

# On the importance of Akhiezer damping to thermal conductivity in silicon at elevated temperatures above 300 K

Cite as: J. Appl. Phys. **135**, 045103 (2024); doi: [10.1063/5.0187032](https://doi.org/10.1063/5.0187032)

Submitted: 13 November 2023 · Accepted: 2 January 2024 ·

Published Online: 25 January 2024



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## AFFILIATIONS

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## ABSTRACT

Recently, Chiloyan *et al.* [Appl. Phys. Lett. **116**, 163102 (2020)] have reported that phonon transport could exceed bulk heat conduction if low-frequency phonons with long mean free path (MFP) remain in the nonthermal regime in silicon. To gain a better understanding of their findings, we investigated the effects of temperature-induced anharmonicity on both Landau–Rumer damping and Akhiezer damping, including polarization. To do this, we follow a rigorous procedure for calculating the Akhiezer model and use phonon kinetic theory based on the Boltzmann transport equation. Consequently, we find that in the Akhiezer regime, the longitudinal acoustic phonon modes (LA) are strongly suppressed by phonon anharmonicity compared to the transverse acoustic phonon modes. Therefore, the low-frequency phonons with a long MFP of LA can help to exceed bulk heat conduction if they remain in the regime of nonthermal phonon transport where there are no appreciable scatterings with other phonons. It is also shown that Akhiezer damping eliminates thermal conductivity by 16.8% at 500 K, which is higher than the observed reduction (12.6%) at 300 K in silicon, uncovering a novel regime where the Akhiezer damping, previously deemed insignificant in the thermal conduction of bulk silicon, becomes crucial.

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## I. INTRODUCTION

Understanding phonon transport provides insights into the thermophysical properties of materials, encompassing free energy, thermal expansion, and thermal conductivity.<sup>1</sup> This understanding can be harnessed for optimizing materials in various silicon-based technologies, including energy conversion in thermoelectrics and thermal management in electronics.<sup>2,3</sup>

Recently, Chiloyan *et al.*<sup>4</sup> have reported that nonthermal phonon transport could exceed bulk heat conduction if the main phonon modes generated from the local heat source are low-frequency phonons with long mean free path (MFP) at room temperature (RT) in silicon. However, their proposal remains elusive due to the significant magnitude of heat transfer enhancement, exceeding fivefold, which cannot be sufficiently explained by only the heat conduction from long MFP phonons.

To comprehend phonon transport by low-frequency phonons with a long MFP, it is crucial to grasp the phonon scattering mechanism, which can be classified into two fundamental categories:

Landau–Rumer (L–R) damping and Akhiezer damping.<sup>5</sup> Akhiezer damping pertains to sound absorption in the range  $\omega\tau_{\text{th}} \ll 1$ , where  $\omega$  is the phonon angular frequency and  $\tau_{\text{th}}$  is the average relaxation time of the thermally excited phonons over the Brillouin zone (BZ) in a crystal.<sup>6–9</sup>

Akhiezer damping occurs at  $\omega \ll k_B T/\hbar$ ,<sup>5</sup> which is associated with low-frequency phonons at a given temperature  $T$ , where  $k_B$  is the Boltzmann constant and  $\hbar$  is the reduced Planck constant. In contrast, in the high-frequency regime at low temperatures, L–R damping plays a significant role.<sup>5</sup> Therefore, one would expect a transition frequency between Akhiezer and L–R regimes will be dependent on the temperature. For example, at high temperatures in pure crystals, phonons with a frequency of  $> 100$  GHz correspond to the L–R damping, while those with  $> 10$  GHz correspond to it at lower temperatures.<sup>10</sup> So far, research on the temperature dependence of the attenuation of ultrasonic waves has been confined to temperatures below 300 K.<sup>10</sup> Meanwhile, it has only just begun to explore the effect of these low-frequency phonons on heat conduction in materials.<sup>6–9</sup> However, even in these studies,<sup>6–8</sup> the

accurate determination of parameters associated with the Akhiezer damping has been lacking because these parameters have been fitted by comparing with experimental data.<sup>6–8</sup>

In this study, we aim to expand the temperature range above 300 K and provide a systematic methodology for the determination of parameters associated with the Akhiezer damping to investigate how temperature-induced phonon anharmonicity influences both Akhiezer damping and L–R damping and to gain a better understanding on recent findings of Chiloyan *et al.*,<sup>4</sup> which are unconventional findings. To do this, we conducted a rigorous procedure for calculating the Akhiezer damping instead of the simple fitting method mentioned above and used phonon kinetic theory based on the Boltzmann transport equation (BTE).

## II. METHODOLOGY

Integrating the Akhiezer damping effect into first principles calculations remains challenging, primarily due to the requirement for large supercell sizes, higher-order scattering, and substantial computational resources.<sup>6</sup> Furthermore, applying the Boltzmann transport equation (BTE) to bulk materials is computationally intensive and, therefore, typically limited to micro-/nanoscale regimes.

Because of the substantial computational load involved in simulating bulk materials, we employ here an analytic theory based on the BTE within the frame of the relaxation time approximation under the assumption of thermal equilibrium.<sup>11,12</sup> This well-known classical kinetic theory reads<sup>13,14</sup>

$$K = \frac{1}{3} \sum_j \int C_j v_j \Lambda_j d\omega, \quad (1)$$

where  $K$  is the thermal conductivity,  $j$  is the polarization (LA: longitudinal acoustic phonon modes; TA: transverse acoustic phonon modes),  $C$  is the spectral phonon specific heat,  $v$  is the spectral phonon group velocity, and  $\Lambda$  is the spectral phonon MFP. Among these, obtaining a reliable value  $\Lambda$  is crucial in calculating  $K$  of Eq. (1) because the simplistic kinetic theory based on an averaged phonon MFP value can be very misleading.<sup>15</sup> To obtain a reliable spectral phonon MFP calculation, we incorporated semi-experimental thermal spectroscopy data<sup>16,17</sup> into the estimations of  $\Lambda_j$ . However, the thermal spectroscopy data<sup>16,17</sup> do not account for phonon polarization. To address this limitation, we applied  $\tau_j(\omega_j, T)$  at a given  $T$  proposed by Zhu *et al.*<sup>18</sup> using the lattice dynamics method, where  $\tau$  is the phonon relaxation time. Assuming a known  $v_j$ , this relationship yields  $\Lambda_j(\omega_j, T)$  as  $\Lambda_j = v_j \tau_j$ . Consequently, a fitting function can be found from the function  $\Lambda_j(\omega_j)$ , facilitating the determination of  $\omega_j(\Lambda_j, T)$  as a dependent variable for  $\Lambda_j$ . Knowing  $\omega_j(\Lambda_j, T)$  allows us to determine  $\omega_j$  at  $\Lambda_j$  and  $T$ , which means we can calculate spectral phonon properties with  $\omega_j$  at given  $\Lambda_j$  from the thermal spectroscopy data.<sup>16,17</sup> According to Kim *et al.*'s experimental work,<sup>19</sup> the phonon density of states (DOS) for silicon can be considered to be temperature-independent in the range of 100–600 K. Therefore, in our study, we used phonon DOS provided at 300 K by Zhu *et al.*<sup>18</sup> for silicon, which was given as a function of phonon frequency. We obtained a polynomial function

to fit the data of Zhu *et al.*<sup>18</sup> with an assumption that the phonon DOS is temperature-independent in the range we considered (100–500 K) in this study. We performed our calculations on all of the phonon properties related to  $\omega_j$ , using the experimental dispersion in the [001] direction and assuming that the crystals are isotropic. In addition, the thermal transport by optical phonons and the temperature dependence of  $v_j$  were neglected in this study.<sup>1,20–22</sup>

