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Akhiezer mechanism limits coherent heat conduction in phononic crystals

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Heat in phononic crystals (PnCs) is carried by phonons, which can behave coherently (wave-like) or incoherently (particle-like) depending on the modes, temperature, and length scales. By comparing the measured thermal conductivity of PnCs with theories, recent works suggest that thermal conductivity of PnCs can be explained by considering only surface and boundary scatterings, which not only backscatter phonons but also break their coherence. The logic here is that since average phonon wavelength at room temperature is only a few nanometers, the roughness at the surfaces and boundaries make the scattering diffusive (break the phase coherence of phonons), and thus only very long wavelength (low frequency) phonons with negligible contribution to total thermal conductivity remain coherent. Here, we theoretically show that in a thin film PnCs, the low frequency coherent phonons could significantly contribute to thermal conductivity when assuming the three-phonon scattering model for intrinsic scattering because of their extremely large density of states that result from the low dimensional nature. Yet, further analysis shows the contribution of the low frequency coherent phonons is still negligible within temperature range from 130 to 300 K due to the Akhiezer mechanism, which properly answers the question why the thermal conductivity of PnCs can be explained by considering only scattering of incoherent phonons at these temperatures.

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I. INTRODUCTION

Phononic crystals (PnCs) with specifically designed periodic structures are meant to manipulate propagation of phonons using the coherent effect. In such a case, phonons follow the dispersion relation of PnCs, whose branches are folded and have band gaps, which reduce group velocity, and hence result in reduction of thermal conductivity [1]. The benefit of manipulating thermal conductivity of PnCs using the coherent effect is that it has smaller influence on electrons. Therefore, they are regarded as attractive candidates for enhancing the figure of merit of thermoelectric materials.

The promising prospect of controlling phonons by using the coherent effect in periodic structures has triggered many experimental measurements of the thermal conductivity of PnCs. The most popular class of PnCs is silicon thin films with periodic holes as they can be fabricated by conventional microfabrication technique. However, the reduced thermal conductivity in PnCs can be attributed to phonon coherent effect only for temperatures below 10 K [2,3], and recent theoretical works have confirmed that thermal conductivity of PnCs seen at room temperature in some of the early works can be explained by considering only scattering of incoherent phonons (i.e., phonons that lose phase and follow the dispersion relation of bulk crystals instead of the dispersion relation of PnCs), that is to say that coherent phonons have negligible effect on the thermal conductivity of PnCs at room temperature [4,5].

A possible theoretical explanation for the negligible contribution of coherent phonons is that the coherent transport requires atomically smooth boundary surfaces, and the absence of impurities and defects [6–9], which can only be realized in limited structures such as superlattices [10,11]. As a result, in PnCs, coherence of thermal phonons, whose wavelengths are only a few nanometers at room temperature, is lost when scattered by nanoscale roughness and disorders. Therefore, room temperature coherent transport only occurs for long-wavelength or low frequency phonons ($<$200 GHz [3,8]) with large relaxation time, but their small density of states makes the contribution to thermal conductivity negligible. The logic of the above explanation is true for bulk crystals but fails in the case of the usual PnCs that take the form of films because these low frequency coherent phonons have extremely large density of states owing to the low dimensional nature of the PnCs [12,13]. This leads to the result that even very low frequency coherent phonons could significantly contribute to thermal conductivity when only considering the three-phonon scattering mechanism for evaluating intrinsic phonon relaxation time (here, intrinsic relaxation time is referred to as the lifetime due to phonon-phonon interaction [14]), as will be discussed later. That is to say thermal conductivity of PnC cannot be explained by a boundary scattering of incoherent phonons if only considering three-phonon scattering. Therefore, the reason for the negligible contribution of coherent phonons remains unclear.

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In fact, the large contribution of low frequency coherent phonons suggests that more detailed discussion should be given to their relaxation time. Indeed, for low frequency phonons at room temperature, the consideration of only the three-phonon scattering mechanism is not sufficient. Experimental measurements and theoretical works have shown that relaxation time of low frequency phonons (sound waves) is dominated by Akhiezer damping rather than the three-phonon scattering mechanism (Landau-Rumer theory) in a variety of bulk materials [14–19]. The mechanism of Akhiezer damping is a coupling of the strain of sound waves and thermal phonons: sound wave strain disturbs the local occupation of thermal phonons whose frequencies depend on strain, and the thermal phonons then collide with one another, returning the system to local thermal equilibrium as energy is removed from the sound waves [15]. Such a mechanism should also affect the relaxation process of coherent phonons in PnCs, which have frequencies within the subterahertz (sub-THz) range, and are basically sound waves. Since the original work of Akhiezer, the mechanism of Akhiezer was found to be important for the absorption of sound waves, as well as for energy dissipation in mechanical nanoresonators [20,21], however, few studies have noticed its importance in the field of heat conduction.

In this paper, we show that the Akhiezer mechanism plays an important role in heat conduction for low dimensional materials like PnCs. We theoretically illustrate that the Akhiezer mechanism significantly reduces contribution of coherent phonons to thermal conductivity of PnCs at the temperature regime from 130 to 300 K to the extent that it becomes intrinsically small even when there is no roughness, thus, properly answering the question why the thermal conductivity of PnC can be explained by considering only scattering of incoherent phonons.

