the optimization of complex thermal transport processes

In complex structures such as polycrystalline materials, grain boundaries are sites of enhanced atomic motion, but the

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complexity of the atomic structures within a grain boundary network makes it difficult to link the structure and atomic dynamics. In particular, thermal transport across grain boundaries is still not well understood,[125] due to challenges in obtaining experimental data and limitations in simulation studies that rely on employing empirical potentials. In the past two decades non-equilibrium molecular dynamics (NEMD) simulations have mainly been performed to examine thermal conductivities of individual grain boundaries.[126–130] General results revealed that thermal conductivity normally varies with misorientation angle and grain boundary energy; however, the underlying physical mechanism has not been elucidated in terms of the detailed grain boundary structures. The difficulty resides in that, grain boundaries are actually high dimensional space which makes the single or few structural parameters proposed in a previous study[131] insufficient for explaining the structure–property relationships. In this regard, machine learning, in particular deep learning, could have great potential to quantitatively predict thermal transport in polycrystalline structures and also extract/identify the dominant structural factors that govern the thermal transport process. A schematic of using deep learning to study the phonon scattering process across grain boundaries and bridge the nano-scale and meso-scale simulation is illustrated in Fig. 7. This framework is composed of integrating (i) machine learning of interatomic potentials for the local atomic potential landscape at a nano-meter or atomic scale; (ii) NEMD method for probing phonon scattering/transmission at grain boundaries (up to a hundred nanometers), such as using FDDDM as mentioned above; and (ii) the Peierls–Boltzmann transport theory for simulating phonon transport in much larger length scales (up to a hundred microns).

Specifically, step (i) involves training interatomic potentials for nano-scale MD simulations (see the right-bottom panel of Fig. 7).

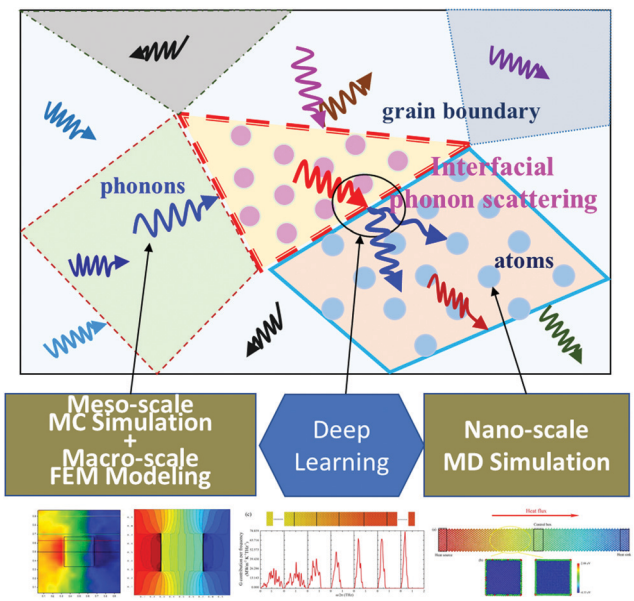

Fig. 7 Schematic of using deep learning to study the phonon scattering process across grain boundaries and bridge the nano-scale and meso-scale simulation.

Lots of machine learning techniques can be used in this regard, *e.g.*, the latest techniques of DPMD[112] and SDNNFF[113] as mentioned above. For step (ii), it is worth emphasizing here that SDNNFF[113] is promising for the study of thermal transport across grain boundaries. The SDNNFF method proposed by the Hu group was originally established on training total forces of each individual central atom i (denoted as $\boldsymbol{f}_i$) according to its local environment or spatial distributions of neighbors. This method focuses on the usage of a three-dimensional mesh of density functions, which together act as a mapping of the atomic environment and provides a physical representation of the forces acting on the central atom. Currently, the Hu group is extending the SDNNFF method to train interatomic forces, *i.e.*, interaction between a central atom i and its neighbor pair j (denoted as $\boldsymbol{f}_{ij}$). It should be noted that $\boldsymbol{f}_{ij}$ is directly relevant to the atomistic heat current $\boldsymbol{q}_{ij}$ between two atoms i and j[116]