The “Akhiezer model” for the relaxation time  $\tau_A$  of Akhiezer damping including polarization is given by<sup>5–9</sup>

$$\tau_{A,j} = \tau_{\text{inf},j} \left( 1 + \frac{1}{\omega_j^2 \tau_{\text{th}}^2} \right). \quad (2)$$

Now, we need to obtain the functional form having the temperature dependence of  $\tau_{\text{inf},j}$  and  $\tau_{\text{th}}$  to obtain a closed form for the above Akhiezer model. So far, as mentioned before,  $\tau_{\text{inf},j}$  and  $\tau_{\text{th}}$  have been fitted by comparing with experiment data.<sup>6–8</sup> Instead of fitting here,  $\tau_{\text{th}}$  is taken as

$$\tau_{\text{th}} = 96.85 \times (2\pi\hbar/k_B T), \quad (3)$$

from Ref. 23 for Si that is derived by considering the relation between the collision rate of thermal phonons and the thermal energy of the interacting thermal phonons in Si.<sup>23</sup>

The attenuation  $\alpha$  of the acoustic wave including polarization can be written as<sup>5</sup>

$$\alpha_j = \frac{C_j T}{2\rho v_{a,j}^3} \frac{\omega_j^2 \tau_{\text{th}}^2}{1 + \omega_j^2 \tau_{\text{th}}^2} \Gamma_j, \quad (4)$$

where  $\rho$  is the density and  $v_{a,j}$  is the acoustic wave velocity taken as  $v_{a,LA} = 8400$  m/s and  $v_{a,TA} = 5800$  m/s from Ref. 7. Here,  $\Gamma_j$  is given by  $\Gamma_j = \langle \gamma_j^2 \rangle - \langle \gamma_j \rangle^2$ , where  $\gamma$  represents the mode Grüneisen parameter and angular bracket ( $\langle \rangle$ ) denotes the averages taken over the entire spectrum of thermally excited phonons including the propagation direction and  $\Gamma_j$  is the mean square deviation about  $\langle \gamma_j \rangle$  that is related to the mean square deviation of  $\gamma_j$  about its mean  $\langle \gamma_j \rangle$ ; therefore,  $\Gamma_j$  is associated with an increment in the modulus of elasticity of the solid.<sup>24</sup> With the relation of  $\alpha_j = 1/\Lambda_{\text{Akhiezer},j} = 1/(\tau_{A,j} v_{a,j})$ ,<sup>25</sup> we get the following equation:

$$\tau_{A,j} = \frac{2\rho v_{a,j}^2 \tau_{\text{th}}}{C_j T \Gamma_j} \left( 1 + \frac{1}{\omega_j^2 \tau_{\text{th}}^2} \right). \quad (5)$$

By comparing Eqs. (2) and (5), finally,  $\tau_{\text{inf},j}$  can be defined as

$$\tau_{\text{inf},j} = \frac{2\rho v_{a,j}^2 \tau_{\text{th}}}{C_j T \Gamma_j}, \quad (6)$$

which is a formula independent of the phonon frequency.

Now, we need to determine  $\Gamma_j$  to calculate  $\tau_{\text{inf},j}$  of Eq. (6). According to Klemens,<sup>26</sup> the relaxation time of acoustic phonons can be approximated as  $\tau_j^{-1} = 2\gamma_j^2 (k_B T / M \bar{v}_j^2) (\omega_j^2 / \omega_{\text{max},j})$ . From

this,  $\gamma_j^2$  can be rearranged as

$$\gamma_j^2 = \frac{M\bar{v}_j^2 \omega_{\max,j}}{2k_B T \omega_j^2} \tau_j^{-1}, \quad (7)$$

where  $M$  is the atomic mass for Si ( $M = 46.6 \times 10^{-27}$  kg),<sup>27</sup>  $\omega_{\max,j}$  is the largest frequency of the mode  $j$ , and  $\bar{v}_j$  is the average group velocity associated with the mode  $j$ . Here, we calculate  $\bar{v}_j$  as a weighted average of phonon mode specific heat, resulting in  $\bar{v}_{LA} = 5615.9$  m/s and  $\bar{v}_{TA} = 1532.3$  m/s.

While estimating  $\tau^{-1}$  of Eq. (7), we adopted the following model provided by Minnich *et al.*:<sup>28</sup>

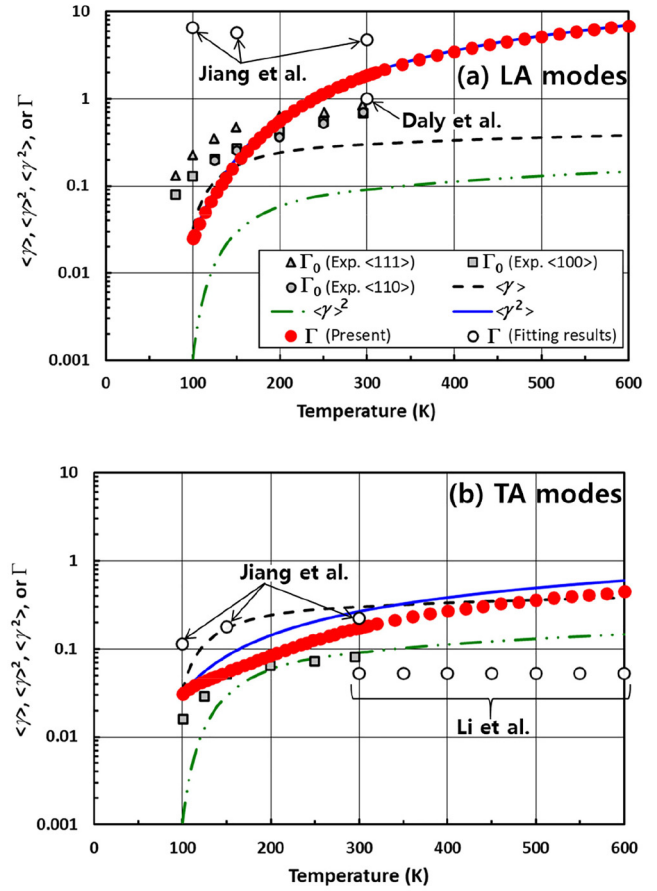
$$\tau_j^{-1} = \lambda \omega_j^2 T^\epsilon \exp(-\theta_j/T), \quad (8)$$