II. THEORY FOR THERMAL CONDUCTIVITY OF COHERENT AND INCOHERENT PHONONS

The total thermal conductivity κ_{Total} of thin film and PnCs includes the contributions of both coherent (κ_{coh}) and incoherent phonons (κ_{inc}), which is expressed as [22]

$$\kappa_{\text{Total}}(\omega_s) = \kappa_{\text{coh}}(\omega_s) + \kappa_{\text{inc}}(\omega_s), \tag{1}$$

where ω_{s} is the upper frequency bound of the coherent regime, in other words, the switching frequency between the coherent and incoherent regimes.

We calculate the contribution from incoherent phonons (κ_{inc}) of thin films and PnCs based on the kinetic theory, which is expressed as

$$\kappa_{\text{inc}}(\omega_s) = \int_{\omega_s}^{\infty} C(\omega) D_{\text{bulk}}(\omega) v_{\text{bulk}}(\omega) l(\omega) d\omega, \tag{2}$$

where ω is the frequency; $C(\omega)$, $D_{\text{bulk}}(\omega)$, $v_{\text{bulk}}(\omega)$ denote the frequency dependent heat capacity, the bulk density of states, and group velocity, respectively. $l(\omega)$ is the effective mean free path (MFP) of incoherent phonons obtained by Monte Carlo ray-tracing method [23].

Similarly, contribution of coherent phonons to thermal conductivity (κ_{coh}) of thin films and PnCs is calculated by

$$\kappa_{\text{coh}}(\omega_s) = \int_{0}^{\omega_s} C(\omega) D(\omega) v_g(\omega)^2 \tau(\omega) d\omega, \tag{3}$$

where $D(\omega)$, $v_{\text{g}}(\omega) = \partial\omega/\partial q$ and $\tau(\omega)$ denote the frequency dependent density of states, group velocity, and relaxation time for coherent phonons in thin films and PnC, respectively.

The parameters $C(\omega)$, $D(\omega)$, and $v_{\text{g}}(\omega)$ in Eq. (3) can be calculated from phonon dispersion of thin films and PnCs, which is obtained by solving the continuum-based elastic wave equation using the finite element method (FEM) [13]:

$$\mu\nabla^2 u + (\mu + \lambda)\nabla(\nabla \cdot u) = -\rho\omega^2 u, \tag{4}$$

where u is the displacement vector, $\rho = 2329\,\text{kg}\,\text{m}^{-3}$ is the mass density of the silicon crystal, and $\lambda = 69.3\,\text{GPa}$ and $\mu = 81.3\,\text{GPa}$ are the Láme parameters of the silicon crystal.

As discussed in Sec. I, relaxation of coherent phonons is expected to take two forms: the three-phonon scattering mechanism (Landau-Rumer theory) and Akhiezer damping. The relaxation time due to the three-phonon scattering mechanism is approximated by the Klemens model, which has been widely used and validated [24,25]

$$\tau_K^{-1} = BT\omega^2, \tag{5}$$

where T is the temperature, and B is a constant often quantified empirically.

It should be noted that Landau-Rumer theory is also based on the concept of three-phonon scattering, however, it only includes sound-phonon-phonon interactions. Here, we use the three-phonon scattering model instead as it also includes sound-sound-sound and sound-sound-phonon interactions, which is a more accurate description. On the other hand, the relaxation time of Akhiezer damping is modeled using the expression derived by Maris [17]

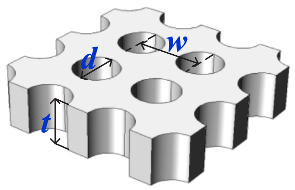
$$\tau_A^{-1} = \frac{C_v T}{\rho v^2} \cdot \frac{\omega^2 \tau_{ph}(\langle\gamma^2\rangle - \langle\gamma^2\rangle)}{1 + \omega^2 \tau_{ph}^2}, \tag{6}$$

where C_{v} is the specific heat capacity per volume, γ is the Grüneisen parameter, v is the phonon phase velocity, and τ_{ph} is the averaged relaxation time of thermal phonons.

Here, we include the mechanisms of both three-phonon scattering and Akhiezer damping into phonon relaxation time τ by using Matthiessen's rule as [14,16]

$$\tau^{-1} = \tau_K^{-1} + \tau_A^{-1}. \tag{7}$$

Equation (6) shows that phonon relaxation time first yields a quadratic frequency dependence for the lower frequencies, with a factor almost three orders of magnitude smaller than the three-phonon scattering, and in the high-frequency limit of the Akhiezer model (around tens of GHz), the lifetime is independent of frequency, and becomes constant [14]. This and Eq. (7) indicate that the relaxation time of phonons first follows the Akhiezer model and then transits to three-phonon scattering when phonon frequency becomes higher. The transition frequency between three-phonon scattering and Akhiezer's damping is expected to happen around several hundred GHz, which was experimentally observed by Hasson and Many [19]. The transition zone of the two scenarios was observed by Maznev *et al.* at room temperature [14].



