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Phonon polariton-mediated heat conduction: Perspectives from recent progress

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It has been well-accepted that heat conduction in solids is mainly mediated by electrons and phonons. Recently, there has been a strong emerging interest in the contribution of various polaritons, quasi-particles resulting from the coupling between electromagnetic waves and different excitations in solids, to heat conduction. Traditionally, the polaritonic effect on conduction has been largely neglected because of the low number density of polaritons. However, it has been recently predicted and experimentally confirmed that polaritons could play significant roles in heat conduction in polar nanostructures. Since the transport characteristics of polaritons are very different from those of electrons and phonons, polariton-mediated heat conduction provides new opportunities for manipulating heat flow in solid-state devices for more efficient heat dissipation or energy conversion. In view of the rapid growth of polariton-mediated heat conduction, especially by phonon polaritons, here we review the recent progress in this field and provide perspectives for challenges and opportunities.

Polaritons are quasi-particles resulting from strong coupling between electromagnetic waves and excitations in matter that carry electric or magnetic dipoles [1]. Many different types of polaritons have been identified and studied in the general field of light-matter interactions. Transport of these quasi-particles carries energy with them, which can contribute to thermal transport. However, the contribution of polaritons to heat conduction has been largely neglected because of the much lower number density of polaritons as compared to those of electrons and phonons. Recently, there has been rapidly growing interest in the contributions of two of the most investigated polaritons, phonon polaritons (PhPs) and surface plasmon polaritons (SPPs), to heat conduction. Pioneering theoretical and experimental studies have shown promising results with highly intriguing observations that deserve further exploration.

So far, most studies of polariton-mediated heat conduction have focused on the implementation of PhPs, quasi-particles resulting from coupling between electromagnetic waves (photons) and polar optical phonons. When long wavelength (far- to mid-infrared) photons propagate in polar solids, they modulate and interact with the vibrations of the polar lattice within the unit cells [Fig. 1(a)]. This coupling generates new modes that result from the hybridization between photons and phonons, which are called phonon polaritons. These hybrid modes are closely related to SPPs [2], which are derived from a strong coupling between photons and coherent free electron oscillations in conductors, and exciton polaritons [3], resulting from the coupling between photons and electron–hole pairs. In all forms, these polaritonic modes offer the ability to compress the free-space wavelength and serve as a key base for infrared nanophotonics.

The properties of PhPs are well-described by their dispersion relation as shown in Fig. 1(b) [4]. In the bulk of polar materials, PhPs can exist below the transverse optical (TO) phonon frequency and above the longitudinal optical (LO) phonon frequency, as shown by the two curved black lines in the dispersion. Within the field of nanophotonics, polaritons on these two dispersion lines are often referred to as 'bulk' PhPs due to their extension into the volume of the polaritonic medium. Yet, between the TO and LO phonon frequencies, the real part of the complex permittivity tensor of polar materials becomes negative and they exhibit high reflectivity to infrared photons within this regime. As a result, propagation of infrared photons in this frequency range (named Reststrahlen band) in the bulk of

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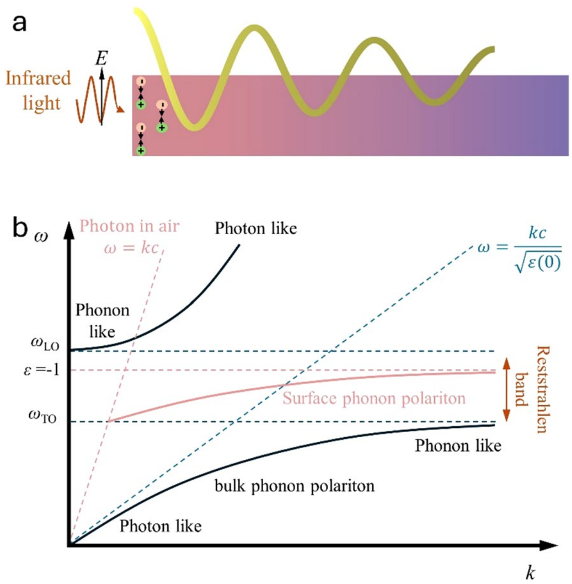

