

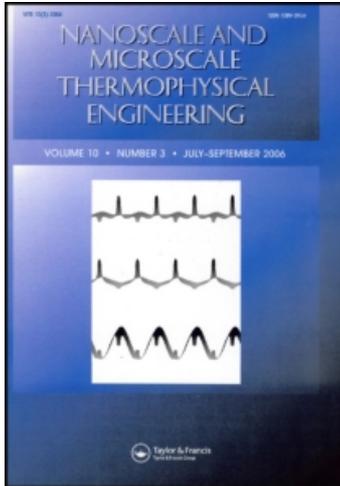
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PHONON SCATTERING AND VELOCITY CONSIDERATIONS IN THE MINIMUM PHONON THERMAL CONDUCTIVITY OF LAYERED SOLIDS ABOVE THE PLATEAU

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The minimum thermal conductivity of amorphous solids is investigated by implementing Einstein's original assumption of scattering at every lattice point within the framework of nonlocalized coupled oscillators. By separating the phase velocity and group velocity, thermal conductivity predictions agree well with experimental results indicating that it is necessary to account for both energy transport via hopping of localized modes (fractons) as well propagation of nonlocalized modes (phonons). The further reduction of thermal conductivity in layered structures is then examined by considering the scattering rates by atoms with a different binding force within the context of these minimum conductivity models.

KEY WORDS: minimum thermal conductivity, phonons, interfaces, disorder, layered structures

INTRODUCTION

Thermal properties of amorphous and disordered solids have been the central topic of many theoretical studies as a means to decipher the exact phonon mechanisms contributing to the experimental anomalies observed in the heat capacity at low temperatures and the trends in thermal conductivity at various temperatures. At high temperatures, above the plateau, models for the thermal conductivity of amorphous and disordered solids are typically based upon the idea of a minimum thermal conductivity [1] in which the mean free path, l , is restricted to the interatomic spacing of solid, as originally proposed by Einstein [2]. In this model, Einstein assumes that thermal energy propagates via a random walk between localized, uncoupled atomic oscillators all vibrating at the same frequency and transferring energy at a rate of one half of the period of the oscillation. To remove the uncertainty inherent in choosing a frequency of this atomic oscillation (i.e., the Einstein frequency), this idea was later

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extended to include oscillators having a spectrally dependent size ($\lambda/2$) that responded in a Debye fashion by Cahill and Pohl (CP model) [3]. The CP model assumes, like Einstein, that propagating modes are scattered at a rate of one half of the period of their oscillation, but unlike Einstein, due to their spectrally dependent size, do not necessarily scatter at every lattice site. In the minimum limit, this difference removes much of the localized energy propagation inherent in the original Einstein model even as several studies have shown that both localized (fractons) as well as propagating (phonons) modes can contribute to the thermal conductivity of amorphous solids [4–8]. In addition, recent experimental [9, 10] and theoretical [11] investigations have shown that layered materials can further reduce the thermal conductivity below the CP limit, thus raising questions regarding the effects of the localized contribution as such solids approach the lower limit to thermal conductivity.

In response, the theoretical minimum to thermal conductivity for layered materials is investigated at temperatures above the plateau by implementing the assumptions originally applied by Einstein in the framework of the CP model in order to leverage the advantages of each. Specifically, by assuming that all modes scatter at a rate of one half of their period of oscillation and that these events are separated in the lower limit by the lattice spacing, a response that exhibits characteristics of energy propagation via fracton- and phonon-like behavior is demonstrated. Such an approach leads to a linear temperature trend in heat capacity at low temperatures and improvement in the temperature trend of the thermal conductivity for amorphous solids above the plateau. Subsequently, the minimum conductivity model derived in this work is then applied to layered materials by considering scattering rates driven by different binding forces as a means to investigate materials having experimentally measured conductivities below the lower limit derived by the CP method.

BACKGROUND

The basis of the minimum models for thermal conductivity is derived from the standard kinetic theory expression given by

$$\Lambda = \frac{1}{3} \int_{\omega} C_{\omega} v_g l d\omega, \quad (1)$$

where C_{ω} is the spectral heat capacity, v_g is the group velocity, and ω is the angular phonon frequency. The CP model assumes that $l = v_g \tau$, where τ is the minimum scattering time, which is assumed as one half of the period of oscillation, $\tau = \pi/\omega$. Therefore, taking a Debye assumption, the CP model is given by [3, 12]:

$$\Lambda_{CP} = \left(\frac{\pi}{6}\right)^{1/3} k_B n^{2/3} \sum_j v_{g,j} \left(\frac{T}{\Theta_j}\right)^2 \int_0^{\Theta_j/T} \frac{x^3 \exp[x]}{(\exp[x] - 1)^2} dx, \quad (2)$$

where k_B is the Boltzmann constant, n is the atomic density, and T is the temperature. Θ_j is the Debye temperature, which for each polarization j is given by

$$\Theta_j = v_{g,j} \hbar (6\pi^2 n)^{1/3} / k_B, \text{ thereby implicitly defining the material as a Debye solid}$$