(a) Phononic Crystals

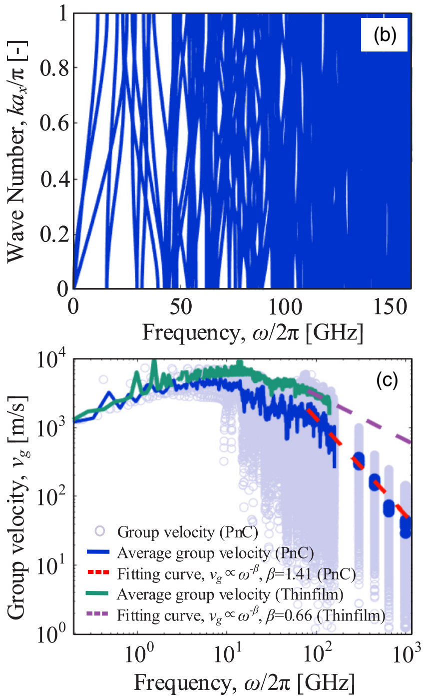


FIG. 1. (a) Schematic of a two-dimensional silicon phononic crystal (PnC). $t = 150$ nm, $w = 100$ nm, and $d = 80$ nm denote the height, width, and hole diameter of the PnC, respectively. (b) Phonon dispersion relation of PnC along G-X. (c) Frequency-dependent phonon group velocity of PnCs and thin film.

III. COHERENT HEAT CONDUCTION

A. Structures, Dispersion Relation, and Group velocity

We considered a two-dimensional (2D) silicon thin film with periodic cylindrical holes, which is the most frequently studied representative PnC [3] [Fig. 1(a)]. The height t, width w, and hole diameter d of the PnC are set to 150 nm, 100 nm, and 80 nm, respectively. A folded dispersion relation in the frequency range of 0–160 GHz [Fig. 1(b)] is obtained by solving Eq. (4) with 2D periodic boundary conditions. It is shown that the folded dispersion curves become flatter as frequency increases, which indicates reduction in group velocity [Fig. 1(c)]. Furthermore, although the frequency-dependent profile of group velocity is scattered, when smoothed by averaging the group velocities for each frequency, the profile in the range between 80 and 160 GHz shows a clear power-law frequency dependence. An exponent of -1.41 is obtained by fitting a power law to the data in this frequency range. The fitting curve was then extrapolated to obtain average group velocity in higher frequency regimes. Note that the extrapolation is needed because the computational load to calculate full dispersion relations of higher frequency phonons would become too large. The validity of the extrapolation was confirmed by calculating tens of branches of dispersion around given frequencies within 1THz, and the average group velocities around given frequencies were confirmed to agree with the fitting curve. With the same approach as for PnC, we also obtained group velocity of a thin film with the same thickness (150 nm), and only average group velocity is plotted [Fig. 1(c)]. It is shown that the average group velocity of a thin film is larger than that of the PnC because periodic structures in PnC cause larger bandgaps, which reduce group velocity.

B. Temperature Dependent Phonon Relaxation Time

First, to show the validity of the calculation, we obtained the phonon relaxation time of bulk silicon crystals from first-principles-based lattice dynamic calculation, which agrees with experimental data at a temperature range of 130–300 K [Fig. 2(a)]. The maximum difference between our calculation and experimental data is 25%. It is clear that, for a given temperature, phonon relaxation time deviates from the three-phonon scattering scenario, and transits to the Akhiezer damping scenario when phonon frequency becomes GHz. As a result, the relaxation time of low frequency phonons is reduced by 3 orders. It should be noted that the transition between the two scenarios has been investigated only at room temperature for Si and GaAs-AlAs superlattices [14]. Here, in Fig. 2(a), by comparing experiment data with our calculation, we observed that the transition takes place at $\sim$200 GHz for 200–300 K, and $\sim$100 GHz for 130 K.

Now that the calculation is validated, we obtained the relaxation time of the acoustic branches ($<$12 GHz) for PnCs, as plotted in Fig. 2(b), taking the case of 300 K as an example. It is shown that the trend of the relaxation time for the longitudinal mode of PnCs agrees with that of bulk crystals, however, the magnitude is smaller due to the folding effect, which yields phonon bandgaps and reduces phase velocity v. Other acoustic branches show similar characteristics. For optical phonons, instead of replacing phase velocity v in Eq. (6) by group velocity v_g, as in the work of Chavez-Angel *et al.* [26], we approximated v by the average phase velocity of all acoustic branches considering that optical branches are folded acoustic ones. One can observe that phonon relaxation time transits to that of three-phonon scattering as frequency increases, which indicates that three-phonon scattering mechanism is the dominant phonon decay process for high frequency phonons. Similar relaxation time transitions also happens for the 2D thin film; the difference is that the average relaxation time of the thin film is larger than that of PnC due to a larger phase velocity v, which resulted from smaller bandgaps in dispersion relations of thin films.