where  $\lambda = 2 \times 10^{-19}$  and  $\epsilon = 1.49$  for LA and  $\lambda = 1.2 \times 10^{-19}$  and  $\epsilon = 1.65$  for TA, and  $\theta_j = \hbar \omega_{\max,j}/k_B$ . While the application of alternative forms of the model for  $\tau$  is possible (for example, see Refs. 18, 29, and 30), determining physically rigorous values to the parameters involved in these models<sup>29,30</sup> for  $\tau$  is exceedingly challenging, especially, as a function of temperature, and for Zhu *et al.*'s model,<sup>18</sup> the inclusion of the phonon DOS in the model presents a significant challenge in accurately calculating  $\tau$  including temperature variations due to the nonlinearity of phonon DOS in frequency dependence. We can now derive an explicit expression for  $\langle \gamma_j^2 \rangle$  from Eq. (7). This yields a formula for the phonon frequency independent, as  $\omega_j^2$  cancels out during the calculation process.

To obtain  $\langle \gamma_j \rangle$ , we employed the results  $\langle \langle \gamma \rangle \rangle$  by Cuffari and Bongiorno<sup>31</sup> for Si. It is important to note that  $\langle \gamma_j \rangle$  is a weighted average of mode specific heat for both polarization and frequency and  $\langle \gamma \rangle = \langle \gamma_{LA} \rangle + \langle \gamma_{TA} \rangle$ . Given that the phonon specific heat for TA accounts for nearly 70% of the total phonon specific heat in Si over the temperature above 100 K<sup>30</sup> and that  $\gamma_{LA} > \gamma_{TA}$ ,<sup>32–34</sup> we can approximate  $\langle \gamma_{TA} \rangle = \langle \gamma_{LA} \rangle = \langle \gamma \rangle / 2$  with an approximation of linear combination between them. While this approach represents an apparent rough approximation, as can be seen in Fig. 1, it is evident that the magnitude of  $\langle \gamma_j \rangle$  for both LA and TA is  $\sim 0.1$ , indicating a value less than 1, resulting in  $\langle \gamma_j \rangle^2$  having a magnitude of  $\sim 0.01$ . Conversely, in the high-temperature range exceeding 300 K, where the Akhiezer effect is expected to be prominent,<sup>5</sup>  $\langle \gamma_j^2 \rangle$  attains a value of  $\sim 0.1$  for LA and  $\sim 1$  for TA. Consequently, this order of magnitude comparison shows that the variability in the value of  $\langle \gamma_j \rangle$  itself minimally affects the value of  $\Gamma_j$ , defined as  $\Gamma_j = \langle \gamma_j^2 \rangle - \langle \gamma_j \rangle^2$ , or subsequently the phonon relaxation time ( $\tau_{A,j}$ ) caused by Akhiezer damping. Furthermore, Cuffari and Bongiorno<sup>31</sup> only provided  $\langle \gamma \rangle$  up to 300 K. While the temperature dependence of the average  $\gamma$  is reported to vary among different research groups and analysis methods, the temperature dependence of the average  $\gamma$  is found to be very similar up to 900 or 1000 K.<sup>34,35</sup> Based on this fact, an extrapolation was employed here for temperatures above 300 K. Now to incorporate  $\tau_A$  into Eq. (1) for each polarization  $j$ , we use Matthiessen's rule:<sup>6,9</sup>  $\Lambda^{-1} = (\nu \tau_{L-R})^{-1} + (\nu \tau_A)^{-1}$ .

### III. RESULTS AND DISCUSSION

Now, we calculated the temperature-dependent variations of  $\Gamma_j$  together with relevant parameters ( $\langle \gamma_j \rangle$ ,  $\langle \gamma_j \rangle^2$ , and  $\langle \gamma_j^2 \rangle$ ), and



**FIG. 1.** Results of the calculation of  $\Gamma_j$  and the calculated results of the relevant factors used in the calculation of  $\Gamma_j$ : (a) for LA and (b) for TA. The experimental data  $\Gamma_0$  are from Ref. 33, which is the averaged Grüneisen parameter in each of three polarization directions averaged over 0.2–1.0 GHz frequency at 80–300 K. Results of Jiang *et al.*, Daly *et al.*, and Li and Cahill are from Refs. 6, 5, and 36, respectively. The legend in (b) corresponds to the one presented in (a).

they are shown in Fig. 1. Notably, the temperature dependence of the fitting results ( $\Gamma$ ) of Jiang *et al.*<sup>6</sup> is different. It might be attributed to their oversimplified methodology, which relies only on experimental thermal conductivity data and overlooks the averaging of phonon contributions across thermal phonon branches and directional characteristics.

Values for temperature-dependent  $\rho$  and  $C_j$  are required as inputs to calculate Eq. (5):  $\rho$  is taken from Ref. 37, and temperature-dependent  $C_j$  for acoustic phonons is computed following a formula introduced in Ref. 30, which is given by

$$C_j = \int \frac{\hbar^2 \omega_j^2 q_j^2 e^{\hbar \omega_j / k_B T} dq_j}{2\pi^2 k_B T^2 (e^{\hbar \omega_j / k_B T} - 1)^2}, \quad (9)$$

where  $q_j$  is the wave vector of given polarization  $j$ . Figure 2 shows the calculation results of  $\tau_{\text{inf}}$  of Eq. (6) for LA and TA compared