Figure 1: Basic properties of phonon polaritons. (a) Polariton formation at the polar solid surface resulting from the interaction between polar lattice vibrations and infrared photons. (b) Phonon polariton dispersions as compared to the photon dispersion.

polar materials is prohibited. However, within the Reststrahlen band, the formation of sub-diffractional PhPs can be realized at the surface of polar materials or their interfaces with dielectric materials, which are called surface phonon polaritons (SPhPs). These SPhPs are highly confined to the surface or interface with their electric field intensity attenuating strongly following an exponential decay along the normal direction of the surface/interface, which is a widely known behavior of evanescent waves. It is important to note that due to the evanescent nature of these SPhP modes, they can offer dramatic compressions of the wavelength, yet this shrinking of the polariton wavelength (i.e., increasing the polariton wavevector) requires overcoming a significant momentum mismatch with free-space light. This can be accomplished via scattering, nanostructuring of the material, using a high index prism, or diffraction grating [5].

The existence of PhPs was first predicted by Huang in 1951 [6] and they were experimentally observed in 1965 [7]. SPhPs were initially studied as non-radiative modes existing along material surfaces [8]. For thermal sciences, SPhPs promise to play an important role in radiative heat transfer [9–13]. In fact, for near-field radiation, when the distance between two neighboring surfaces becomes close enough to allow for coupling between the evanescent fields of SPhPs on the two surfaces, the radiative thermal conductance between these two surfaces can be enhanced by several orders of magnitude. This enhanced near-field radiation, as a result of the remarkably enhanced SPhP density of states in the Reststrahlen band, has a strong potential for thermal photovoltaics [14] and radiative cooling [15, 16].

Inspired by the important SPhP contribution in near-field radiation, Chen et al. [17] first predicted in 2005 that transport of SPhPs along thin SiO_2 films could enhance their thermal conductivities by several-fold [Fig. 2(a)] and identified this as the only known approach to counteract the classical size effect that leads to reduced lattice thermal conductivity in thin films. Two distinct transport characteristics render SPhPs particularly attractive as energy carriers for heat conduction. First, due to the optical phonon and light origin, these modes are of higher energy as compared to acoustic phonons, while the photon nature imparts a group velocity to SPhPs that could be orders of magnitude higher than that of acoustic phonons. In addition, the SPhP decay length can be very long [18], up to millimeters or even centimeters, which are also orders of magnitude larger than the phonon mean free path (from a few nm up to ~ 10 μm) [19]. These distinct features provide the potential for effective heat conduction along thin films or nanowires so long as the excited SPhPs reach a small fraction of the number density of phonons.

In Chen et al.'s study [17], the contributions of anti-symmetric and symmetric modes were discussed and it was suggested that it is the light-line portion of the anti-symmetric modes that make the strongest PhP contribution to the thermal conductivity of thin SiO_2 films. This is mainly due to the large group velocity and long decay length of these anti-symmetric modes. Importantly, Chen et al. pointed out in this seminal study that the reliance on the long decay length for significant PhP-mediated heat conduction renders the effect only noticeable over a long distance on the order of centimeter scale.

The pioneering work by Chen et al. stimulated many follow-up theoretical studies. For example, Ordonez-Miranda et al. [20] predicted that adjusting the permittivity values of the substrate and superstrate of a SiO_2 thin film could effectively extend the SPhP propagation length, to tens of millimeters, which could lead to a SPhP-mediated thermal conductivity of 2.5 W/m·K for a 30 nm thick SiO_2 film, ~ 1.8 times that of the phonon-mediated value for bulk SiO_2. More recently, through theoretical calculations, Li and Shin [21] further suggested that through tuning the permittivity of the surrounding media of 50 nm thick SiO_2 films to push most of the SPhP energy field out of the SiO_2 surface, the propagation/decay length could be extended beyond one meter long, which could lead to an anomalously high PhP thermal conductivity of 248 W/m·K [Fig. 2(b)]. While these predictions are exciting, again these impressive values could only be achieved with ultralong sample lengths, which put severe limitations to the demonstration and real-world applications of these theoretical predictions.