C. Influence of Akhiezer Damping on Thermal Conductivity of Coherent Phonons

Next, we discuss how much the transition from three-phonon scattering to Akhiezer damping can affect thermal conductivity of both bulk crystals, thin films, and PnCs. We first verified that such a transition has a negligible effect on the total thermal conductivity of bulk crystals when temperature is below 300 K. As for thin films and PnCs, we assume

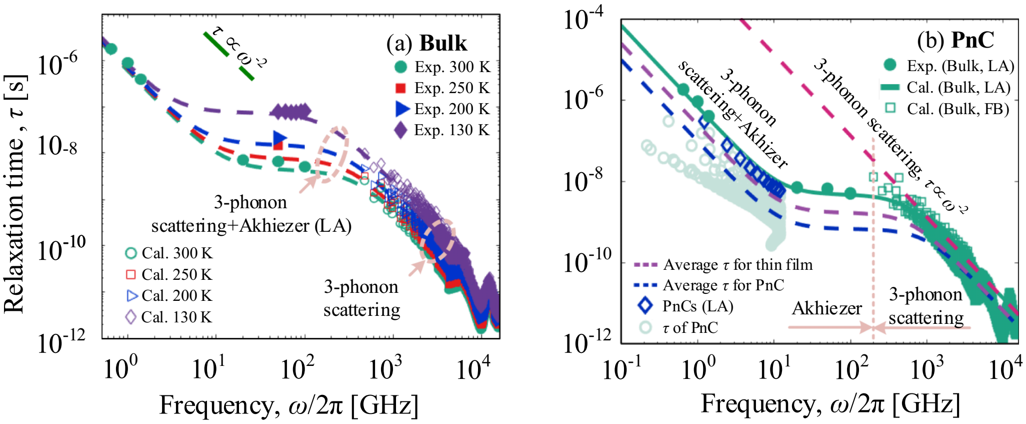


FIG. 2. (a) Frequency and temperature dependent phonon relaxation time for bulk silicon crystals. The dashed lines are calculated results for longitudinal modes (LA) at different temperatures. Experimental data are measured results for LA modes and are taken from Refs. [15,27–29] (b) Frequency-dependent relaxation time of thin film and PnCs at 300 K, and a comparison with bulk phonon relaxation time for LA modes and modes in full Brillion zone (FB). The boundary of Akhiezer and three-phonon-scattering regimes at 300 K is around 200 GHz. Note that "three-phonon scattering+Akhiezer" in Fig. 2 means the relaxation time calculated using Eq. (7).

the switching frequency ω_s as 0.2 THz, i.e., the coherent regime is 0–0.2 THz, and leave the discussion of frequency dependence on the coherent regime for later, as it does not affect the discussions here. It should be noted that the coherent regime here (0–0.2 THz) is not taken randomly, but matches with the case that the thin film and PnC have a 2-nm-surface roughness [3,8], which is the average value of the most frequently measured roughness in experiment (1–3 nm). The method for determining the coherent regime by roughness is discussed in the Appendix. The κ_{inc} is obtained by Monte Carlo ray-tracing calculation, in which boundary scattering of incoherent phonons is included. In the case of coherent phonons behaving as waves, the boundary effect is included as the folded dispersion of coherent phonons (Fig. 1), which are formed when the phonons are reflected without dephasing at the periodic boundaries.

A comparison of thermal conductivity of a thin film and a PnC with two different relaxation time τ models (with and without Akhiezer, as in Eq. (6) and Eq. (7), respectively) for 130 and 300 K is shown in Fig. 3. First, we discuss the results when there is only three-phonon scattering. In this case, κ_{coh} of the thin film within 0–0.2 THz is 5 $\text{W m}^{-1}\,\text{K}^{-1}$ at 300 K and 17.8 $\text{W m}^{-1}\,\text{K}^{-1}$ at 130 K, which contributes about 7.4% and 15.2% of κ_{Total} for 300 and 130 K, respectively [Fig. 3(a)]. The proportion of κ_{coh} in κ_{Total} becomes even larger for PnCs due to larger density of states, which will be shown later. At 300 K, κ_{coh} of PnC is 9.5 $\text{W m}^{-1}\,\text{K}^{-1}$, which contributes 53% of κ_{Total} for PnC. At lower temperature of 130 K, κ_{coh} of PnC reaches 33.3 $\text{W m}^{-1}\,\text{K}^{-1}$, and contributes to 81.1% of κ_{Total} for PnC. If this is the case, the total thermal conductivity of PnCs cannot be explained by scattering of incoherent phonons, which is not the actual situation of previous theoretical and experimental results [3–5]. As discussed in Secs. I and III B, for low frequency phonons, only considering three-phonon scattering is not enough, Akhiezer damping should be considered as a key issue for the relaxation process of these phonons, and it can be included in phonon relaxation time using Eqs. (5) and (6). For the case where Akhiezer damping is considered, κ_{coh} of both thin film and PnC is smaller than 0.5 $\text{W m}^{-1}\,\text{K}^{-1}$ for 300 and 130 K, and the proportion of κ_{coh} in κ_{Total} is less than 1%, which indicates that κ_{coh} is negligible in both thin film and PnC for 130–300 K [Fig. 3(b)], and that κ_{Total} is almost dominated by the incoherent part κ_{inc}. The implication here is that, for low dimensional materials like thin films and PnCs, it is important to take Akhiezer damping into account to properly evaluate the relaxation time of low frequency phonons, otherwise, their contributions to thermal conductivity can be hugely overestimated by only considering three-phonon scattering.