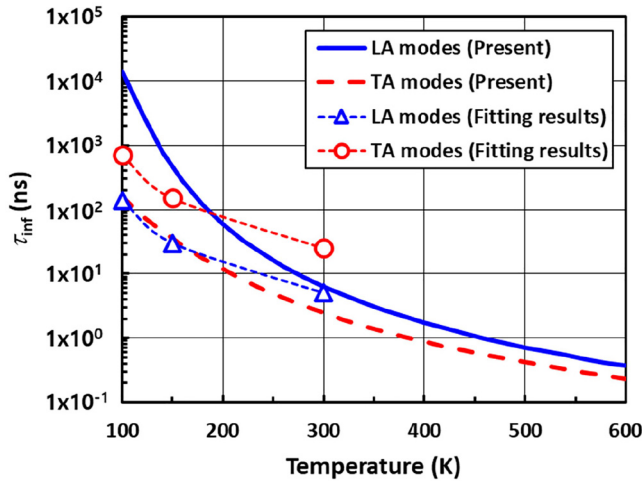


FIG. 2.  $\tau_{\text{inf}}$  as a function of temperature. Fitting results from Jiang *et al.*<sup>6</sup>

with the fitting results of Jiang *et al.*<sup>6</sup> When considering  $\tau_A \sim \tau_{\text{inf}}$ , it is evident that for low-frequency phonons related to Akhiezer damping,  $\tau_{\text{LA}} \gg \tau_{\text{TA}}$  at 100 K because  $\alpha_{\text{TA}} \gg \alpha_{\text{LA}}$  at low temperatures.<sup>10,38</sup> However, Jiang *et al.*<sup>6</sup> employed a simple approach:  $\tau_{\text{inf, TA}} = 5\tau_{\text{inf, LA}}$ .<sup>7</sup> With a more rigorous procedure in this study, we have demonstrated that  $\tau_{\text{inf, LA}}$  decreases more rapidly than  $\tau_{\text{inf, TA}}$  with increasing temperature, as illustrated in Fig. 2. This would be attributed to the participation of more excited high-energy phonons in the LA as an increase in the temperature.<sup>39</sup> It is clearly shown that as temperature increases, the accelerated reduction of  $\tau_{\text{inf, LA}}$  compared to  $\tau_{\text{inf, TA}}$  eventually leads to a decrease in the difference between them, as already reported in Ref. 10.

From the results of Eqs. (3) and (6), the calculated  $\tau_{A,j}$  of Eq. (2) allows us to examine the influence of temperature-induced phonon anharmonicity on phonon relaxation times with increasing temperature. As shown in Fig. 3, for both LA and TA, as expected, the decrease in phonon relaxation times is observed due to the temperature-increased phonon anharmonicity. However, in the case of LA, as indicated by the pink ellipse in Fig. 3(a), it is observed that the phonon relaxation time for 300 K is smaller than that for 500 K at the frequency  $< \sim 0.4$  THz. This difference is attributed to the utilization of distinct spectroscopy techniques: Minnich<sup>16</sup> employed thermal conductivity spectroscopy ( $T = 300$  K), while Hua and Minnich<sup>17</sup> utilized transient grating spectroscopy ( $T = 500$  K). This will be discussed later. For the case of  $T = 100$  K, since  $\tau_A > \tau_{\text{L-R}}$  holds throughout the entire phonon frequency range both for LA and TA, one can expect that the Akhiezer effect on phonon transport in Si can be safely negligible. To show this clearly, we calculated the accumulated and differential thermal conductivity ( $\Delta K$ ) with and without the Akhiezer damping as a function of phonon MFP and phonon frequency (see Appendix A). According to previous studies,<sup>10</sup> Akhiezer damping is dominant for phonons with frequencies much below  $\sim 0.1$  THz at 300 K, aligning closely with the present results for both LA and TA when considering the dotted curve shown in Fig. 3. However,

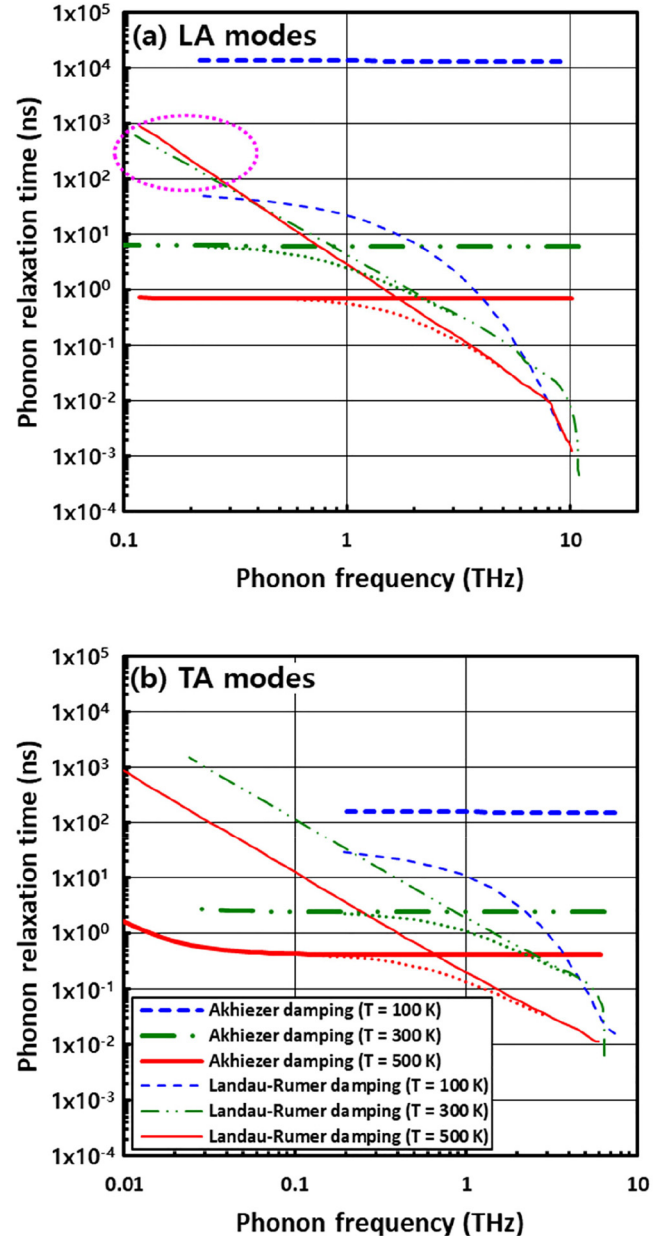


FIG. 3. Phonon relaxation time as a function of phonon frequency for Akhiezer damping and Landau-Rumer damping: (a) for LA and (b) for TA. The pink oval indicates the regions where  $\tau_{500\text{ K}} > \tau_{300\text{ K}}$ . The dotted curve shows a possible transition between the Akhiezer and Landau-Rumer regimes. The legend in (a) corresponds to the one presented in (b).