[13]. For the solids considered in this work, j represents 1 longitudinal and 2 transverse polarizations. Equation (1) gives a T^2 temperature dependency of thermal conductivity at low temperatures where amorphous solids exhibit a temperature dependency of around $T^{1.8}$ [14]. Although the CP model gives approximately the correct temperature dependency of thermal conductivity, it is derived under that assumption of a Debye heat capacity [1] that yields a T^3 dependence at low temperatures. However, amorphous solids exhibit a low-temperature heat capacity that is linear with temperature (i.e., $C \propto T$) [15].

This linear trend with temperature has been associated with scattering off of tunneling states (TS) that allow for atoms to have two equilibrium positions and, therefore, must be considered as a localized phenomenon [16, 17]. With such a perspective, predictions of the specific heat in amorphous solids must then somehow capture this localized nature. Additionally, as previously mentioned, the thermal conductivity above the plateau has been associated with a combination of fracton hopping and phonon diffusion further necessitating a model that captures each of these contributions.

Localized Vibrations in the Minimum Thermal Conductivity

To capture each of these aspects of the transport, begin by considering, as do both the Einstein and the CP models, that heat-carrying modes scatter at a time rate of one half their period of oscillation; i.e., $\tau = \pi/\omega$. In addition, assume, like Einstein but unlike the CP model, that these scattering events are, in the lower limit, separated by the interatomic spacing; i.e., $l = n^{-1/3}$. The ramifications of these assumptions make it imperative that the phase velocity be reconsidered. Specifically, because the time between scattering events is given by τ and the distance between these events is l , phase information may then be deduced to be carried at a rate of $\nu_p = l/\tau = \pi/(n^{1/3}\omega)$, where ν_p is the phase velocity. In a material exhibiting Debye characteristics like that assumed in the CP model, however, the energy is assumed to propagate at the speed of sound within the material and, as such, the group velocity ν_g will no longer be equal to the phase velocity ($\nu_g \neq \nu_p$) as is typically assumed. This fact, in turn, will cause a modification in the density of states.

The density of states per unit volume in three dimensions is given by $D = q^2 dq / (2\pi^2)$ where q is the wavevector. Converting to frequency space, and recognizing that the phase velocity, $\nu_p \equiv \omega/q$, is different from the group velocity, $\nu_g \equiv \partial\omega/\partial q$, yields a spectral density of states given by $D = \omega^2 d\omega / (2\pi^2 \nu_p^2 \nu_g)$. Given the atomic spacing limited phase velocity in the minimum limit, the density of states becomes $D = n^{2/3} d\omega / (2\nu_g)$. This result, like the TS model, exhibits a density of states that is spectrally constant.

Note that the phase velocity, which is termed here as an *atomic-spacing limited phase velocity*, specifies the rate in which phase coherency is destroyed in the minimum limit. This is related to the spatial extent of the crystal coherency (which is the interatomic

spacing in the minimal limit). The assumption of a group velocity in the solid, which in this analysis is defined as spectrally constant (Debye), allows the model to account for the incoherent coupling of the oscillators driving thermal diffusion (phonon transport). Although more rigorous methods could be used to determine the spectral phonon velocity in a heavily disordered solid, such as molecular dynamics simulations, defining both an atomic-spacing limited phase velocity to capture the contribution of localized fracton hopping and a group velocity to describe the incoherent coupled oscillators driving phonon diffusion accounts for much of the unique lattice properties of amorphous solids.

Using these definitions for the phase and group velocities, the heat capacity is calculated by $C = \int \hbar\omega D(\partial f/\partial T) d\omega$ where f is the Bose-Einstein distribution. By then taking into account an atomic spacing limited phase velocity and the resulting density of states obtained above, the heat capacity for amorphous materials in the lower limit may be described as:

$$C = \left(\frac{3\pi^2}{8}\right)^{1/3} n k_B \sum_j \frac{T}{\Theta_j} \int_0^{\Theta_j/T} \frac{x^2 \exp[x]}{(\exp[x] - 1)^2} dx \quad (3)$$

which, at low temperatures, yields $C \propto T$. Note that although the trends in Eq. (3) agree with low-temperature data [15], the values do not match. However, as the theory developed in this work is applied to results above the plateau, it is expected that the calculations will not capture many of the quantum aspects characteristic of glassy transport at temperature below the plateau. The change in the trends in heat capacity from the typical Debye or CP models' low-temperature trends stems from the spectrally constant phonon density of states derived from the interatomic spacing limited phase velocity; again, this spectrally constant density of states is consistent with the TS model.