D. Phonon Density of States

From Sec. III C, we see that Akhiezer damping does not influence the total thermal conductivity of bulk silicon crystals but has a large influence on thermal conductivity of PnCs. The reason lies in density of states, $D(\omega)$ (Fig. 4). In bulk crystal, $D(\omega)$ is proportional to ω^2, which indicates that $D(\omega)$ of low frequency phonons is very small. Therefore, even relaxation time of these phonons is overestimated by

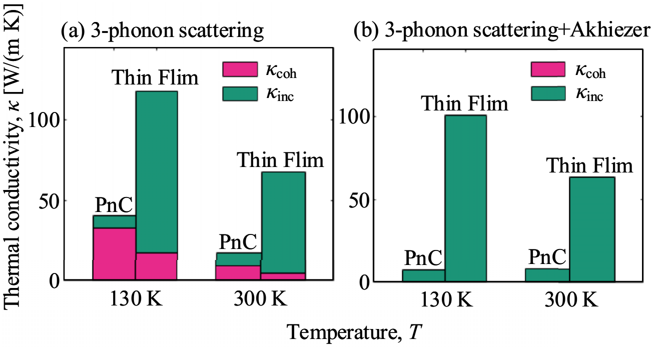


FIG. 3. A comparison of thermal conductivity of thin films and PnCs with two different relaxation time τ models at 130 and 300 K for a 2-nm roughness. (a) τ model: only three-phonon scattering; (b) τ model: three-phonon scattering and Akhiezer damping. The coherent regime is determined as 0–0.2 THz for 300 and 130 K when there is a 2-nm roughness.

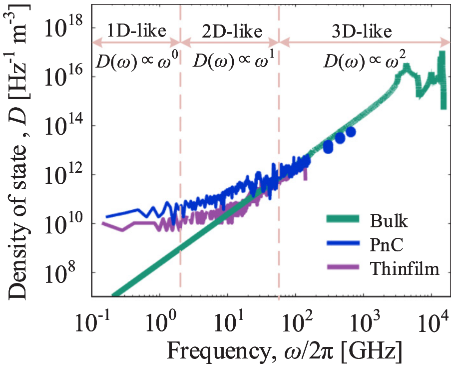


FIG. 4. Frequency-dependent density of states $D(\omega)$ of thin films and PnCs, and a comparison with density of states of bulk silicon crystals.

only considering the ω^{-2}-dependent three-phonon scattering; their contributions to the total thermal conductivity of bulk silicon crystal is still negligible, in other words, we do not need to consider the Akhiezer damping effect on the thermal conductivity of bulk crystal.

However, in PnCs, $D(\omega)$ transits from 3D to 2D to 1D as frequency decreases due to coherent effect, and accordingly, the frequency dependence of $D(\omega)$ changes from ω^{-2} to ω^{-1} to ω^{0}. As a result, $D(\omega)$ of low frequency phonons in the thin film and PnC is much larger than $D(\omega)$ in bulk crystals, which leads to a significant overestimation of κ_{coh} in thin films and PnCs when only considering the ω^{-2}-dependent three-phonon scattering for intrinsic relaxation time. We also noticed that, in the case without Ahkiezer damping, κ_{coh} for PnC is larger than κ_{coh} of the thin film [Fig. 3(a)], even phonon group velocity is larger for the thin film (Fig. 1). This is because the $D(\omega)$ of the acoustic phonon (<12 GHz) in PnCs is four times larger than the $D(\omega)$ of the acoustic phonon in thin films, which would lead to severe overestimation κ_{coh} for PnC.

The conclusion for Sec. III is that, although the existence of a 2-nm-surface roughness makes the coherent regimes very small (0–0.2 THz), the density of states of the low frequency coherent phonons is much larger than that of bulk silicon crystals, therefore, these phonons have a large potential to contribute to thermal conductivity when only three-phonon scattering is considered. However, when the Akhiezer mechanism is involved, relaxation time of these phonons is hugely reduced, and as a result, the proportion of κ_{coh} in κ_{Total} for both the thin film and PnC is negligible (<1%), which is why the total thermal conductivity of the thin film and PnC, κ_{Total} can be explained by considering only the contributions of incoherent phonons κ_{inc}.

The current result is consistent with the recent experimental and theorertical works on thin film PnCs [3–5]. They have successfully reproduced the experimental results with Monte Carlo calculations by ignoring the contribution of the sub-THz phonon to thermal conductivity. The fact that a calculation could reproduce the experiments means that the Akhiezer damping has suppressed the phonon relaxation time, and that the results are consistent with our work. Although the actual geometry of our PnC and these works are different, the above discussion on the dimension and contribution of sub-THz phonon contribution should be applicable to PnC with thickness and holes on the order of 100 nm.

IV. SWITCHING-FREQUENCY-DEPENDENT COHERENT AND INCOHERENT HEAT CONDUCTION

So far, our discussion has been based on the assumption that the roughness of the thin film and PnC is 2 nm, and the switching frequency ω_{s} is $\omega_0 (= 0.2\,\mathrm{THz})$. However, the ω_{s} can change when roughness on the surface is modified. Therefore, in what follows, we consider ω_{s} as a variable to investigate contributions of coherent phonons to thermal conductivity. Note that this also helps gain understanding of the case with no roughness, which is the theoretical upper limit of the contribution of coherent phonons. Nevertheless, we can determine the maximum value of ω_{s} using the criterion that the MPF of bulk phonons should be at least larger than several periods of periodic structures in PnCs (here, it is 100 nm). The reason is that coherent length should be smaller than MFP bulk phonons, and MFP are required to be sufficiently long such that they can cross several periodicities, thereby creating many secondary waves to achieve the interference effect, which results in the folded dispersion relation of PnCs [6]. The minimum number of periodicity is two (the phonon passes though the PnC and then is reflected back), which gives the upper bound of ω_{s}.