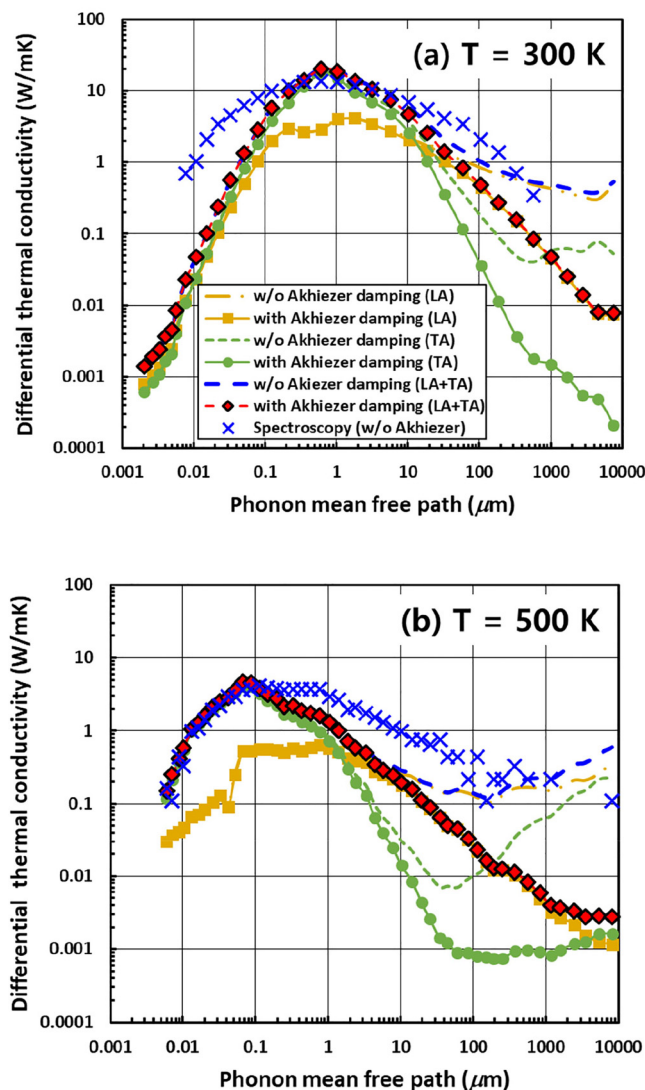
the impact of phonon anharmonicity on LA increases with frequency for L-R damping, even not significantly, while TA exhibit a more substantial impact of phonon anharmonicity compared to LA across the entire frequency spectrum. Analyzing the transition

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frequency from Akhiezer damping to L–R damping reveals that LA phonons experience an almost twofold increase, from 0.83 THz at 300 K to 1.54 THz at 500 K, while TA phonons exhibit a decrease from 0.81 THz at 300 K to 0.61 THz at 500 K. Therefore, with increasing temperature, the contribution of LA to thermal conductivity is anticipated to surpass that of TA in the Akhiezer regime.

To provide a more quantitative comparison, we present the phonon  $\Delta K$  as a function of phonon MFP at 300 and 500 K as shown in Fig. 4. For comparison, we also plotted the thermal



**FIG. 4.** Comparison of  $\Delta K$  between the thermal spectroscopy results and present results showing as a function of phonon MFP at 300 (a) and 500 K (b). Data of thermal spectroscopy are from Ref. 16 for  $T = 300$  K and Ref. 17 for  $T = 500$  K. The legend in (b) corresponds to the one presented in (a).

spectroscopy results<sup>16,17</sup> that do not consider the influence of Akhiezer damping. Regarding the thermal transport impact of Akhiezer damping, previous research by Liao *et al.*<sup>9</sup> indicated a negligible thermal contribution of Akhiezer damping in bulk Si at 300 K, while Jiang *et al.*<sup>6</sup> reported a 15 W/m K ( $\sim 10\%$ ) contribution. In this study, Fig. 4 shows that the thermal contribution of Akhiezer damping was 18.0 W/m K (12.6%) at 300 K [LA 3.4 W/m K (2.4%) and TA 14.6 W/m K (10.2%)], comparable to the results of Ref. 6, and 12.4 W/m K (16.8%) at 500 K [LA 7.6 W/m K (10.3%) and TA 4.8 W/m K (6.5%)].

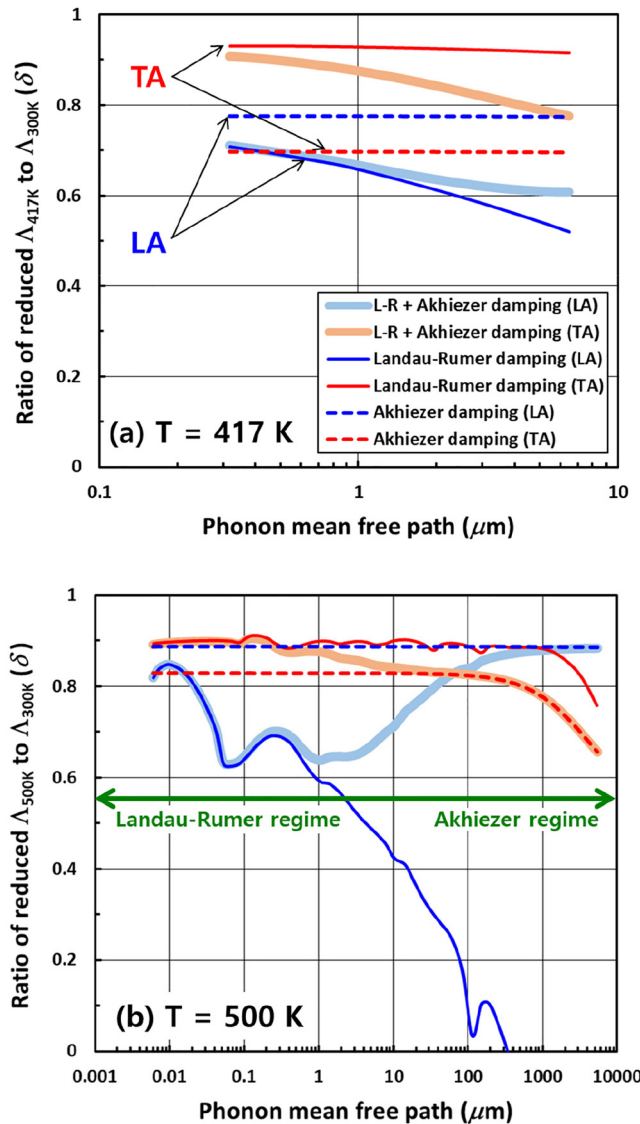
In Fig. 4(a), a significant difference between our results and spectroscopy is shown for  $MFP < \sim 0.2 \mu m$ . Minnich<sup>16</sup> showed a significant disparity between the “actual MFP distribution” provided by Minnich<sup>16</sup> and the thermal spectroscopy results below  $\sim 0.3$  THz.<sup>16</sup> It is believed that this error is reflected in Fig. 4(a). To investigate its frequency dependence, we calculate  $\Delta K$  as a function of phonon frequency with and without the Akhiezer damping at 300 and 500 K using the present method, and they are provided in Appendix B.

To further reveal the effect of temperature-induced anharmonicity in the phonons on L–R damping and Akhiezer damping, we calculated  $\delta = (\Lambda_{300 K} - \Lambda_T) / \Lambda_{300 K}$ , where the subscript  $T$  is 417 or 500 K, and the results are shown in Fig. 5. Due to the error mentioned before, i.e.,  $\tau_{500 K} > \tau_{300 K}$  at the frequency  $< \sim 0.4$  THz in Fig. 3(a), the phonon MFP in Fig. 5(b) exhibits a negative  $\delta$  above approximately  $300 \mu m$ . In this study, the interpretation of this aspect was entirely excluded to prevent confusion arising from such errors.