In addition to amorphous SiO_2 thin films, Ordonez-Miranda et al. [26] modeled the SPhP contribution to thermal conductivity of quartz, in which they gave the relationship between the SPhP-mediated thermal conductivity and the film thickness. High thermal conductivity on the order of 100 W/m·K was

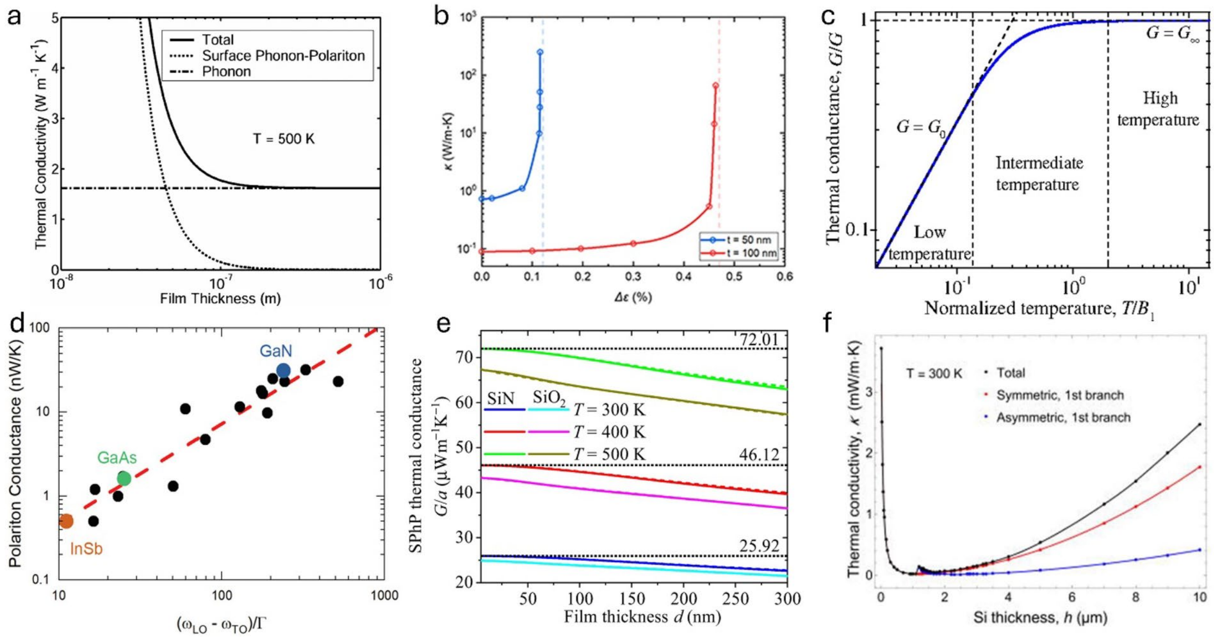

Figure 2: Theoretical results on PhP-mediated heat conduction. (a) Thermal conductivity of amorphous SiO_2 thin films due to phonons and surface phonon polaritons as a function of film thickness at 500 K [17]. (b) The thermal conductivity of SiO_2 thin films medicated by SPhPs with varied Δε and the thin film thickness [21]. (c) Thermal conductance vs. temperature for a nanowire with a single transmitted SPhP mode [22]. (d) PhP-mediated conductance vs the Figure of Merit for each of the examined materials [23]. (e) SPhP thermal conductance per unit width of 1-mm-long SiN_x and SiO_2 thin films as a function of their thickness [24]. The solid and dashed lines represent the respective analytical and numerical predictions, while the dotted ones stand for the quantum of thermal conductance. (f) Si thickness dependence of the in-plane SPhP thermal conductivity for the SiO_2/Si/SiO_2 sandwich structure [25]. Figures reproduced from Refs. 17, 21–25 with permission. Copyright American Physical Society, AIP Publishing.