If $\omega_s < \omega_0$, κ_{coh} within $0-\omega_{\mathrm{s}}$ can be directly calculated by Eq. (3) with their full dispersion relations, whereas if $\omega_{\mathrm{s}} > \omega_0$, the contributions of coherent phonons from ω_0 to ω_{s} is estimated by the averaging method due to a lack of information in the full dispersion relation. In the averaging method, we approximate the phonon group velocity and density of states in Eq. (1) with the averaged group velocity obtained by the fitting curve [Fig. 1(c)], and bulk phonon density of states (Fig. 4), respectively. The latter approximation is based on the observation that density of states of PnCs and bulk crystal are roughly the same for frequencies above ω_0 (Fig. 4). Then, the switching-frequency dependent $\kappa_{\mathrm{coh}}(\omega_{\mathrm{s}})$ of PnCs or thin films can be expressed as

$$\kappa_{\mathrm{coh}}(\omega_s) = \kappa_0 + \int_{\omega_0}^{\omega_s} C(\omega) D_{\mathrm{bulk}}(\omega) v_{av}(\omega)^2 \tau(\omega) d\omega, \qquad (8)$$

where κ_0 is the contribution of coherent phonons with frequencies between 0 and ω_0, and the second term represents the contribution of coherent phonons with frequencies between ω_0 and ω_{s}, where $v_{\mathrm{av}}(\omega)$ is the averaged group velocity and $D_{\mathrm{bulk}}(\omega)$ is the bulk phonon density of states.

For a thin film and PnC at 300 K [Figs. 5(a) and 5(c)], the maximum coherent regime is determined as 0–3.0 THz. We found that κ_{coh} of both the thin film and PnC are negligible when compared to κ_{inc}, even when ω_{s} reaches its maximum (3 THz). The results indicate that κ_{coh} contributes a very small proportion of κ_{Total} for both thin films and at 300 K, even though there is no roughness effect. This is because of two reasons: one is that the Akhiezer damping significantly reduces the relaxation time of phonons in the Akhiezer regime [<0.2 THz for 300 K, Fig. 3(b)], and the other is that the group velocity of phonons in the three-phonon-scattering regime [0.2–3.0 THz, Fig. 3(b)] is small due to phonon bandgaps that

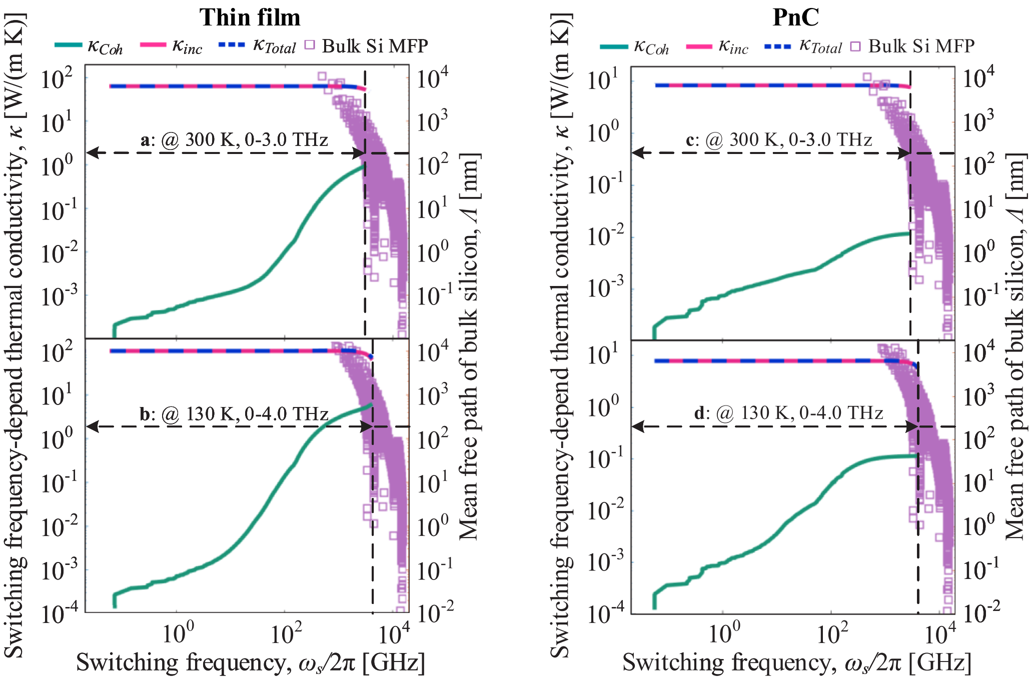


FIG. 5. Contributions of coherent and incoherent phonons to total thermal conductivity of thin films and PnCs as a function of switching frequency at 300 and 130 K. The maximum coherent regime is determined as 0–3 THz at 300 K, and 0–4 THz at 130 K.

are caused by the folding effect. As a result, the contributions of phonons within the whole coherent regime are limited.

