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TOPICAL REVIEW — Phononics and phonon engineering

Tuning thermal transport via phonon localization in nanostructures*

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Localization, one of the basic phenomena for wave transport, has been demonstrated to be an effective strategy to manipulate electronic, photonic, and acoustic properties of materials. Due to the wave nature of phonons, the tuning of thermal properties through phonon localization would also be expected, which is beneficial to many applications such as thermoelectrics, electronics, and phononics. With the development of nanotechnology, nanostructures with characteristic length about ten nanometers can give rise to phonon localization, which has attracted considerable attention in recent years. This review aims to summarize recent advances with theoretical, simulative, and experimental studies toward understanding, prediction, and utilization of phonon localization in disordered nanostructures, focuses on the effect of phonon localization on thermal conductivity. Based on previous researches, perspectives regarding further researches to clarify this hectic-investigated and immature topic and its exact effect on thermal transport are given.

Keywords: thermal conductivity, phonon localization, nanostructure

PACS: 65.80.–g, 63.22.–m, 63.50.–x, 63.20.Pw **DOI:** 10.1088/1674-1056/abb7fa

1. Introduction

Wave (coherent) and particle (incoherent) transport are two of the most basic properties for physical entities, such as electrons, photons, and phonons, which directly determine the properties, ranging from electronic, optical to thermal, of materials. For coherent transport, the phase information of energy carriers must be considered, which can give rise to interference. One of the most attractive phenomena is the multiple scattering of waves by disorders,[1] which can bring transport to a complete halt.[2–6] This phenomenon, named Anderson localization, was first proposed by Phillip Anderson when investigating the coherent transport of electrons through crystals with a random potential.[1] The localization of electron can result in a metal–insulator transition.[1] As the electron localization originates from the interference of multiple scatterings of electronic waves, Anderson localization is expected to be ubiquitous in coherent transport. Later, Anderson localization has been demonstrated for photon[3,4] and acoustic waves[5,6] by both theoretical and experimental studies in low- and high-dimensional systems.[7]

Phonons are quantized vibration modes in materials, which are the main heat carrier in semiconductors and insulators.[8] During the past decades, thermal engineering by manipulating the phonon transport has attracted great attention due to the low efficiency of energy utilization and rapid increase in power density of microelectronics.[9–12] On one hand, thermoelectric devices can convert heat into electricity directly, which are expected to reutilize waste heat and enhance the efficiency of energy utilization.[13–17] An effective way to enhance the efficiency of thermoelectric devices[14,18–25] is reducing lattice thermal conductivity while keeping the electric properties of the materials the same. On the other hand, heat dissipation becomes one of the critical factors that limit the further development of microelectronic devices,[26,27] which lies in the decreasing of the interface thermal resistance (Kapitza resistance)[28–34] and exploring materials with high thermal conductivity.[35–37] What is more, electronic similar devices, such as thermal diode,[38–43] thermal transistor,[44–49] and thermal logic gates,[50] enable the constructive control of heat flow.[51] Phononics would therefore lead to abundant application of heat.[51,52]

Owing to the progress of nanotechnology, materials can be fabricated with nanoscale precision in morphology and composite.[53] Comparing with macroscale thermal transport in traditional bulk materials governed by the Fourier's law, thermal transport in nanostructures can be hugely changed due to the modulation of phonon relaxation time and group velocity in different ways.[9,21,54–68] Generally, the exercised approaches to control phonon transport through nanostructures are based on coherent and incoherent nature of phonons, or a combination of both two. Over the past decades, most of the commonly adopted approaches, such as diffusive interface/surface scattering,[54,69–71] defects scattering,[72–74] and

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high-order phonon scattering,[75,76] are mainly based on the phonon incoherent picture. This is because the characteristic scale of nanostructure should be on the order of phonon wavelength, which is about 1–10 nm at room temperature.[77,78] In addition, boundaries of nanostructures should be smooth enough to maintain the phase information of phonon.[79,80] These pose great difficulties for materials fabrication and experiment measurement. Thus, tuning phonon transport via its coherent nature is challenging.

Since the investigating of thermal transport in superlattice,[55,56,81–83] phonon coherent transport becomes an active area of research. It is found that band-folding by periodic phononic crystals (PnCs)[21,84–90] and resonance hybridization by nanopillars[20,58–61,91–93] or embedded nanoparticles[94–96] via phonon interference can effectively modulate the thermal conductivity. Since the Anderson localization is essence a wave effect,[1] which should also occur in phonons. During the past decade, more and more researchers have focused on phonon localization, and found that large reduction of thermal conductivity can be achieved. A lot of reviews have been published on the topic of band-folding based periodic PnC[78,80,97] and resonance hybridization based metamaterials,[98] few have been focused on phonon localization. Considering phonon localization as an emerging topic, it is necessary to review the present progress, and provide an outlook for further work.

In this review, the recent progress and understanding on phonon localization are summarized. In Section 2, a brief outline about the coherent nature of phonons is given. In Sections 3–5, recent studies about phonon localization by surface morphology, disorder of impurities, and secondary artificial periodicity in PnC are summarized, which focus on the change of thermal conductivity. In Section 6, the conclusions are given, and brief perspectives on the challenges and outlook are presented.

2. The coherent nature of phonon

As mentioned above, the coherent nature of phonon works only when the characteristic scale of a nanostructure is on the order of phonon wavelength, which is much smaller than the phonon mean free path.[11,69,99] In the early years, due to the limitation of nanotechnology, the characteristic scale of the nanostructure is only on the order of the phonon mean free path.[100] Thus, phonon transport cannot be tuned by its coherent nature. The change in thermal conductivity of nanostructures can be well explained by the incoherent nature of phonon through involving diffuse scattering of boundaries or interfaces.[9,10,77,81,86] In 2000, Mahan *et al.* investigated the thermal conductivity of superlattice on the assumption of coherent phonon transport, and found a minimum thermal conductivity with the varying periodic length.[55] The findings attracted great attention[82] due to the potential application of superlattice on thermoelectrics.[14,83] Later, the results were demonstrated by molecular dynamics simulation[84,101,102] and experiment.[56] In 2012, the coherent nature of phonons was experimentally confirmed.[79]