predicted when the length scale extends to > 2 m. In addition, a recent report [27] calculated SPhP-mediated thermal conductivity of $SrTiO_3$ thin films and suggested that for 100 nm thick films at 300 K, the polaritonic thermal conductivity could reach 18.60 W/m·K, which represents over twice of the lattice thermal conductivity mediated by phonons. Beyond thin films, it has been suggested that in super crystals composed of periodically packed spheroidal nanoparticles of SiC, the SPhP contribution to the thermal conductivity could surpass that of phonons [28], which is attributed to the ultra-high surface-area-to-volume ratio in these structures. Moreover, it was proposed that the SPhP thermal conductance along a polar nanowire was independent of the material characteristics and quantized [Fig. 2(c)] [22]. Interestingly, the authors argued that the quantized transport was not limited to ultralow temperature (< < 1 K) as is the case for electrons and phonons, but could even extend to room temperature, which could facilitate its observation and applications.

The above-mentioned pioneering theoretical studies have inspired additional modeling efforts. For example, the Boltzmann transport equation was used to solve for SPhP-mediated thermal conductivity along SiO_2 films of different thicknesses and lengths, and the results indicate a more pronounced SPhP contribution in thinner and longer films [29]. Minyard and Beechem [23] recently compared the SPhP-mediated thermal conductance for InSb, GaAs, and GaN thin films. Based on their calculations, it was suggested that in addition to longer SPhP lifetimes or decay lengths, higher optical phonon energies and wider Reststrahlen bands would help boost the SPhP contribution to the film thermal conductance. The polariton thermal conductance is shown to be correlated with a figure of merit defined as $(\omega_{LO} - \omega_{TO})/\Gamma$, where Γ is a parameter related to the polariton decay length [Fig. 2(d)]. Interestingly, another recent theoretical study [24] suggested that for sufficiently thin films, the thermal conductance per unit width mediated by SPhPs is independent of the material properties [Fig. 2(e)] and given by $12z(3)k_B^3T^2/ch^2$, where k_B and h are the Boltzmann and Planck constants, respectively, while c is the free-space speed of light, T is the temperature, and $z(3)$ is the Riemann zeta function. Clearly, it would be interesting to explore whether the material characteristic-dependent SPhP thermal conductance would converge as the film thickness is reduced, and if so, what is the critical thickness for this to occur.

In addition to SPhP-mediated thermal transport along free-standing polar thin films, studies have been performed on multilayer films such as those made of SiO_2/Si/SiO_2 sandwich structures [25]. It was suggested that as the Si layer thickness transitions from the sub-micron regime to beyond 1 μm regime, the electromagnetic field intensity shifts from the free-space to the Si layer. As a result, the SPhP-mediated thermal conductivity of the SiO_2 layers reaches a minimum value at 1 μm Si spacer layer thickness [Fig. 2(f)]. Studies of PhP-mediated thermal transport in multilayer structures are important as most solid-state devices are composed of multiple layers of different films.

Experimental studies to explore the role of SPhPs in the thermal conductivity of polar nanostructures have been made only in the past decade. Tervo et al. reported the first experimental attempt towards observing SPhP-mediated thermal transport in packed SiO_2 nanoparticle beds [30]. It was suggested that when a sub-nm thick water or ethylene glycol coating exists on the nanoparticle surfaces, the measured thermal conductivity is significantly higher than the corresponding value for bare particle beds and the relevant liquids [Fig. 3(a)]. The authors proposed that this enhancement could be due to the contribution of SPhPs. While the experimental observation is intriguing, the attribution to SPhP-mediated thermal transport was not conclusive as that material system is extremely complex with many factors that could affect heat conduction. Importantly, analysis with conventional SPhP-mediated conduction models could not explain the experimental results. Moreover, the new model proposed by the authors, which yielded better fitting to the experimental data, is not fully consistent with the material system.