On a trial basis, we apply our method to applicable data from Ref. 40 conducted at 417 K in the confined MFP range of approximately  $0.3$ – $6.5 \mu m$ , and we calculated the parameter  $\delta$  [see Fig. 5(a)]. Although the results of Regner *et al.*<sup>40</sup> cover only a limited range of phonon MFP, it is evident from Fig. 5(a) that temperature-induced anharmonicity has a distinct impact in longer MFP regions where Akhiezer damping prevails.

To facilitate a more quantitative analysis, we calculated  $\delta$  using the data provided by Hua and Minnich,<sup>17</sup> which offered nearly complete frequency information. This result is depicted in Fig. 5(b), and we see that LA is  $\delta_{AK} \gg \delta_{L-R}$ , whereas TA is  $\delta_{AK} < \delta_{L-R}$ , concluding that LA is more affected by phonon anharmonicity than TA in the Akhiezer regime. In other words, for LA, phonon anharmonicity more significantly influences Akhiezer damping compared to L–R damping, leading to a considerable increase in the  $K$  contribution of LA if it remains in a nondiffusive transport regime where phonon scattering is not important. Conversely, for TA, phonon anharmonicity has a stronger effect on L–R damping than on Akhiezer damping, resulting in a relatively minor impact of phonon anharmonicity on Akhiezer damping. Consequently, the removal of Akhiezer damping does not lead to a substantial increase in the  $K$  of TA. As a result, as previously mentioned, the thermal contribution of Akhiezer damping increases from 12.6% at 300 K to 16.8% at 500 K, with LA making a significant contribution of 10.3%. However, due to differences in the methods employed by Regner *et al.*<sup>40</sup> and Hua and Minnich<sup>17</sup> it is not appropriate to compare Figs. 5(a) and 5(b) directly.

It is well known that, in bulk Si, the contributions to  $K$  from optical phonons having high-frequency as well as from low-frequency



**FIG. 5.** Ratio of the reduced phonon MFP to the phonon MFP at  $T = 300$  K as a function of phonon MFP at 417 (a) and 500 K (b). The legend in (b) corresponds to the one presented in (a).

phonons corresponding to the Akhiezer regime are not particularly significant.<sup>6–9</sup> However, in nanostructures such as nanowires, contributions of such high-frequency phonons to heat conduction have been observed to increase by up to 20% at 300 K due to the unaffected short MFP phonons.<sup>20</sup> Moreover, in thin film materials such as phononic crystals, the contribution of low-frequency acoustic phonons to heat conduction becomes significant due to the modification of the phonon DOS in low-dimensional materials.<sup>9</sup> Here, we newly uncovered a novel regime where the Akhiezer damping, previously deemed insignificant in the thermal conduction of bulk Si,<sup>6–9</sup> becomes crucial: it is at higher temperatures above 300 K.

#### IV. SUMMARY

In summary, Chiloyan *et al.*<sup>4</sup> have recently reported that non-thermal phonon transport in Si could exceed bulk heat conduction if the main phonon modes generated from the local heat source are low-frequency phonons with a long MFP at RT. The findings of Chiloyan *et al.*<sup>4</sup> are supported by the present results, suggesting that it is possible to achieve thermal conduction performance well beyond that of the bulk material when LA corresponding to Akhiezer damping occurs in a nonthermal phonon transport where there are no appreciable scatterings with other phonons. It is also observed that the temperature-induced phonon anharmonicity results in the range of phonon frequencies corresponding to the Akhiezer damping for LA widens, while for TA, it narrows in bulk Si. Additionally, for TA, anharmonicity notably influences L–R damping more than Akhiezer damping, while for LA, anharmonicity has a more pronounced effect on Akhiezer damping than on L–R damping. Consequently, the thermal conductivity was eliminated by 16.8% (12.4 W/m K) from  $K_{\text{bulk}}$  at 500 K, which seems nonnegligible in thermal phonon transport in bulk Si.

#### ACKNOWLEDGMENTS

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (Grant No. 2022R1F1A1068162).

#### AUTHOR DECLARATIONS

##### Conflict of Interest

The authors have no conflicts to disclose.

#### Author Contributions

**Jae Sik Jin:** Conceptualization (lead); Data curation (lead); Formal analysis (lead); Funding acquisition (lead); Investigation (lead); Methodology (lead); Project administration (lead); Resources (lead); Software (lead); Supervision (lead); Validation (lead); Visualization (lead); Writing – original draft (lead); Writing – review & editing (lead).

#### DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

#### APPENDIX A: THERMAL CONDUCTIVITY BY THERMAL PHONON TRANSPORT AT 100 K

In Fig. 3 in the main text, it is shown that  $\tau_A > \tau_{L-R}$  holds throughout the entire phonon frequency range for both LA and TA at  $T = 100$  K. From this, we can expect that the Akhiezer effect on phonon transport in Si can be safely negligible. To confirm this, we calculated the differential thermal conductivity with and without the Akhiezer damping as a function of phonon MFP and phonon frequency. The results are shown in Fig. 6.

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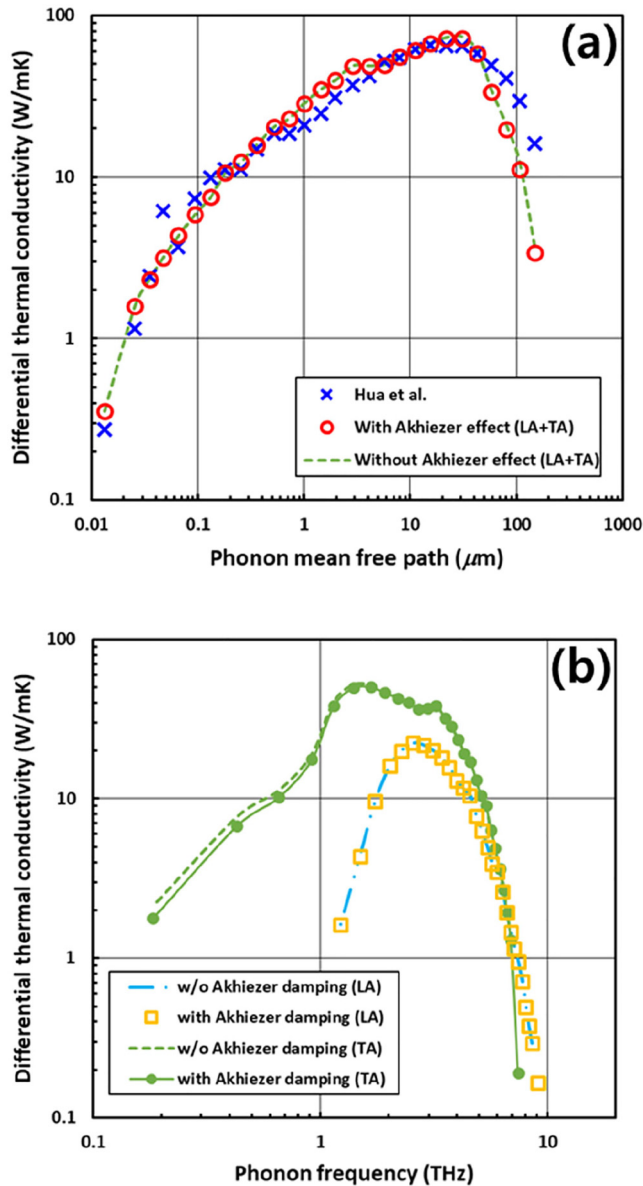