To verify the coherent nature of phonon, Luckyanova *et al.*[79] investigated thermal transport in AlAs/GaAs superlattice (Fig. 1(a)) through time-domain thermoreflectance method by increasing the number of periods while the periodic length is fixed. In AlAs/GaAs superlattice, if the phonons are diffusively scattered, they will lose phase information, and incoherently transport. In this case, the phonon mean free path is limited by each period, and the thermal conductivity will keep constant as the number of periods increases due to diffusive transport. However, if the interfaces do not destroy the phase information of phonons, the superlattice can be treated as a homogeneous material. Due to the size effect,[63–65] the thermal conductivity will linearly increase with the increasing number of periods. As shown in Fig. 1(b), the measured thermal conductivity of the AlAs/GaAs superlattice linearly increases below 150 K when the number of periods increases from 1 to 9. This is an evidence of phonon coherent transport. From 250 K to 296 K, the dependence of the thermal conductivity on the number of periods deviates from the linear relationship. This is because more high frequency phonons,[99] which have short wavelengths, take part in the thermal transport and show incoherent nature.

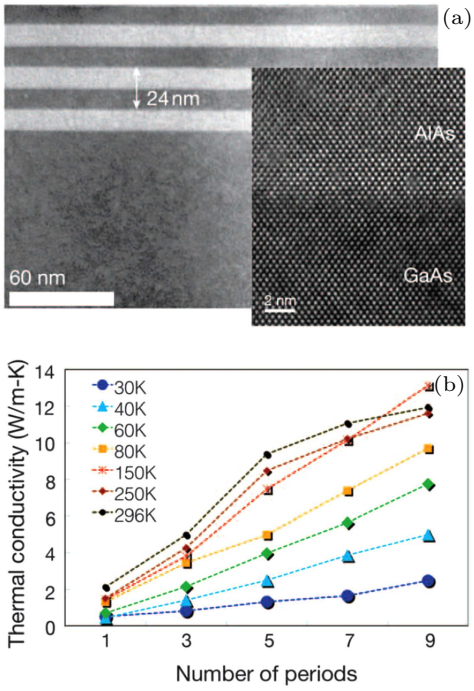


Fig. 1. (a) Transmission electron microscope image of AlAs/GaAs superlattice. (b) The dependence of thermal conductivity of AlAs/GaAs superlattice on number of periods. Reproduced with permission from Ref. [79].

In order to understand the coherent nature of phonons, an important parameter, phonon coherence length, needs to be

clarified. The phonon coherence length measures the distance that a phonon can travel while still keeping a specified degree of phase information. Latour *et al.* proposed that the coherence of phonons depends on the spatial correlations of atomic fluctuations (shown in Fig. 2).[99] Thus, the spatial phonon coherence length can be defined as[99]

$$l_c^2 = \frac{\sum_{k=0}^{N_\parallel - 1} |C(k\Delta z, \omega)|^2 (k\Delta z)^2}{\sum_{k=0}^{N_\parallel - 1} |C(k\Delta z, \omega)|^2} - \left(\frac{\sum_{k=0}^{N_\parallel - 1} |C(k\Delta z, \omega)|^2 k\Delta z}{\sum_{k=0}^{N_\parallel - 1} |C(k\Delta z, \omega)|^2} \right)^2, \tag{1}$$

where $N_\parallel$ and Δz are the cell number and lattice parameter in $e_\parallel$ direction (shown in Fig. 2(a)), respectively. $C(k\Delta z, \omega)$ is the cross correlation function which can be calculated as[99]

$$C(k\Delta z, \omega) = \frac{1}{N_\parallel - 1} \sum_{i=1}^{N_\parallel - k} \mu(z_i, z_{i+k-1}, \omega), \tag{2}$$

$$\mu(z_m, z_n, \omega) = \frac{W(z_m, z_n, \omega)}{[W(z_m, z_m, \omega)]^{1/2} [W(z_n, z_n, \omega)]^{1/2}}. \tag{3}$$

Here $\mu(z_m, z_n, \omega)$ is the two-point (z_m, z_n) cross spectral density function, which can be obtained by taking Fourier transformation of mutual coherent function $\Gamma^{\alpha\beta}(z_m, z_n, \tau)$ that is defined as[99]

$$\begin{aligned} &\Gamma^{\alpha\beta}(z_m, z_n, \tau) \\ &= \frac{1}{k_B T N_\perp} \sum_{i=1}^{N_c} \sum_{j=1}^{N_c} \sum_{\gamma=1}^{N_b} \sqrt{m_i^\gamma m_j^\gamma} \left\langle v^\alpha(r_i^{0\gamma}, t) v^\beta(r_j^{0\gamma}, t+\tau) \right\rangle \\ &\times \delta[(r_i^0 - r_j^0) \cdot e_{\perp 1}] \delta[(r_i^0 - r_j^0) \cdot e_{\perp 2}] \\ &\times \delta[(r_i^0 \cdot e_\parallel - z_m] \delta[(r_j^0 \cdot e_\parallel - z_n]. \end{aligned} \tag{4}$$

Here m, v, and r are the atomic mass, group velocity, and position, respectively. γ and i are the atomic and cell numbers, respectively. The phonon coherence length of argon calculated by this method is shown in Fig. 2(b). Obviously, the phonon coherence length decreases as the frequency increases. Thus, low frequency phonons will be more likely to show coherent nature. At low temperatures, the thermal conductivity is only contributed by low frequency phonons, it will be more easier to observe the coherent nature of phonons. That is why, the coherent nature of phonon was only observed by Luckyanova *et al.*[79] in experiment at low temperature.