Tranchant et al. reported an endeavor [31] to characterize the SPhP contribution to the thermal conductivity of SiO_2 films of 20 to 300 nm thick and a few mm long. The results show that the thermal conductivity increases as the film becomes thinner, which was attributed to the more pronounced effect of SPhPs for thinner films. However, the measurement results carry large uncertainties, and were suggested [32] as not fully conclusive because of the presence of the large error bars.

Wu et al. [32] measured the thermal conductivity of different thickness SiN_x thin films and compared the obtained data with the modeled SPhP-mediated thermal conductivity. It was shown that for a 30 nm thick film, the SPhP contribution dominated the thermal conductivity [Fig. 3(b)]. Later, in a separate study [33], the same group of authors measured SiN_x films at two lengths of 100 μm and 200 μm, respectively. Their results showed that the thermal conductivity of the 200 μm long film was up to 10% higher than the corresponding value for the 100 μm long film, which was attributed to the ultra-long SPhP decay length. While these two publications provide interesting data that could result from SPhP contributions to heat conduction along polar thin films, the measurement results are not fully consistent with

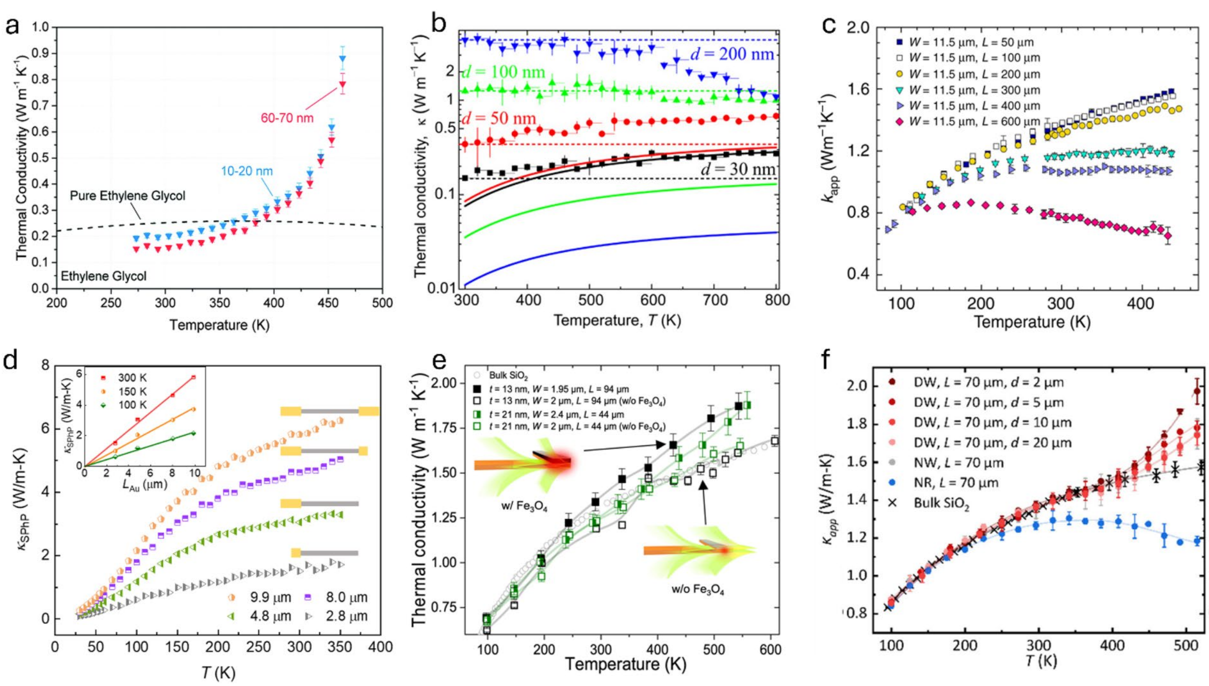

Figure 3: Experimental results on PhP-mediated heat conduction. (a) Thermal conductivity of ethylene glycol coated SiO_2 nanoparticles 10–20 nm (blue inverted triangles) and 60–70 nm (red inverted triangles) in diameter as a function of increasing temperature. The magnitude and temperature dependence have been attributed to contributions from SPhPs [30]. (b) In-plane thermal conductivity of SiN_x thin films (dots) and the modelled SPhP thermal conductivities (solid lines) [32]. (c) Measured apparent thermal conductivity (κ_{app}) vs. temperature for SiO_2 nanoribbon [34]. The reduced κ_{app} for longer ribbon samples is attributed partially to the radiation heat loss mediated by SPhPs. (d) SPhP thermal conductivity (κ_{SPhP}) vs. temperature for SiC nanowires with a short Au-coating at the end of the nanowire [35]. The inset shows the dependence of κ_{SPhP} on the length of the Au-coating. (e) Comparison of thermal conductivity of SiO_2 nanoribbons with and without the Fe_3O_4 absorber to promote the coupling between SPhPs and the thermal reservoirs/temperature sensors [36]. (f) Temperature dependent κ_{app} of SiO_2 samples with 70 μm suspended lengths [37]. SPhP contributions can be detected when the sample is aligned vertically as a nanowall (NW) but cannot when the sample is aligned horizontally as a nanoribbon (NR) because of the different coupling strengths with the thermal reservoirs. Figures reproduced from Refs. 30,32,34–37 with permission. Copyright Royal Society of Chemistry, AAAS, Nature, American Chemical Society.