$$\boldsymbol{q}_{ij} = \frac{1}{2}\left\langle \boldsymbol{f}_{ij}\left(\boldsymbol{v}_i + \boldsymbol{v}_j\right)\right\rangle, \tag{4}$$

where $\boldsymbol{f}_{ij}$ is the force between two atoms i and j, $\boldsymbol{v}$ is the velocity of atoms, and $\langle\cdot\rangle$ denotes the time average. Then, training effective $\boldsymbol{f}_{ij}$ will enable quantitative characterization of atomistic heat current and summing $\boldsymbol{q}_{ij}$ over the grain boundary, which can be regarded as a special interface, will enable investigation of interfacial heat transport across grain boundaries. Note that the number of atoms in the grain boundaries is huge and that the training data for the atomic pairs (i and j) and associated atomistic heat current $\boldsymbol{q}_{ij}$ are also huge, which will enable a well-trained deep learning neural network model, a similar strategy as we used in the previous SDNNFF method.[113] Once the neural network model for $\boldsymbol{f}_{ij}$ is trained, it is then straightforward to couple with the previously developed FDDDM[116] approach to evaluate the phonon spectrum of cross-boundary thermal transport in the framework of NEMD (see the schematic in the middle-bottom panel of Fig. 7). Another promising approach is the neural network force field by direct energy decomposition,[102] which is based on extracting atomic energies from DFT calculations. It has been used for calculating the thermal conductivity of amorphous silicon based on long molecular dynamics simulations. As per-atom energy is trained, it is highly expected that this approach can be extended to the NEMD framework as well (atomistic heat flux can be derived from the spatial change of atomic energies or energy densities), and then the above phonon spectrum of interfacial heat flux can be extracted from the NEMD simulation, which brings us to step (iii). In step (iii), deep learning can be used for "learning" the patterns of the frequency dependent interfacial heat spectrum and correlating the interfacial thermal conductance or resistance to the features of local atomic structures at the grain boundaries (see the left-bottom panel of Fig. 7). Deep learning could also be powerful in identifying the dominant phonon scattering process across numerous grain boundaries when using an empirical model of phonon-grain boundary scattering as input parameters for higher level BTE modeling, since in this case the space of adjustable computational parameters is so huge that studying the effect of combinations of all parameters

is impossible. Identifying the governing phonon-grain boundary scattering events is critical for tailoring the atomic structures for achieving desirable thermal transport properties. The computational framework proposed in Fig. 7 is expected to establish a quantitative understanding of thermal transport across various types of grain boundaries using the high predictive power of atomistic simulations with first-principles level accuracy, and also has the potential to keep the computational cost several orders-of-magnitude cheaper than that of the direct atomistic simulations.

Very recently, machine learning with data derived from the smooth overlap of atomic positions (SOAP) descriptor and perturbed MD has been utilized to quantify the relationship between local atomic structure and overall thermal conductivity in standard- and high-pressure symmetric tilt grain boundaries, twin, twist, and asymmetric tilt grain boundaries of MgO.[132] A simple metric based on the SOAP descriptor, namely local distortion factor (LDF), was proposed and analyzed to correlate with atomic thermal conductivity of the polycrystalline MgO in a non-linear fashion. The importance of structural disorder at grain boundaries for phonon transport in polycrystals is consistent with a previous study on nanocrystalline diamond.[133] Having developed the machine learning model, it is straightforward to explore the entire high-dimensional space of grain boundaries to precisely control or tailor the thermal transport, provided that the trained model is capable of correctly representing the local structural distortion to the largest extent. It should be noted that, this kind of method is limited to the system size due to the nature of full atomistic simulation.