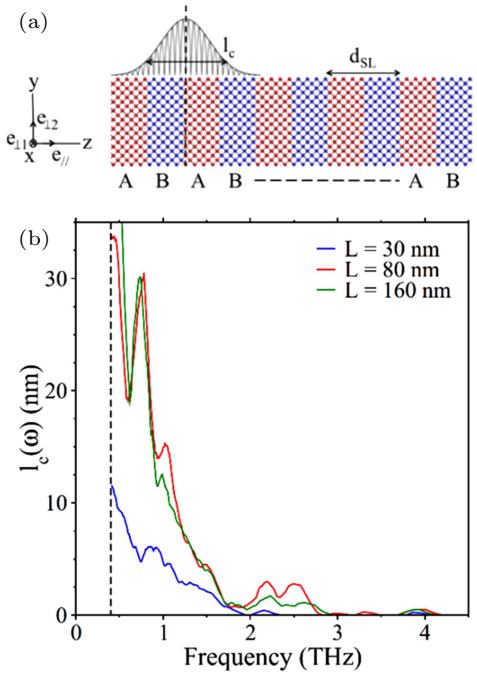


Fig. 2. (a) The schematic diagram for the extension of a wave packet on the z direction. (b) The frequency dependent phonon coherence length of argon. Reproduced with permission from Ref. [99].

3. Phonon localization by surface morphology

The nanowires, especially silicon nanowire (SiNW), have attracted great attention because of their excellent physical and chemical properties, which show huge potential in the applications of electronics,[103,104] biosensors,[105] and photovoltaics.[106,107] In 1999, through molecular dynamics simulation, Volz *et al.* predicted that the thermal conductivity of SiNW is about two orders of magnitude lower than that of bulk silicon due to boundary scattering.[54] Later, the findings were demonstrated by Li *et al.* through thermal bridge method.[108] This indicates that SiNW is a good candidate for thermoelectric application.[18] Since the reduction of thermal conductivity in SiNW is due to surface scattering,[54,108] researchers found that the increase of surface roughness can further reduce its thermal conductivity.[18,108–110] However, for the early researches, surface scattering was purely attributed to the phonon incoherent effect. The results by assuming diffusive boundary scattering based on the incoherent nature of phonons,[109,111–113] have good agreement with experiment results for SiNW with large cross section area.[109,113] However for SiNW with small cross section area, deviations occur between results from simulation and experiment.[109,113] This discrepancy simulates researchers to investigate the effect of phonon coherent transport.

A lot of researches have revealed that coherent phonon scattering by rough boundaries can hugely decrease the thermal conductivity of SiNW.[114–119] Through simultaneously manipulating the amplitude and correlation length of roughness, thermal conductivity of SiNW can even be much lower than the Casimir limit,[117] which is the low limit of incoherent transport by assuming completely diffuse phonon scattering. In the cases when the phonon coherent effect dominates the thermal transport, phonon localization is expected. Through Landauer formalism and DMPK (named for Dorokhov, Mello, Pereyra, and Kumar) model,[120,121] Murphy *et al.* found that phonon localization plays an important role in the thermal transport for nanowires with strongly disordered surfaces when the diameter is less than 20 nm.[114] The theoretical results here are in accordance with previous results from simulation and experiment. As shown in Fig. 3, when phonon localization is taken into account, the transmission functions

for middle and high frequency phonons are much smaller than those without localization. This is because the low frequency phonons have long localization length, which are hard to be localized. As a result, the thermal conductance is effectively reduced except at ultra-low temperature where the thermal conductivity is mainly contributed by low frequency phonons due to the Bose–Einstein distribution function. When the temperature increases, the difference becomes larger. However, this does not imply that phonon localization can be obvious at high temperature. Here, Murphy *et al.* assumed only coherent nature of phonons, which is just related to phonon transport at low temperatures.

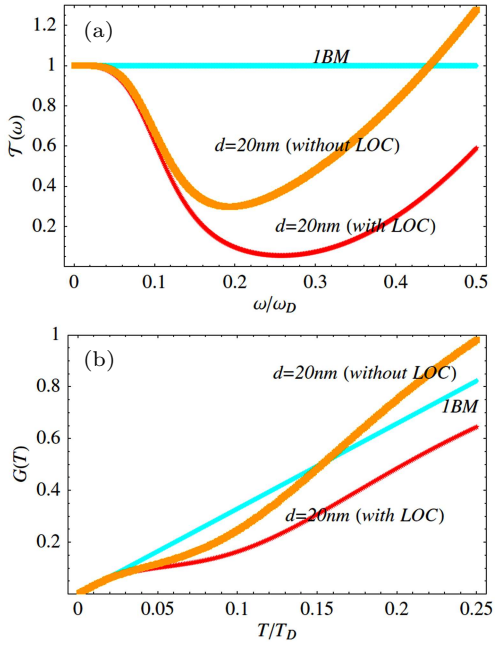


Fig. 3. (a) Frequency dependent transmission function and (b) temperature dependent thermal conductance for a surface-disorder nanowire with and without localization (LOC). "1BM" refers to one ballistic mode. Reproduced with permission from Ref. [114].

To investigate phonon localization at room temperature, Donadio *et al.* adopted the molecular dynamics simulation, lattice dynamics, and Boltzman transport equation.[115] It is found that, in SiNW with amorphous boundaries, only high frequency phonons with frequency larger than 530 cm^{-1} are localized. As the thermal conductivity of SiNW is mainly contributed by phonons with low and middle frequencies.[113,115] The localization of these high frequency phonons has marginal effect on the thermal conductivity.[115,116] The reduction of thermal conductivity for SiNW is due to the decreased relaxation time of propagating phonons and the presence of nonpropagating modes. By analyzing the polarization vectors and group velocity, it is found that phonons with frequency between 70 cm^{-1} and 530 cm^{-1} show the extended, nonpropagating characteristic. Further analyzing by Sadhu *et al.* indicates that this kind of phonons as localized modes, however, have a small spatial extension and can still propagate particlelike between scatters.[117,122] The contribution of these phonons to the thermal conductivity is small due to their ultralow group velocity. Except in nanowires, Wang *et al.* investigated phonon localization in another kind of one-dimensional structures, graphene nanoribbons.[123] It is found that the thermal conductivity of graphene nanoribbon with zigzag edges is much higher than that with armchair edges. This difference in thermal conductivity decreases as the temperature increases, and cannot be explained by the roughness scattering theory. After further analyses on the normalized cumulative thermal conductance of phonon wave length and participation ratio, Wang *et al.* indicated that this difference in thermal conductivity is not induced by edge roughness scattering, but strong phonon localization near and at the edges.