Consider now the minimum conductivity in an amorphous material. Given that energy is transferred by scattering of phonons at every lattice point, the mean free path is given by $l = v_p \tau = n^{-1/3}$ so that Eq. (1) becomes $\Lambda = \frac{1}{3} \int_{\omega} C_{\omega} v_g v_p \tau d\omega$. In the minimum limit, the thermal conductivity then becomes

$$\Lambda_{MOD} = \left(\frac{\pi^2}{36}\right)^{1/3} n^{2/3} k_B \sum_j \frac{T}{\Theta_j} v_{g,j} \int_0^{\Theta_j/T} \frac{x^2 \exp[x]}{(\exp[x] - 1)^2} dx \quad (4)$$

which was presented previously without derivation [11]. Equation (4) assumes that the heat is propagating through the solid at a group velocity that is different than the phase velocity that is limited by the atomic oscillations. Equation (4) is linearly proportional to T at low temperatures. Previous studies have theorized that the thermal conductivity of amorphous solids is due to a combination of fracton hopping leading to a linear dependence on T at low temperatures and phonon transport leading to a T^2 dependence [18]. Equation (4) accounts for fracton hopping through the phase velocity considerations and phonon propagation through the group velocity, whereas the CP model treats thermal transport purely as phonon propagation.

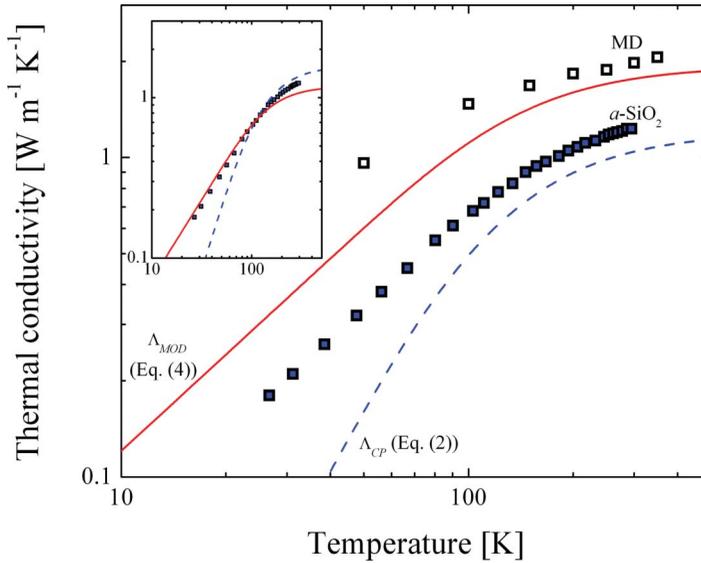


Figure 1. Measured thermal conductivity of $a\text{-SiO}_2$ [19] compared to MD simulations [4] and the lower limit thermal conductivity predictions using the CP model (dashed lines) and the model given by Eq. (4) (solid lines). The MD simulations on $a\text{-SiO}_2$ account for localized modes, which explains the agreement between Eq. (4) and the MD results. Capturing the measured thermal conductivity in the measured $a\text{-SiO}_2$, meanwhile, necessitates some combination of the two models. For model calculations, the longitudinal and transverse phonon velocities were taken as 5,800 and 3,700 m s^{-1} , respectively, and the atomic density was $6.61 \times 10^{28} \text{ m}^{-3}$ [19]. (inset) Models normalized to the data at $T = 110 \text{ K}$. The linear trend in thermal conductivity of $a\text{-SiO}_2$ is dominant and captured by the minimum conductivity model derived in this work up to temperatures approaching the Debye temperature ($T < 100\text{K}$), whereas at higher temperatures, the trends are captured by some combination of both the CP model and Eq. (4).

Figure 1 shows calculations of Λ_{CP} and Λ_{MOD} from this work as a function of temperature for $a\text{-SiO}_2$ along with experimental data [19] and molecular dynamics (MD) simulations [4]. The experimentally obtained thermal conductivities show qualitative trends with temperature that are similar to the model in this work, whereas the quantitative values seem to be described by some fraction of both the current and the CP models. In addition, the MD simulations indicate that thermal transport in this amorphous glass is due to both propagating (phonons) and hopping of the localized modes (fractons) in $a\text{-SiO}_2$ (see Shenogin et al. [4]). Because the current approach accounts for this localized contribution in addition to the phonon propagation, it is more capable of describing the results of this simulation than the CP model.

To compare the qualitative trends in the thermal conductivity apart from the quantitative offsets between the models and data, the inset of Figure 1 shows the results normalized to their value at $T = 110 \text{ K}$. The trend in thermal conductivity of $a\text{-SiO}_2$ is dominant and captured by the minimum conductivity model derived in this work up to temperatures approaching the Debye temperature ($T < 100\text{K}$). At higher temperatures, however, the trends seem to be described by a combination of both the CP model and the current approach (Eq. (4)). This indicates that localized fracton hopping still dominates the thermal conductivity in amorphous materials even above the plateau, with the contribution from diffusive phonon transport strongly influencing thermal conductivity at temperatures approaching and above the Debye temperature.