each other, which requires further studies with more controlled experiments to validate.

An interesting and more recent report by Shin et al. [34] demonstrated that the measured thermal conductivity of SiO_2 nanoribbons is reduced as the ribbon length extends beyond 100 µm [Fig. 3(c)]. Through extensive modeling, the authors suggested that the lower measured thermal conductivity for longer ribbons is due to far-field radiation loss from scattering of near-field SPhPs propagating along the film. While this study presents an interesting observation, the explanation of the result relies on modeling of the radiation loss and how SPhPs influence the effective emissivity of the nanoribbons.

One common challenge in all above-mentioned experimental studies is that only the total thermal conductivity with contributions from both phonons and SPhPs is measured. Theoretical modeling has to be used to calculate the phonon- or polariton-mediated contribution to the thermal conductivity to extract the PhP-mediated value. More recently, three experimental studies [35–37] have been reported in which contrast between the results with and without PhP contributions have been shown, allowing for more direct extraction of the PhP-mediated thermal conductivity.

In a recent report on the PhP-mediated thermal conductivity of polar 3C-SiC nanowires [35], it has been shown that a short segment (a few microns long) of Au-coating at the end of the SiC nanowire could enhance the thermal conductivity of the uncoated wire segment from the corresponding value measured without the Au-coating. Here the role of the Au-coating is to thermally launch PhPs to propagate along the uncoated SiC nanowire. This contrast between the case with and without Au-coating provided direct experimental evidence for SPhP-mediated heat conduction along the SiC nanowire. More interestingly, the experimentally derived pre-decay thermal conductance is two to three orders of magnitude higher than the Landauer limit calculated based on ballistic transport of fully excited PhPs of equilibrium number density. To explain this intriguing result, the authors hypothesized that the Au-coating effectively launched and guided the PhPs excited along the entire Au-SiC interface to propagate along the uncoated wire segment. This hypothesis is strongly supported by the experimental observation that the PhP-mediated thermal conductivity is proportional to the Au-coating length [Fig. 3(d)]. Further, it was verified for these SiC nanowires that the largest thermal conductivity enhancement is correlated with the longest SPhP propagation as measured via scattering-type scanning near-field optical microscopy (s-SNOM).