To induce phonon localization at low frequencies and effectively tune the thermal conductivity, Chen *et al.* proposed another localization mechanism.[124,125] Instead of introducing disordered boundaries, Chen *et al.* introduced crystal atomic coating on the surface of nanowire to form the core–shell structure (shown in Fig. 4(a)). Due to the structure confinement in the transverse direction, oscillations appear for transverse modes, which is similar to the standing waves. These oscillations do not propagate in the longitudinal direction, and their eigen-frequencies can be calculated through the elastic medium theory. As a result, multiple oscillation peaks can be observed in the frequency space for Ge/Si core–shell NWs. This can be demonstrated in Fig. 4(b) by that the heat current autocorrelation function (HCACF) of SiNWs decays to zero for enough long time, while the HCACF of Ge/Si core–shell NWs oscillates periodically around zero finally. As the sound velocities of Ge and Si are different, atoms near the core–shell interface are stretched, which induces a strong coupling between the transverse and longitudinal motions. Thus, a resonance forms. Due to the nonpropagating nature of the transverse modes, the coupling will lead to the localization of the longitudinal propagating modes. As shown in Fig. 4(c), the participation ratio, which measures the mode localization, for phonons in Ge/Si core–shell NWs is much lower than that of Ge NWs at low frequencies. As low frequency phonons have a large contribution to thermal transport, the thermal conductivity of Ge/Si core–shell NWs can be decreased compared to that of Ge NWs (shown in Fig. 4(d)). However, when the coating thickness increases, the thermal conductivity of Ge/Si core–shell NWs can also be higher than that of Ge NWs. This is because the surface-to-volume ratio decreases, which weakens the boundary scattering.

Except in core–shell nanostructures, the scenery of coupling between different modes has been found in natural schwarzite based host–guest systems.[126] Mode hybridization can be formed between rattler modes (the local vibrations of guest atoms) and propagating modes, leading to phonon localization.[127] It is found that the "on-center" rattling in-

duces a single hybridized mode, while the "off-center" rattling broadens frequencies for the hybridized modes. As a result, the off-center rattling can strengthen the hybridization and induce strong phonon localization. Based on phonon localization, the concept of phonon storage can be realized by the strong confinement of hybridized modes. In 2020, Zhang *et al.* demonstrated that the phonon nano-capacitor can be built by carbon schwarzite based host–guest system.[128] The storage of phonons from gigahertz to several terahertz can be achieved by changing the host–guest interaction.

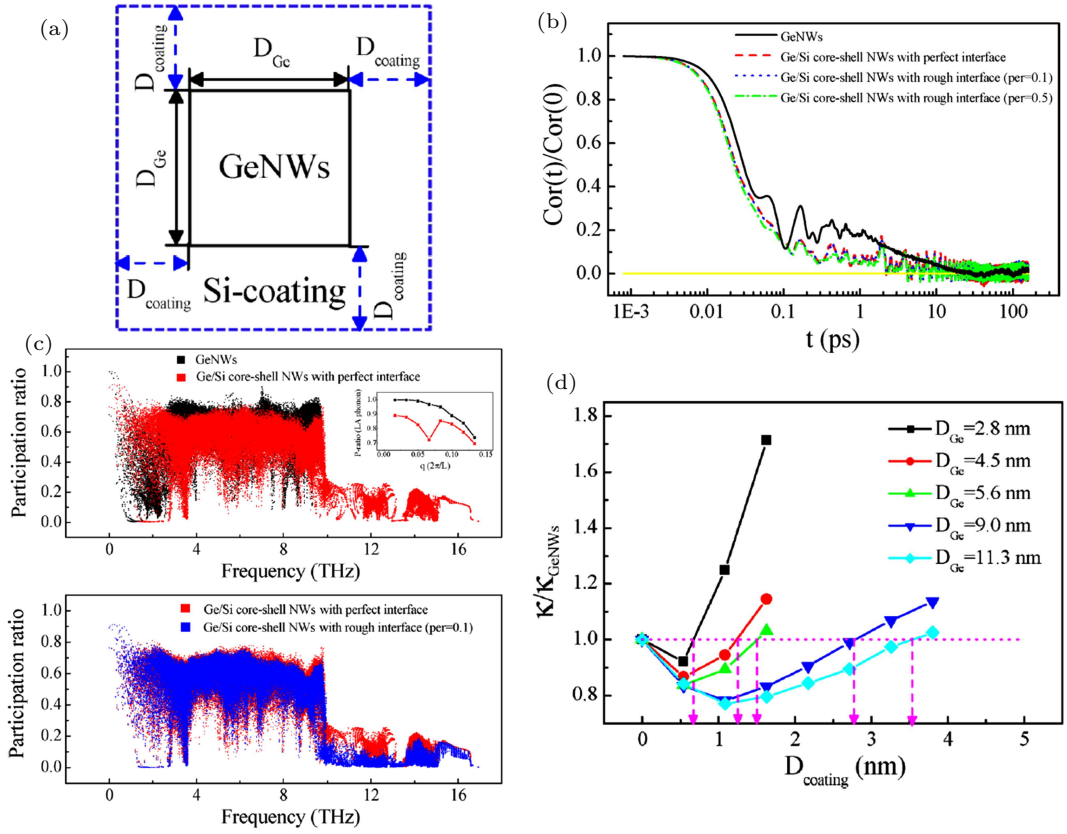


Fig. 4. (a) The schematic diagram of Ge/Si core–shell NWs. (b) The heat current autocorrelation function and (c) participation ratio of Ge/Si core–shell NWs and Ge NWs. (d) The dependence of normalized thermal conductivity on coating thickness for different side length of Ge. Reproduced with permission from Refs. [124,125].

4. Phonon localization by disorder of impurities

The researches of how current, or electron, transport in semiconductors containing impurities are actually the origin of Anderson localization.[129] Except for electrons, impurity scatterings of phonons, for example isotopic doping[84,101,130] and forming alloys,[19,131] are widely used to modulate thermal properties of materials. In 2006, through thermal bride method, Chang *et al.* found that the thermal conductivity of boron–nitride nanotube can be decreased through isotopic doping.[132] They speculated that the reduction of the thermal conductivity is due to phonon localization by isotopic scattering. To clarify this point, Mingo *et al.* investigated phonon interference effect by multiple scattering in carbon and boron–nitride nanotubes for different isotope concentrations through *ab initio* calculations.[133] It is found that thermal conductivity reduction induced by phonon localization can only be observed for isotopic concentrations larger than 10%. This is because localized high frequency phonons coexist with quasi-ballistic low frequency phonons. At low temperatures, only quasi-ballistic low frequency phonons can contribute to thermal conductivity due to Bose–Einstein distribution. High frequency phonons cannot be excited. At high temperatures, the thermal conductivity is dominated by ballistic and super-diffusive phonons, which generally have low and middle frequencies. As a result, although localized high frequency phonons are excited, their contribution to thermal conductivity is quite small.