FIG. 6. Differential thermal conductivity with and without the Akhiezer damping as a function of phonon MFP (a) and phonon frequency (b). The results of Hua and Minnich are taken from Ref. 17 in the main text.

## APPENDIX B: FREQUENCY DEPENDENCE OF AKHIEZER DAMPING AT 300 AND 500 K

We calculated the differential thermal conductivity as a function of phonon frequency with and without the Akhiezer damping at 300 and 500 K using the present method to investigate the frequency dependency of the Akhiezer damping effect. The results are shown in Fig. 7.

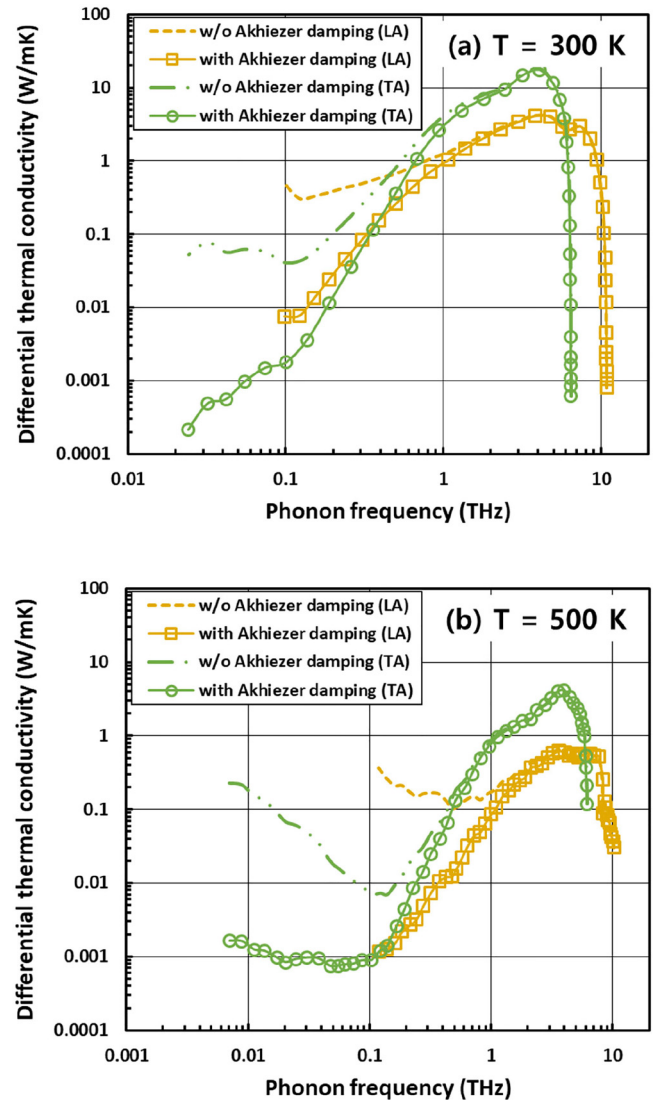


FIG. 7. Differential thermal conductivity as a function of phonon frequency with and without the Akhiezer damping at 300 (a) and 500 K (b).

## REFERENCES

- <sup>1</sup>D. S. Kim, O. Hellman, N. Shulumba, C. N. Saunders, J. Y. Y. Lin, H. L. Smith, J. E. Herriman, J. L. Niedziela, D. L. Abernathy, C. W. Li, and B. Fultz, "Temperature-dependent phonon lifetimes and thermal conductivity of silicon by inelastic neutron scattering and *ab initio* calculations," *Phys. Rev. B* **102**(17), 174311 (2020).
- <sup>2</sup>G. Chen, "Non-Fourier phonon heat conduction at the microscale and nanoscale," *Nat. Rev. Phys.* **3**(8), 555–569 (2021).
- <sup>3</sup>J. M. S. Gordillo, A. Morata, C. D. Sierra, M. Salleras, L. Fonseca, and A. Tarancón, "Recent advances in silicon-based nanostructures for thermoelectric applications," *APL Mater.* **11**(4), 040702 (2023).
- <sup>4</sup>V. Chiloyan, S. Huberman, A. A. Maznev, K. A. Nelson, and G. Chen, "Thermal transport exceeding bulk heat conduction due to nonthermal micro/nanoscale phonon populations," *Appl. Phys. Lett.* **116**(16), 163102 (2020).