4.3 Uncertainty quantification in multi-scale heat transfer modeling

Uncertainty quantification (UQ) and big data analysis have received increasing attention in recent years. Extensive research effort has been devoted to these topics, and novel numerical methods have been developed to efficiently deal with large-scale data sets and complex problems with uncertainty. Both UQ and big data analysis enable us to better understand the impacts of various uncertain inputs (boundary and initial data, parameter values, geometry, network, *etc.*) on numerical predictions. UQ and big data analysis are thus critical to many important practical problems. As the data size and dimensions of parameter space increase, one of the biggest challenges in UQ computations and big data analysis is the computational cost for analyzing the data and running the simulations. From the multi-scale heat transfer modeling point of view, since there are lots of empirical models and/or trained models that will be used to determine the final material property (such as effective thermal conductivity or overall interfacial thermal resistance) or predict phonon transport process in complex structures, UQ will undoubtedly play a critical role in providing confident prediction results to the community. Uncertainty can exist everywhere in multi-scale simulations, from the pseudo plane wave potential used in first-principles,[53,134–136] to the interatomic potentials used for GK-EMD or NEMD simulations,[137–143] to the empirical phonon-boundary/-surface/-impurity scattering formula used in phonon BTE modeling,[144] to the uniform effective thermal resistance used in thermal transport across grain boundaries.[145] One of the intuitive ideas is to combine detailed theoretical models and experiments for selected model systems and/or structures with UQ algorithms such as the Bayesian decision framework to develop better theories, interpret emerging experiments correctly, design better experiments and simulations, and quantify the uncertainty in the predictions.[146,147] To date, few studies have been conducted on systematic UQ analysis of thermal transport,[148] even for mono-scale computational methods. For multi-scale thermal transport simulation, the first significant difficulty lies in that the governing physical mechanism is still not clear. For instance, on which scale is the thermal transport dominated in terms of thermal resistance and what is the corresponding thermal transport mechanism (ballistic or diffusive or hybrid)? What is the relationship between such a dominant thermal transport process and the local structure? In this sense, the UQ analysis is strongly coupled with the algorithm or framework development of multi-scale thermal transport itself. Without a clear understanding of the thermal transport in the entire system, the UQ analysis cannot be performed. The second challenge is due to the complexity of phonons which is usually mode dependent. So far, precise and sophisticated atomistic models for predicting mode dependent phonon-impurity/-surface/-boundary-/interface scattering are very rare. Despite some work that has been initiated for phonon-interface interaction in the framework of NEMD simulations[115,116,149,150] and phonon wave-packet dynamics[151–153] (so far limited to zero temperature), the small scale that current MD simulations can handle limits the wide applications of the relevant algorithms to a larger scale. It is worth pointing out that some multi-scale models such as concurrent atomistic-continuum modeling[154,155] have been used for simulating phonon-dislocation interactions.[156,157] Such models need further development to include intrinsic phonon anharmonicity in the materials. Also, development of other types of phonon-"defect" interaction is still missing. More importantly, how to transfer these algorithms to higher scales so that the phonon-"defect" scattering process at larger length scales can be understood is still an open question. Again, the UQ analysis first calls for the fundamental understanding of these transport phenomena and foundation of the relevant algorithms or frameworks.

4.4 Phonon interaction with other energy carriers

There are four principal energy carriers from an atomistic point of view: (1) ions/molecules; (2) atoms (lattice, phonons); (3) electrons (including magnons); and (4) photons. In the development of energy transport materials, most of the time a single type of energy carrier is functioning. However, sometimes there is strong interaction or coupling between two different energy carriers and sometimes even among more carriers. Considering thermal transport as an example, thermal conductivity measures a material's ability to conduct heat and is intrinsically determined by the quantum behaviors of electrons and phonons. In most semiconductors with a finite band-gap, the heat transport is dominated by phonons, and the lattice thermal conductivity is mainly limited by the phonon–phonon interaction (PPI) and extrinsic scattering due to defects or isotopes, and the contribution of