In another line, John *et al.* suggested that photon localization would occur in the frequency window of certain disordered superlattice.[3] Inspired by this idea, Mendoza *et al.* observed phonon localization by introducing disordered impurities at the interface of superlattice.[134] Through nonequilibrium Green's function, Mendoza *et al.* investigated the dependence of thermal conductivity on the length of GaAs/AlAs superlattice with disordered ErAs impurities at the interface (shown in the inset of Fig. 5(b)). The periodic length is chosen as 5.66 nm, and the pure superlattice can be treated as a homogeneous material (as illustrated in Section 2). At 10 K, the normalized conductivity increases for both cases with 2.38% and 23.8% disorder. This tendency is in accordance with the size

effect of thermal conductivity, which is governed by the ballistic and diffusive phonon transport.[63–65] However, at 100 K, the normalized conductivity first increases and then decreases when the concentration of disorder is 23.8%. This is quite different from the case with 2.38% disorder.

For the superlattice with 2.38% impurities, the degree of disorder is unable to induce phonon localization. Thus, the dependence of the normalized conductivity obeys the traditional rules of phonon diffusive and ballistic transport. The transmission function $g(\omega) \propto L^{-1}$ and $g(\omega) \propto e^{-L/\xi}$ (ξ is the localization length) at the ballistic and localization regions, respectively. The transition of conductance from power law to exponential law at about 1.5 THz (shown in Fig. 5(a)) indicates phonon localization for the superlattice with 23.8% impurity. Since 1.5 THz is the fifth band edge, the appearing of phonon localization obeys the mechanism proposed by John *et al.*[3] However, at 10 K, the phonon population shifts to low frequency where no localization is induced, and the phonons experience ballistic transport. At 100 K, the localized modes become populated. As a result, when the system length is smaller than the localization length, the conductivity increases due to the traditional size effect, and when the system length becomes larger than the localization length, phonon localization is induced and begins to affect the conductivity. As the system length further increases, more phonons will be localized. Thus, the decrease of the thermal conductivity is due to phonon localization when the length is larger than 112 nm.

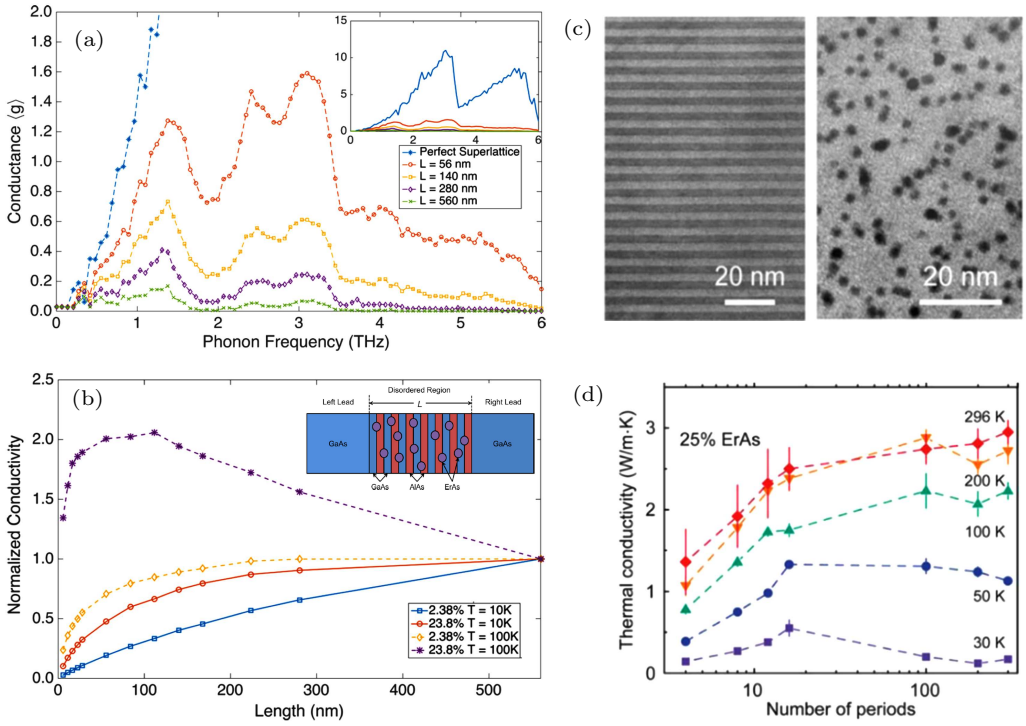


Fig. 5. (a) Frequency dependent dimensionless phonon conductance for GaAs/AlAs superlattices with 23.8% ErAs. (b) Temperature dependent normalized phonon conductivity for GaAs/AlAs superlattices with 2.38% and 23.8% ErAs at 10 K and 100 K by simulation. Inset is the schematic diagram of GaAs/AlAs superlattices with ErAs impurities at interface. (c) The transmission electron microscope image of GaAs/AlAs superlattice with disordered ErAs at interface. (d) The dependence of thermal conductivity of GaAs/AlAs superlattice with 25% ErAs on length from 30 K and 296 K by experiment. Reproduced with permission from Refs. [134,135].

Based on the above predictions by Mendoza *et al.*, Luckyanova *et al.* observed phonon localization in experiment.[135] Luckyanova *et al.* fabricated the GaAs/AlAs superlattice with periodic length of 6 nm (shown in Fig. 5(c)) which is similar to that in the simulation by Mendoza *et al.*[79,134] ErAs dots with concentration of 25% are randomly introduced at the interface (shown in Fig. 5(c)). By the time-domain thermoreflectance method, the decrease of thermal conductivity is found when increasing the length of the system at 30 K and 50 K (shown in Fig. 5(d)). At room temperature, this trendency is not observed. This is because on one hand, the thermal conductivity is contributed by a broader range of phonons. On the other hand, high frequency phonons experience phonon–phonon scattering, which results in the break of phase information.