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- <sup>5</sup>B. C. Daly, K. Kang, Y. Wang, and D. G. Cahill, "Picosecond ultrasonic measurements of attenuation of longitudinal acoustic phonons in silicon," *Phys. Rev. B* **80**(17), 174112 (2009).
- <sup>6</sup>P. Jiang, L. Lindsay, and Y. K. Koh, "Role of low-energy phonons with mean-free-paths  $>0.8\ \mu\text{m}$  in heat conduction in silicon," *J. Appl. Phys.* **119**(24), 245705 (2016).
- <sup>7</sup>A. A. Maznev, J. A. Johnson, and K. A. Nelson, "Onset of nondiffusive phonon transport in transient thermal grating decay," *Phys. Rev. B* **84**(19), 195206 (2011).
- <sup>8</sup>A. A. Maznev, "Onset of size effect in lattice thermal conductivity of thin films," *J. Appl. Phys.* **113**(11), 113511 (2013).
- <sup>9</sup>Y. Liao, T. Shiga, M. Kashiwagi, and J. Shiomi, "Akhiezer mechanism limits coherent heat conduction in phononic crystals," *Phys. Rev. B* **98**(13), 134307 (2018).
- <sup>10</sup>M. Markov, J. Sjakste, N. Vast, R. Legrand, B. Perrin, and L. Paulatto, "Breakdown of Herring's processes in cubic semiconductors for subterahertz longitudinal acoustic phonons," *Phys. Rev. B* **98**(24), 245201 (2018).
- <sup>11</sup>A. Minnich and G. Chen, "Modified effective medium formulation for the thermal conductivity of nanocomposites," *Appl. Phys. Lett.* **91**(7), 073105 (2007).
- <sup>12</sup>J. S. Jin, "Investigation of the full spectrum phonon lifetime in thin silicon films from the bulk spectral phonon mean-free-path distribution by using kinetic theory," *J. Korean Phys. Soc.* **70**(5), 480–488 (2017).
- <sup>13</sup>P. G. Klemens, "Theory of lattice thermal conductivity: Role of low-frequency phonons," *Int. J. Thermophys.* **2**(1), 55–62 (1981).
- <sup>14</sup>G. Chen, *Nanoscale Energy Transport and Conversion: A Parallel Treatment of Electrons, Molecules, Phonons, and Photons* (Oxford University Press, 2005).
- <sup>15</sup>R. Legrand, A. Huynh, B. Jusserand, B. Perrin, and A. Lemaître, "Direct measurement of coherent subterahertz acoustic phonons mean free path in GaAs," *Phys. Rev. B* **93**(18), 184304 (2016).
- <sup>16</sup>A. J. Minnich, "Determining phonon mean free paths from observations of quasiballistic thermal transport," *Phys. Rev. Lett.* **109**(20), 205901 (2012).
- <sup>17</sup>C. Hua and A. J. Minnich, "Transport regimes in quasiballistic heat conduction," *Phys. Rev. B* **89**(9), 094302 (2014).
- <sup>18</sup>Z. Zhu, D. A. Romero, D. P. Sellan, A. Nabovati, and C. H. Amon, "Assessment of the Holland model for silicon phonon-phonon relaxation times using lattice dynamics calculations," *J. Appl. Phys.* **113**(17), 173511 (2013).
- <sup>19</sup>D. S. Kim, H. L. Smith, J. L. Niedziela, C. W. Li, D. L. Abernathy, and B. Fultz, "Phonon anharmonicity in silicon from 100 to 1500 K," *Phys. Rev. B* **91**(1), 014307 (2015).
- <sup>20</sup>Z. Tian, K. Esfarjani, J. Shiomi, A. S. Henry, and G. Chen, "On the importance of optical phonons to thermal conductivity in nanostructures," *Appl. Phys. Lett.* **99**(5), 053122 (2011).
- <sup>21</sup>J. E. Turney, A. J. H. McGaughey, and C. H. Amon, "Assessing the applicability of quantum corrections to classical thermal conductivity predictions," *Phys. Rev. B* **79**(22), 224305 (2009).
- <sup>22</sup>J. V. Goicochea, M. Madrid, and C. Amon, "Thermal properties for bulk silicon based on the determination of relaxation times using molecular dynamics," *J. Heat Transfer* **132**(1), 012401 (2010).
- <sup>23</sup>J. S. Jin, "Temperature-dependent thermal phonon relaxation time and its influence on ballistic-diffusive heat transport in SOI transistor," *Phys. B* **657**, 414828 (2023).
- <sup>24</sup>W. P. Mason and T. B. Bateman, "Ultrasonic-wave propagation in pure silicon and germanium," *J. Acoust. Soc. Am.* **36**(4), 644–652 (1964).
- <sup>25</sup>C. J. Morath and H. J. Maris, "Phonon attenuation in amorphous solids studied by picosecond ultrasonics," *Phys. Rev. B* **54**(1), 203–213 (1996).
- <sup>26</sup>G. Klemens, in *Thermal Conductivity*, edited by R. P. Tve (Academic, London, 1969), Vol. 1.
- <sup>27</sup>A. Balandin and K. L. Wang, "Significant decrease of the lattice thermal conductivity due to phonon confinement in a free-standing semiconductor quantum well," *Phys. Rev. B* **58**(3), 1544–1549 (1998).
- <sup>28</sup>A. J. Minnich, G. Chen, S. Mansoor, and B. Yilbas, "Quasiballistic heat transfer studied using the frequency-dependent Boltzmann transport equation," *Phys. Rev. B* **84**(23), 235207 (2011).
- <sup>29</sup>Y.-J. Han and P. G. Klemens, "Anharmonic thermal resistivity of dielectric crystals at low temperatures," *Phys. Rev. B* **48**(9), 6033–6042 (1993).
- <sup>30</sup>S. V. J. Narumanchi, J. Y. Murthy, and C. H. Amon, "Submicron heat transport model in silicon accounting for phonon dispersion and polarization," *J. Heat Transfer* **126**(6), 946–955 (2004).
- <sup>31</sup>D. Cuffari and A. Bongiorno, "Calculation of mode Grüneisen parameters made simple," *Phys. Rev. Lett.* **124**(21), 215501 (2020).
- <sup>32</sup>E. T. Ritz, S. J. Li, and N. A. Benedek, "Thermal expansion in insulating solids from first principles," *J. Appl. Phys.* **126**(17), 171102 (2019).
- <sup>33</sup>R. Nava, M. P. Vecchi, J. Romero, and B. Fernández, "Akhiezer damping and the thermal conductivity of pure and impure dielectrics," *Phys. Rev. B* **14**(2), 800–807 (1976).
- <sup>34</sup>A. P. Bartók, J. Kermode, N. Bernstein, and G. Csányi, "Machine learning a general-purpose interatomic potential for silicon," *Phys. Rev. X* **8**(4), 041048 (2018).
- <sup>35</sup>H. Babaei, R. Guo, A. Hashemi, and S. Lee, "Machine-learning-based interatomic potential for phonon transport in perfect crystalline Si and crystalline Si with vacancies," *Phys. Rev. Mater.* **3**(7), 074603 (2019).
- <sup>36</sup>D. Li and D. G. Cahill, "Attenuation of 7 GHz surface acoustic waves on silicon," *Phys. Rev. B* **94**(10), 104306 (2016).
- <sup>37</sup>Y. S. Touloukian and E. H. Buyco, "Thermophysical properties of matter," in *Specific Heat: Nonmetallic Solids* (IFI/Plenum, New York, 1970), Vol. 2, p. 166.
- <sup>38</sup>G. G. Sahasrabudhe and S. D. Lambade, "Temperature dependence of the collective phonon relaxation time and acoustic damping in Ge and Si," *J. Phys. Chem. Solids* **60**(6), 773–785 (1999).
- <sup>39</sup>T. Feng, X. Yang, and X. Ruan, "Phonon anharmonic frequency shift induced by four-phonon scattering calculated from first principles," *J. Appl. Phys.* **124**(14), 145101 (2018).
- <sup>40</sup>K. T. Regner, D. P. Sellan, Z. Su, C. H. Amon, A. J. H. McGaughey, and J. A. Malen, "Broadband phonon mean free path contributions to thermal conductivity measured using frequency domain thermoreflectance," *Nat. Commun.* **4**, 1640 (2013).