As temperature decreased to 130 K [Figs. 5(b) and 5(d)], the maximum value of ω_s increased to 4 THz, and Akhiezer damping is weakened due to a reduction in thermal phonon population. Therefore, relaxation time of coherent phonons increases (Fig. 2), however, Ahkiezer damping still has a strong influence on the κ_{coh} of the thin film and PnC. For the PnC at 130 K, the maximum value of κ_{coh} is only $0.1\,\mathrm{W\,m^{-1}\,K^{-1}}$. In thin film, the value of κ_{coh} is larger when comparing with κ_{coh} for PnC, and can contribute to $5.8\,\mathrm{W\,m^{-1}\,K^{-1}}$ when ω_s reaches its maximum (4 THz). This is because group velocity of the three-phonon- scattering regime (0.1–4 THz for 130 K) is larger than that of PnC (Fig. 1). However, the proportion of κ_{coh} in κ_{Total} is still small ($<8\%$). We also noticed that for both thin film and PnC, sub-THz phonons contribute to the most part of κ_{coh}, and above THz coherent phonons do not contribute much to κ_{coh} due to their small group velocity and relaxation time [Figs. 1(c) and 2].

Taking the most extreme case (no roughness) as an example, a comparison of thermal conductivity of a thin film and a PnC with two different relaxation time τ models at 130 and 300 K is shown in Fig. 6. The discussion here is similar to Sec. III C. The coherent part, κ_{coh} can contribute to a large part of κ_{Total} when not considering Akhiezer damping. In a thin film, κ_{coh} can contribute to about 11.2% and 28% of κ_{Total} for 300 and 130 K, respectively. For PnC, contributions of κ_{coh} can even reach to 56% at 300 K and 85% of κ_{Total}. However, Akhiezer damping can reduce the proportion to less than 2% for PnC at both 300 and 130 K, and for the thin film, the proportion is less than 2% at 300 K and around 8% at 130 K. The value of the proportions here is a little larger than that in Sec. III C, however, it is still small. Therefore, we can conclude that the contribution of coherent phonons κ_{coh} to the total thermal conductivity κ_{Total} for both thin film and PnC is very small, even if there is no roughness, or the κ_{Total} for both the thin film and PnC can be explained by the contributions of incoherent phonons.

Finally, it should be noted that currently our calculation cannot deal with the cases for temperatures below 130 K. The reason is that the Ahkiezer mechanism, as originally developed, is only valid at high temperatures, however, even now the exact valid temperature regime is not known. We can extend our calculation to 130 K because the experimental measurements match with theoretical calculations (Fig. 2). More experimental measurements are still needed below 130 K. On the other hand, the equation for Ahkiezer damping

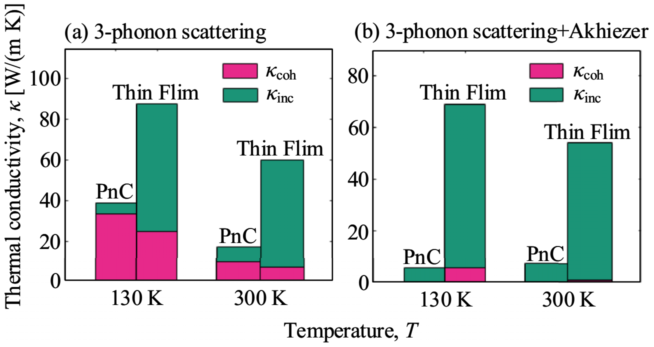


FIG. 6. A comparison of thermal conductivity of a thin film and PnC with two different relaxation time τ models at 130 and 300 K for the most ideal case (no roughness). (a) τ model: only three-phonon scattering; (b) τ model: three-phonon scattering and Akhiezer damping. The coherent regime is, respectively, determined as 0–3 THz and 0–4 THz for 300 and 130 K when there is no roughness.

[Eq. (6)] includes average phonon relation time of the bulk silicon crystal (τ_{ph}), which is often obtained by single phonon relaxation time approximation. However, the single phonon relaxation time approximation for the Boltzmann equation may not be valid at lower temperatures. We hope that lower temperature measurements and deeper theoretical analysis of phonon relaxation time in both bulk and nanostructured materials will soon become available.

V. CONCLUSIONS

In conclusion, we show that average group velocity of high frequency coherent phonons in thin films and PnCs can be approximated by the exponential function $\omega^{-\beta}$ ($\beta = 0.66$ and 1.41 for our thin film and PnC samples, respectively), which indicates that higher frequency phonons have smaller group velocities, and thus contribute less to thermal conductivity. Then, we show that low frequency coherent phonons in low dimensional materials like PnCs have an extremely large density of states due to the low dimensional nature, which could significantly contribute to thermal conductivity when only considering three-phonon scattering. However, by comparing experimental data with our calculation, we show that Akhiezer damping is dominant and should be considered when dealing with relation time of low frequency phonons (<200 GHz for 200–300 K, and <100 GHz for 130 K). Because of Akhiezer damping, the contribution of coherent phonons is reduced to the extent that their contribution to total thermal conductivity of thin films and PnCs at 130–300 K becomes very small (<8%), even if there is no surface roughness. That is why the total thermal conductivity of thin films and PnCs can be explained by considering only the incoherent phonons.