5. Phonon localization by disorder of secondary artificial periodicity in phononic crystal

PnC, analogous to photonic crystal, has been widely used to control phonon transport via coherent interference effect.[21,55,84,101] Generally, the PnC is formed by introducing artificial secondary periodicity to the original structure. Doping to form superlattice[84,101] and introducing periodic holes[21,85] are two commonly used strategies. In PnC, the secondary artificial periodicity will fold the band structures,

leading to the open of band gaps which decreases phonon group velocity and changes phonon density of states.[55,78] This mechanism, called the band-folding effect, can lead to huge decrease in thermal conductivity of PnC, which can be more effective than the diffusive scattering by roughness boundaries.[21,85] Due to this benefit, PnC has been considered as a good candidate for thermoelectrics where low thermal conductivity can enhance the figure of merit.[14,77] Thus, extensive efforts have been made to investigate PnC, among them, factors affecting the thermal conductivity of PnC, including disordered degree of secondary artificial periodicity, attract lots of attention.

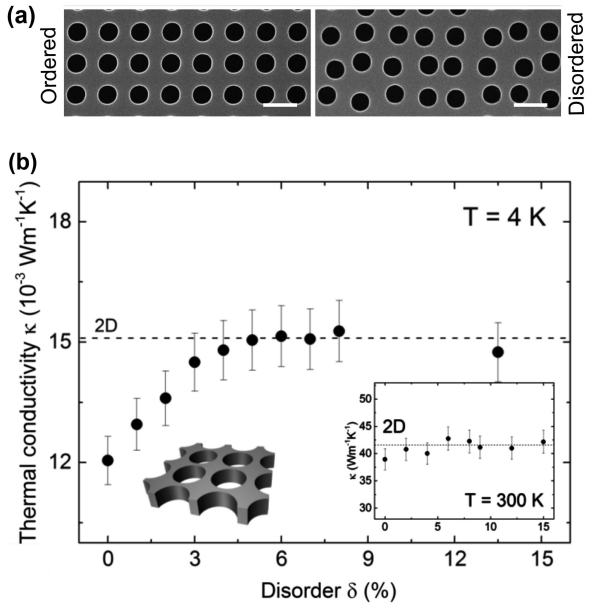


Fig. 6. (a) The transmission electron microscope image of ordered (periodic) and disordered silicon phononic crystal. (b) The disorder dependent thermal conductivity of silicon phononic crystal at 4 K and 300 K. Reproduced with permission from Ref. [136].

As the band-folding effect strongly relies on the periodicity of structure,[78] disorder of secondary artificial periodicity (aperiodicity) has been regarded as the perturbation which will destroy the coherence of phonons and weaken this effect.[79,136] Wagner *et al.* investigated this problem in two-dimensional PnC with holes through time-domain thermoreflectance method.[137] It is found that the introduction of disorder by randomizing the position of holes can hugely modify the hypersonic phonon spectrum. However, the change in thermal conductivity was not observed at room temperature. This may be due to that a broad range of phonons contribute to the thermal conductivity. The change of phonons in hypersonic frequencies due to the disorder of periodic holes contributes little to the thermal conductivity. A more systematic study was performed by Maire *et al.* They focused on one-dimensional and two-dimensional PnCs at both room temperature (300 K) and low temperature (4 K).[136] The results are shown in Fig. 6. With the disorder of the secondary artificial periodicity increases, the thermal conductivity first increases, and then saturates at 4 K. However, at 300 K, the thermal conductivity is insensitive to the change of disorder, which is in accordance with the results of Wagner *et al.*.[137] This is because at low temperature, only low frequency phonons contribute to thermal conductivity due to Bose–Einstein distribution. Thus, the change of phonons in hypersonic frequencies can have observable effects on the thermal conductivity. However, at room temperature, this change is marginal. All in all, the disorder of the secondary artificial periodicity can destroy the band gaps induced by folding, and allow more phonons to take part in thermal transport. The similar results are also observed in the one-dimensional PnC.

However, as suggested by John *et al.*, localization would occur in the frequency gap of certain disordered phononic crystal.[3] If so, the disorder of the secondary artificial periodicity in PnC would decrease its thermal conductivity due to phonon localization. In 2006, this kind of result was presented by Wang *et al.* on anharmonic lattice that subjected to ϕ^4 on-site potential.[138] The secondary artificial periodicity is introduced by changing the on-site potential of selected lattices. When the on-site potential is periodically changed, the system can be treated as PnC. It is found that the thermal conductivity of the system whose on-site potential is randomly changed is lower than that whose on-site potential is periodically changed. Later, Xiulin *et al.* presented similar results in the three-dimensional conceptual Lennard-Jones PnC (superlattice).[139] From 30 K to 150 K, the thermal conductivity of random multilayer is much smaller than that of periodic superlattice due to phonon localization. Juntunen *et al.* extended the conceptual system to real materials, and investigated the same topic in Si/Ge superlattice (shown in Fig. 7(a)) through molecular dynamics simulation at room temperature.[140] To disorder the secondary artificial periodicity, the layer thicknesses of certain atomic layers are randomly sampled. During this process, the layer number and total thickness of Si and Ge are all keep the same. Thus, no extra interfaces are introduced. As shown in Fig. 7(b), the thermal conductivity of the periodic superlattice increases as the system size increases, which indicates the ballistic and diffusive transportation of phonons. However, by disordering the secondary artificial periodicity, the thermal conductivity of the aperiodic superlattice increases first, and then decreases as the system size increases. The non-monotonic change is due to the transition from ballistic transport to localization, which is in accordance with the results of Mendoza *et al.*[134] through introducing disorder impurities at the interfaces. When the system size is small, the thermal conductivities of the aperiodic and periodic Si/Ge superlattices are the same due to the ballistic phonon transport. However, the thermal conductivity of the aperiodic Si/Ge superlattice is much smaller than that

of the periodic Si/Ge superlattice as the system size increases, only 2% of that of the periodic Si/Ge superlattice when the system size is larger than ∼ 250 nm. This is because the localization lengths of most phonons are less than 250 nm. When the system size is larger than 250 nm, no more phonons can be localized. That is to say, two-orders thermal conductivity reduction can be achieved by phonon localization.