A study by Pei et al. [36] that is concurrent with Ref. [35] reported on how a layer of black oxide absorber made of Fe_3O_4 would alter the measured SPhP thermal conductivity along SiO_2 nanoribbons. Without the black oxide absorber, essentially no SPhP contribution can be discerned because of the poor coupling of SPhPs with the heat source and sink. On the other hand, adding a layer of Fe_3O_4 on top of the SiO_2 nanoribbon in the heat source/sink would lead to observable SPhP enhancements to the thermal conductivity [Fig. 3(e)]. This clear contrast between the measurements with and without the absorber also provided direct experimental evidence of SPhP-mediated heat conduction along SiO_2 nanoribbons. Importantly, Pei et al. also demonstrated the ballistic transport nature of SPhPs up to ~ 100 µm.

Another recent study [37] also provided experimental contrast between the cases that SPhPs either contribute or not to the measured thermal conductivity of polar nanostructures. Through tuning the alignment of the SiO_2 nanoribbon with respect to the thermal reservoirs, the coupling strength between the SPhPs with the thermal reservoirs is effectively modulated. Results show that strong coupling between SPhPs and the thermal reservoirs leads to observable SPhP contribution to the thermal conductivity, while weak coupling cannot yield any observable SPhP-mediated thermal conductivity [Fig. 3(f)]. Again, this contrast provides direct experimental evidence for SPhP-mediated heat conduction.

Finally, a study from the Hopkins group [38] has demonstrated that hyperbolic PhPs in the highly anisotropic van der Waals crystal hexagonal boron nitride (hBN) could provide a mechanism for ultrafast interfacial thermal conduction between gold and hBN. In those experiments, an advanced form of time-domain thermal reflectance (TDTR) was employed whereby an ultrafast green (530 nm) pulse was employed to transiently heat the gold pad. Following this excitation pulse, the change in the thermal reflectance of the hBN was monitored as a function of time and probing frequency. It was determined that the TDTR response exhibited large changes within the Reststrahlen band of hBN at sub-10 ps timescales. As the hBN is transparent to the green pulse, this infers the ultrafast thermal transport across the interface between a metal and non-metal, which is typically restricted to timescales on the order of 10 s to 100 s of ps. Thus, this provides significant further evidence for the role of Au near-field radiation as a mechanism for overcoming the momentum mismatch between free-space light and PhPs, as well as the large potential for these hybrid light-matter modes for thermal conductance.

Polariton-mediated heat conduction is still in its infancy, which is especially true with only a handful of publications in experimental studies. A lot of research is still required to provide urgently needed insights into the governing mechanisms of polaritonic heat conduction. Based on the recent research progress, here we provide some perspectives on how the following research topics can move the field forward.

First, while there have been a few theoretical studies indicating that the permittivity of the conduction channel and its surrounding media could have important effects on PhP-mediated heat conduction, no experimental results have been reported to confirm these theoretical predictions. So far, all experimental studies of PhP-mediated heat conduction have been based on measurements of the thermal conductivity of individual thin films/nanowires suspended in vacuum. Considering that most engineering applications that take advantage of polaritonic heat conduction for fast heat dissipation would likely involve polariton transport along the interfaces between multiple layers of thin films or nanowires embedded in certain media, the study of this research topic is of critical significance for both fundamental understanding and practical applications.

One critical issue for PhP-mediated heat conduction is how to effectively launch PhPs to propagate along the conduction channel. For example, the recent publication [35] demonstrating the critical role of a short segment of Au-coating at the end of SiC nanowires poses the intriguing question of how PhPs with high number densities can be thermally stimulated and emitted into the bare SiC nanowire from the Au-coating. A rigorous theoretical analysis based on fundamental theories would provide key insight into the PhP launching mechanisms and effective means of tuning PhP-mediated heat conduction.

The scattering mechanisms and ballistic PhP transport limit are topics of great importance in PhP-mediated heat conduction. So far, in most theoretical analyses and explanations of experimental data, the PhP decay length is determined based on the imaginary part of the in-plane wavevector. This is only valid for thin films/nanowires without structural imperfections. Therefore, it would be of great interest to probe how various factors such as surface roughness, grain boundaries, and stacking faults alter the PhP decay length. In fact, it has been suggested in Ref. 35 that the PhP decay length is dominated by the presence and density of stacking faults in 3C-SiC nanowires with a value much smaller than that reported for most other PhP-supporting media.