APPENDIX

In this Appendix, we evaluate switching frequency ω_s as a function of roughness according to the work of Wagner *et al.* [8]:

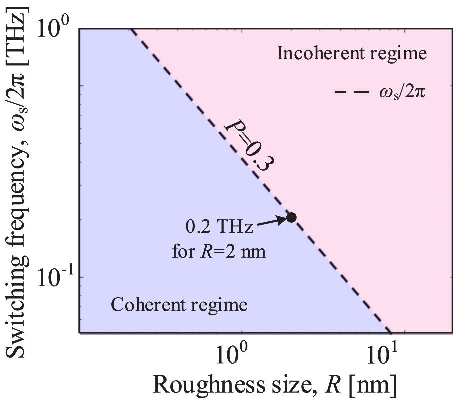


FIG. 7. Switching frequency as a function of roughness size R (surface roughness, hole wall roughness, lattice site displacement, hole disorder, etc.) for selected specularity parameters $P = 0.3$.

$$\omega_s = \frac{2\pi V_s}{R}\sqrt{\frac{-\mathrm{In}(P)}{16\pi^3}}, \tag{A1}$$

where R is the roughness size (including surface roughness, hole wall roughness, lattice site displacement, disorder etc.), P is the specularity, here we use $P = 0.3$ to define the boundary of coherent and incoherent regimes, i.e., the ω_s, and V_s is the longitudinal sound velocity as 8433 m/s [8].

The switching frequency as a function of roughness size R is shown in Fig. 7, from which we can obtain 0.2 THz for ω_s when $R = 2$ nm. In the work of Wagner *et al.*, $P = 0.5$, but here we use 0.3 just for a more conservative estimation.

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[1] J. K. Yu, S. Mitrovic, D. Tham, J. Varghese, and J. R. Heath, Nat. Nanotechnol. **5**, 718 (2010).
[2] N. Zen, T. A. Puurtinen, T. J. Isotalo, S. Chaudhuri, and I. J. Maasilta, Nat. Commun. **5**, 3435 (2014).
[3] J. Maire, R. Anufriev, R. Yanagisawa, A. Ramiere, S. Volz, and M. Nomura, Sci. Adv. **3**, e1700027 (2017).
[4] J. Lee, W. Lee, G. Wehmeyer, S. Dhuey, D. L. Olynick, S. Cabrini, C. Dames, J. J. Urban, and P. D. Yang, Nat. Commun. **8**, 14054 (2017).
[5] K. D. Parrish, J. R. Abel, A. Jain, J. A. Malen, and A. J. H. McGaughey, J. Appl. Phys. **122**, 125101 (2017).
[6] M. Maldovan, Nat. Mater. **14**, 667 (2015).
[7] N. K. Ravichandran and A. J. Minnich, Phys. Rev. B **89**, 205432 (2014).
[8] M. R. Wagner, B. Graczykowski, J. S. Reparaz, A. El Sachat, M. Sledzinska, F. Alzina, and C. M. S. Torres, Nano Lett. **16**, 5661 (2016).
[9] T. Oyake, L. Feng, T. Shiga, M. Isogawa, Y. Nakamura, and J. Shiomi, Phys. Rev. Lett. **120**, 045901 (2018).
[10] M. N. Luckyanova *et al.*, Science **338**, 936 (2012).
[11] J. Ravichandran *et al.*, Nat. Mater. **13**, 168 (2014).
[12] F. Kargar, B. Debnath, J. P. Kakko, A. Saynatjoki, H. Lipsanen, D. L. Nika, R. K. Lake, and A. A. Balandin, Nat. Commun. **7**, 13400 (2016).
[13] R. Anufriev and M. Nomura, Phys. Rev. B **91**, 245417 (2015).
[14] A. A. Maznev, F. Hofmann, A. Jandl, K. Esfarjani, M. T. Bulsara, E. A. Fitzgerald, G. Chen, and K. A. Nelson, Appl. Phys. Lett. **102**, 041901 (2013).
[15] B. C. Daly, K. Kang, Y. Wang, and D. G. Cahill, Phys. Rev. B **80**, 174112 (2009).
[16] A. A. Maznev, J. A. Johnson, and K. A. Nelson, Phys. Rev. B **84**, 195206 (2011).
[17] H. J. Maris, Phys. Rev. **175**, 1077 (1968).
[18] T. O. Woodruff and H. Ehrenreich, Phys. Rev. **123**, 1553 (1961).

[19] J. Hasson and A. Many, Phys. Rev. Lett. **35**, 792 (1975).
[20] K. Kunal and N. R. Aluru, Phys. Rev. B **84**, 245450 (2011).
[21] J. Atalaya, T. W. Kenny, M. L. Roukes, and M. I. Dykman, Phys. Rev. B **94**, 195440 (2016).
[22] S. Alaie, D. F. Goettler, M. Su, Z. C. Leseman, C. M. Reinke, and I. El-Kady, Nat. Commun. **6**, 7228 (2015).
[23] T. Hori, J. Shiomi, and C. Dames, Appl. Phys. Lett. **106**, 171901 (2015).
[24] B. L. Davis and M. I. Hussein, Phys. Rev. Lett. **112**, 055505 (2014).
[25] H. Honarvar and M. I. Hussein, Phys. Rev. B **93**, 081412 (2016).
[26] E. Chavez-Angel, R. A. Zarate, J. Gomis-Bresco, F. Alzina, and C. M. S. Torres, Semicond. Sci. Technol. **29**, 124010 (2014).
[27] J. Y. Duquesne and B. Perrin, Phys. Rev. B **68**, 134205 (2003).
[28] H. Y. Hao and H. J. Maris, Phys. Rev. B **63**, 224301 (2001).
[29] M. Pomerantz, Phys. Rev. **139**, A501 (1965).