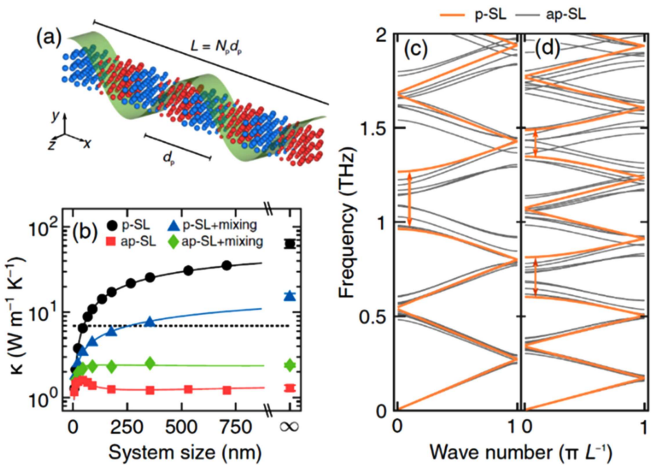


Fig. 7. (a) The schematic diagram of Si/Ge superlattice. (b) The length dependent thermal conductivity of periodic and aperiodic (disorder) superlattice at room temperature. Phonon dispersions of (c) longitudinal and (d) transverse modes for periodic and aperiodic (disorder) superlattice. Reproduced with permission from Ref. [140].

The spectral features of phonon localization have also been explicitly elucidated by Juntunen *et al.* via lattice dynamics,[140] which is shown in Figs. 7(c) and 7(d). For periodic superlattice, obvious band gaps are presented through band folding for both longitudinal and transverse modes. In this case, dispersions near the gaps flatten out, which leads to the reduction of the thermal conductivity.[55] After introducing disorder of secondary artificial periodicity, many milibands are formed and the phonon group velocity can be further reduced at the newly formed Brillouin zone edges.

Except in superlattice, disordering periodic holes in PnC (shown in Fig. 8(a)) can also induce strong phonon localization, and result in huge reduction of thermal conductivity.[141,142] This has been demonstrated in both graphene PnC[141] and C_3N PnC[142] through molecular dynamics simulation by Hu *et al.* To better clarify phonon localization, adopting a two-phonon model developed by Wang *et al.*,[139] the thermal conductivities of graphene PnC with ordered and disordered holes are decomposed to contributions from incoherent and coherent phonons. As shown in Fig. 8(b), when the system length is shorter than 10 nm, thermal conductivities of ordered and disordered graphene PnC are the same for molecular dynamics simulation and Green's function method as a result of ballistic transport. For ordered PnC, coherent phonons contribute a large proportion (more than 50%) to the total thermal conductivity. However, in disordered PnC, coherent phonons are strongly localized, their contributions to the total thermal conductivity decrease rapidly. When the system length is larger than 200 nm, the contributions of coherent phonons are negligible because they are fully localized. For incoherent phonons, the absolute values of their contributions to the thermal conductivity do not change for ordered and disordered PnC. This is because incoherent scattering does not rely on periodicity. What is more, the appearing of the maximum thermal conductivity when the system length increases is only contributed by coherent phonons (shown in the inset of Fig. 8(b)). This further demonstrates the phonon localization mechanisms here. The similar results have also been discovered in the conceptual Lennard–Jones superlattice by Wang *et al.*[139]

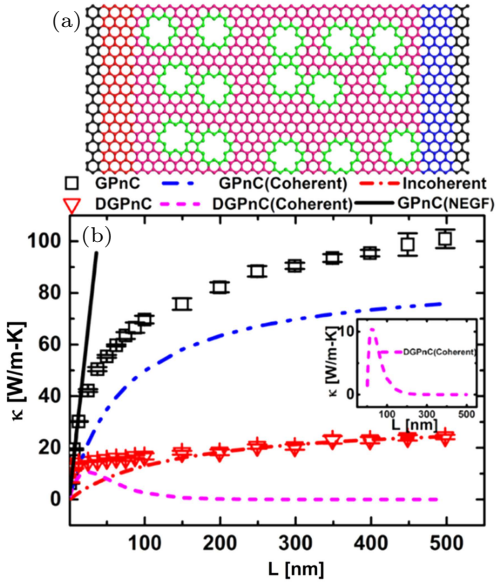


Fig. 8. (a) The schematic diagram of disordered graphene phononic crystal. (b) Decomposition of the total thermal conductivity of periodic and disorder graphene phononic crystal to the contributions of coherent and incoherent phonons at different length. Reproduced with permission from Ref. [141].

To validate this thermal conductivity reduction by phonon localization, Shiomi *et al.* investigated thermal conductivities of periodic and aperiodic GaAs/AlAs superlattices through both machine learning and time-domain thermoreflectance method from 77 K to 300 K. At first, the thermal conductivity of each structure is calculated through the Green's function. Then, Bayesian optimization[143,144] is used to predicted the structure that has the minimum thermal conductivity with both smooth and rough interfaces. Finally, the samples of the predicted structure are fabricated, and the thermal conductivities are measured. GaAs and AlAs are denoted as 0 and 1, respectively. The optimal structure is 1001010101101101, which is obviously an aperiodic superlattice. As shown in Fig. 9, the thermal conductivity of the optimized aperiodic superlattice is quite lower than that of periodic superlattice for both smooth and rough interfaces from 77 K to 300 K. The experiment re-

sults for cases with roughness interfaces agree well with results from the Green's function. This confirms the reduction of thermal conductivity by disordering secondary artificial periodicity. Through mapping the localized density of states (LDOS) for single frequency to each layers, it can be found that the LDOS are relatively uniform for the periodic structure. However, in the disordered structure, large LDOS can be found in AlAs layers, indicating the localization of phonons.