In terms of the effect of exotic features of PhPs, one example could be polariton-mediated heat conduction in hyperbolic materials. PhPs can have a hyperbolic dispersion with multiple branches in the Reststrahlen band. For example, both boron nitride [39, 40] and MoO_3 [41] are naturally occurring van der Waals crystals supporting hyperbolic phonon polaritons (HPhPs). In fact, recent experimental studies have suggested that in bulk hBN and MoO_3, radiative transfer via HPhPs could make contributions to the thermal conductivity that is comparable to or even larger than those from phonons [42, 43]. It would be interesting to see that in the temperature range corresponding to the spectral bandwidth of the Reststrahlen band, whether multiple dispersion branches of HPhPs can be efficiently stimulated by thermally excited infrared photons and contribute to the thermal conductance; and if so, how these HPhPs alter the temperature dependence of polaritonic thermal conductivity. It could be expected that excitation of multiple branches of HPhPs would increase the number density of PhPs, and hence the polaritonic heat capacity. Direct experimental evidence for this could not only demonstrate the possibility of thermally stimulating multiple branches of HPhPs, but also provide distinct signatures of HPhP-mediated heat conduction.

The effect of PhP-mediated heat conduction in various thermal measurements is also a topic of great interest. For example, in the two popular thin film and nanowire thermal conductivity measurement techniques, the time-domain thermal reflectance (TDTR) and microthermal bridge methods, potential PhP effects might have been neglected in the measurements of polar thin films/nanowires. In the TDTR technique, a thin pad of Au or Al is often deposited at the surface as the transducer. If this metal layer is on top of polar materials, it may help launch SPhPs to laterally spread from the metal pad, which could carry away some heat. For the microthermal bridge method, electron beam-induced deposition (EBID) of Pt is often performed locally at the contact between the nanowire and the suspended membranes. For polar nanowires/nanoribbons, this EBID Pt might effectively launch SPhPs that could enhance the thermal conductivity of the uncoated polar nanowires/ribbons.

While the main purpose of this paper is to review relevant previous efforts to provide perspectives on the thermal effects

TABLE 1: Challenges and Opportunities for PhP-mediated Heat Conduction.

Topics	Challenges and opportunities
Measurements of PhP-mediated heat conduction	(1) To distinguish contributions from phonons and polaritons (2) To achieve effective coupling between SPhPs and thermal reservoirs (3) To realize effective launching of SPhPs (4) To verify theoretical predictions such as quantized SPhP transport
Theoretical aspects	(1) Rigorous theoretical understanding of the launching mechanisms for non-equilibrium SPhPs (2) SPhP scattering by surface roughness and structural defects (3) SPhP interactions with thermal reservoirs
Materials	(1) Effects of relevant material properties on SPhP-mediated conduction (2) Size effects in different materials (3) SPhP-mediated heat conduction in hyperbolic materials (4) SPhP-mediated heat conduction in multi-layer structures

of PhPs in polar solids, it should be noted that other types of polaritons such as SPPs could also contribute to energy transport in solids. For example, in a study to explore the thermal effects of SPPs, [44] Kim et al. reported a SPP thermal conductivity of > 2 W/m·K in Ti thin films, which is ~ 25% of the thermal conductivity for bulk Ti.

Overall, polariton-mediated heat conduction, which has been largely neglected in the past due to the low polariton number density in solids, could serve as an effective heat dissipation channel in properly designed nanostructures of polar materials. While the contribution of polaritons to thermal conductivity has been recently confirmed, many scientific questions remain to be answered in this emerging research field, with some examples discussed above and summarized in Table 1. Further studies and better understanding of PhP-mediated heat conduction could have important implications in fast heat dissipation for various engineering devices.

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Author contributions

D.L.: conceptualization, writing the original draft, review and editing. Z.P.: review and editing. J.D.C.: review and editing.

Data availability

Not applicable.

Declarations

Conflict of interest The authors declared no conflicts of interest.

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