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electron–phonon interaction (EPI) or electron–phonon coupling (EPC) is usually negligible. However, recent studies found that EPC can be significant in affecting the thermal conductivity of a highly polarized GaN crystal with strong Fröhlich EPC for the longitudinal optic phonons with a long wavelength,[135] bulk silicon with high carrier concentrations,[158] layered metal oxide,[159] pure metals,[160] and even low-dimensional materials.[161] EPC has also long been believed to play a critical role in determining superconductivity.[162–168] Therefore, a promising and interesting research direction is to include the EPC effect into the existing mono-scale computational methods, such as phononic or electronic BTE, so that the energy transport mechanisms or phenomenon can be probed in a more realistic and accurate way that is comparable with experiments and mimics the situation in practical applications. It is highly expected that, by considering EPC in phononic and electronic BTE, the root reason for excess heat generation in micro-/nano-electronics and the heat dissipation on a larger scale can be deeply understood and then novel device-level architectures pertaining to more efficient thermal management will be designed and fabricated.

Moreover, due to the inherent coupling or interaction between phonons and other principal energy carriers, phonon-assisted photonic quantum transport phenomena have been extensively observed.[169–175] Reindl *et al.*[176] showed an unprecedented two-photon interference from remote strain-tunable GaAs quantum dots emitting on-demand photon-pairs by exploiting the full potential of a novel phonon-assisted two-photon excitation scheme. This study marks an important milestone for the practical realization of advanced photonic quantum technologies and complex multiphoton entanglement experiments involving lattice vibrations such as dissimilar artificial atoms. The experiments will probe theorists to simulate the relevant phenomena by quantifying phonon–photon interactions. Analogously, one can extend this idea to other energy carriers such as ions, *i.e.* explore how the lattice vibrations will assist (better enhance) ionic transport, which is extremely important for fast ionic conductors in the energy storage field. Kraft *et al.*[177] investigated the influence of interatomic bonds (lattice vibrations) on the ionic conductivity in the lithium superionic argyrodites Li_6PS_5X (X = Cl, Br, I). They found that the lattice softness has a striking influence on the ionic transport: the softer bonds lower the activation barrier and simultaneously decrease the prefactor of the moving ions, which provide a useful guidance for tailoring the lattice stiffness of materials in order to maximize ionic conductivity. Still, much more needs to be done, *e.g.* systematic atomistic simulations of concurrent ionic and thermal (phononic) transport to uncover the entangled relation between phononic transport and ionic transport, and embedding these atomistic models into large scale modeling framework to investigate the effect of large scale inhomogeneity.

5. Summary

Understanding and controlling subcontinuum phonon conduction has emerged as a critical issue in recent decades, not only for the cooling of field-effect transistors with gate lengths less than 100 nm, but also for developing high performance energy conversion and storage systems, where the multiscale simulation method can play a central role in quantitatively predicting phonon transport across different length scales and time scales. Another motivation for multiscale simulation methods is that it is always neither possible nor necessary to calculate the full atomistic information in the whole simulation domain. This perspective describes the state-of-the-art multiscale thermal modeling and also points out some future research directions in this regard. Similar to the multiscale methods in other fields, the current issue of these approaches resides in providing a seamless bridge between the atomistic and continuum approaches, even if sometimes intermediate ''mesoscopic'' methods of simulation were introduced. Phonons are inherently highly dimensionally dependent (wave vector and polarization dependent), and the phonon mean free path can span orders of magnitude in space. This adds too much complexity and difficulty when dealing with phonon-inhomogeneity interaction at different length scales. We highlight the potential of artificial intelligence such as machine learning techniques in future multi-scale modeling development for identifying the dominant role or mechanism when multiple phonon-inhomogeneity interaction occurs simultaneously. We also emphasize the importance of uncertainty quantification in the multi-scale thermal transport simulation, which is closely related to our understanding of heat conduction across different scales. Additionally, phonon coupling or entangling with other principal energy carriers is foreseen to be a new area in the near future, where more fundamental and previously unexplored thermal transport mechanisms and physics, such as ultrafast phonon dynamics[178–181] and multi-channel thermal transport,[122,123,182,183] will be discovered and the relevant technologies will be promoted.

Conflicts of interest

There are no conflicts to declare.

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