It needs to be noticed that a seeming discrepancy arises between the results of Maire *et al.*[136] and others.[139–141,145] The results of Maire *et al.* predict that thermal conductivity will increase after disordering the secondary artificial periodicity of PnC, while, the results by others show a tendency of decrease. This discrepancy can be well solved by machine learning method.[144] Different researches have been performed to investigate phonon localization by disordering the secondary artificial periodicity of PnC through machine learning method.[143,146–149] Most of them focused on maximizing phonon localization, and exploring minimum thermal conductivity[143,146–148] for thermoelectric application. The increase of thermal conductivity by disorders has been clarified by Wei *et al.*[149] Through machine learning searching, it is found that the thermal conductivity of disordered PnC can be both lower and higher than that of periodic PnC. The enhancement of phonon transport is due to the particular pore arrangements that facilitate the transport of incoherent phonons. Wei *et al.* proposed that two factors, the shape factor and channel factor which describe the pore arrangement parallel and perpendicular to the heat flux direction, respectively, can affect the thermal conductivity. The structure that has a more spread pore arrangement perpendicular to the heat flux direction will have a smaller channel and larger thermal conductivity.

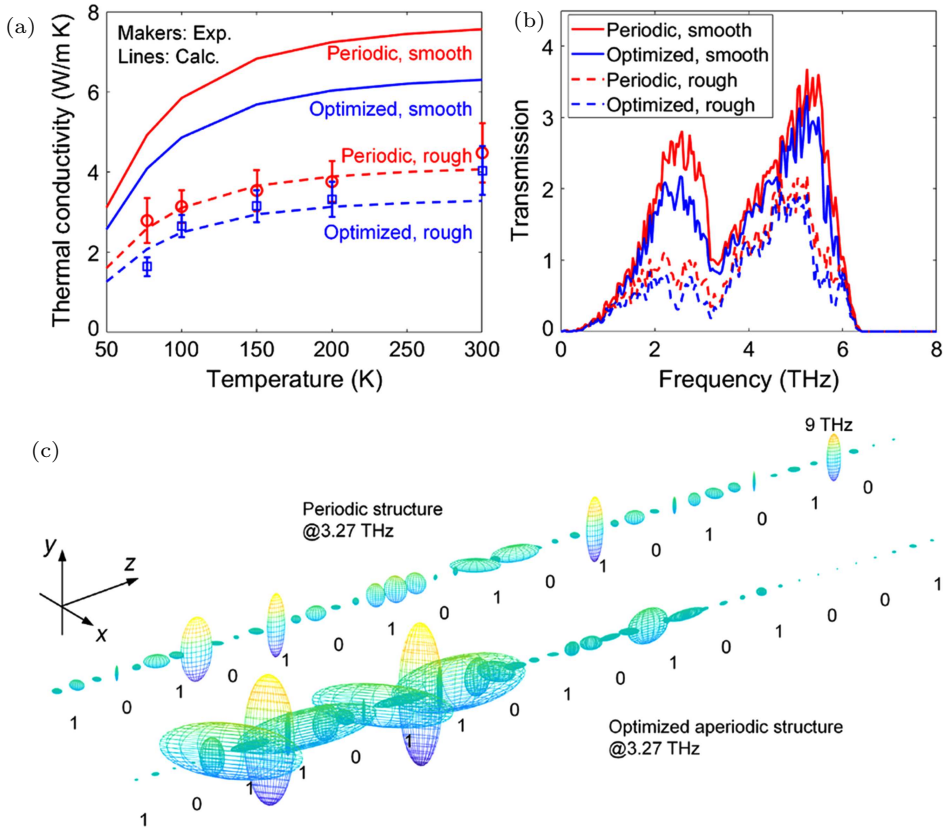


Fig. 9. (a) Thermal conductivity and (b) phonon transmission function for periodic and optimized aperiodic GaAs/AlAs superlattice at different roughness. (c) Phonon localization (LDOS) of periodic and aperiodc GaAs/AlAs superlattice at 3.27 THz. Reproduced with permission from Ref. [145].

6. Conclusion and perspectives

In this review, recent progresses on phonon localization and its effect on thermal conductivity of nanostructures are summarized. By changing the surface morphology, introducing disordered impurities, and disordering the secondary artificial periodicity in phononic crystal, localized modes can be presented. However, phonon localization induced by surface disorder can only reduce the thermal conductivity of nanowire at low temperature. Only with high concentration, phonon localization induced by disordered impurities can have an observable effect on thermal conductivity at room temperature. By disordering the secondary artificial periodicity in phononic crystal, the thermal conductivity can be heavily reduced, even up to two orders. Overall, localization can hinder the transport of phonons in certain frequencies. Due to the fact that phonons in a broad frequency range from about 0.1 THz to tens of THz can contribute to thermal conductivity, thermal conductivity reduction induced by phonon localization can be

observed only when a large number of phonons are localized, especially at low and middle frequencies. Actually, this is one of the challenges in utilizing phonon localization to tailor thermal conductivity. Except that, the phonon coherence length is quite small, which is no more than 10 nm for most phonons at room temperature. To utilizing coherent nature of phonons, the characteristic length of nanostructure should be less than the wavelength. This brings great difficulties for materials synthesizing, and thus, limits its practical application.

Considering the present successes and challenges, we conclude some further directions which need to be investigated. Firstly, the decrease of thermal conductivity by phonon localization implies its potential application in thermoeletrics. Except thermal conductivity, electric properties are also important factors to be considered. However, till now, few researches about the electric properties on this spatial size and structure features have been reported. Multifunctional optimization by changing structure sizes and degree of disorder to hugely reduce the thermal conductivity and silently affect the electric properties is quite important. Secondly, unlike photons and acoustic waves, which are essentially noninteracting, high order phonon scattering will destroy the phase information of phonons. Thus, how anharmonicity or nonlinearity affects phonon localization and the spectral features need to be systematic clarified theoretically. Thirdly, most of present researches are theoretical works, experiment observations and researches of phonon localization are expected. Especially, the reduction of thermal conductivity by phonon localization is just demonstrated by simulation, which is still under debate. Convincing experiment results by isolating the effect of phonon localization and other mechanisms, for example diffusive scattering, on thermal conductivity are needed.[150] At the same time, to better understand the wave nature and localization of phonons, exploring strategies to directly probe phonon coherence length as that of phonon mean free path is of great importance.

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