Minimum Thermal Conductivity of Layered Solids

The necessity of re-examining the CP approach extends to layered materials as well due to recent studies of such solids that have exhibited thermal conductivities below the limit proposed by this model [9, 10]. For such structures, it is necessary to account for bonding in, and between, the layers while determining the velocities and scattering times that led to the resulting thermal conductivity. In the minimum limit for thermal conductivity, the phase velocity is expressed as $v_p = \omega / \left(n^{1/3} \pi \right)$ as previously discussed, and the density of states becomes isotropic and spectrally constant. However, the layered nature of the solid will influence the scattering time. Hopkins and Piekos [11] accounted for this modified scattering time in the localized minimum conductivity model by taking into account scattering by atoms with a different binding force, as originally proposed by Klemens [20]. In this work, the minimum scattering time by an atom bound by different forces must be re-derived to account for different phase and group velocities utilized to predict the minimum conductivity.

Recasting Klemens' [20] original expression for relaxation time on a per polarization and per frequency basis yields [11]:

$$\frac{1}{\tau_j} = 8\pi V N c_{qq',j}^2 \frac{1}{M^2 \omega^2} \frac{\pi}{(2\pi)^3} \frac{dq'}{d\omega} q'^2 \quad (5)$$

where V is the volume of the crystal, N is the number of atoms in the volume, c is the scattering matrix element in the perturbation Hamiltonian, and M is the atomic mass. Assuming conservation of energy, $\omega = \omega'$, and recognizing a difference between the phase and group velocities yields

$$\frac{1}{\tau_j} = \frac{VN}{\pi M^2} \frac{1}{v_{pj}^2 v_{gj}} c_j^2. \quad (6)$$

The key then is to determine c , which in the case of layered solids where atomic oscillations are driven by different binding forces should be related to the changes in energy due to atoms scattering by an atom with a different binding force.

Following a previous derivation [11], the contribution of normal linkages to the unperturbed Hamiltonian of an oscillator is

$$H_j = -\frac{1}{2} \frac{\hbar}{\omega} \left(\frac{v_{g,j}}{a} \right)^2 (qa)^2 \quad (7)$$

so that the Hamiltonian of the oscillator when perturbed by an atom with a different binding force is

$$\dot{H}_j = -\frac{\hbar q^2}{\omega} v_{g,j} \delta v_{g,j} \quad (8)$$

where $\delta v_{g,j}$ is the difference in phonon velocities in the layers and between the layers; $\delta v_{g,j}$ represents the difference between the intra- and intersheet atomic binding forces

since $v_g \propto \sqrt{K}$, where K is the spring constant binding two atoms. The elements of the perturbation energy matrix take the form of $H_j = \hbar N c_j / (M\omega)$ so that the coefficient due to scattering by an atom with a different binding force is

$$c_j = -\frac{M\omega^2}{N v_p^2} v_{g,j} \delta v_{g,j}^*. \quad (9)$$

Inserting Eq. (9) into Eq. (5) yields the scattering rate of a phonon by atoms with different binding forces (*bf*)

$$\frac{1}{\tau_{bf,j}} = \frac{1}{n} \frac{v_{g,j}}{v_{p,j}} \left(\frac{\delta v_{g,j}^*}{v_{p,j}} \right)^2 \frac{\omega^4}{\pi v_{p,j}^3}. \quad (10)$$

In the case of the CP model when $v_p = v_g$, Eq. (10) reduces to the scattering rate derived by Hopkins and Piekos [11].

To determine the minimum scattering rates, first Eq. (10) must be modified to reflect that in the minimum limit $v_p = \omega / (n^{1/3} \pi)$, which yields

$$\frac{1}{\tau_{bf,j}} = \frac{\pi^5 n}{\omega^2} v_{g,j} (\delta v_{g,j})^2. \quad (11)$$

Therefore, in a layered solid, there are two scattering phenomena affecting the minimum conductivity—the minimum scattering time with similar atoms in the layer (π/ω) and the minimum scattering time described by Eq. (11) causing phonon propagation between layers. Therefore,

$$\frac{1}{\tau_{bf,\min,j}} = \frac{\pi^5 n}{\omega^2} v_{g,j} \left(v_{g,j} - \frac{\omega}{n^{1/3} \pi} \right)^2 \quad (12)$$

because the group velocity will propagate modes through and between the layers, but in the minimum limit, the phonons modes are scattered at a distance of the interatomic spacing. Therefore, the total minimum scattering rate is given by

$$\tau_{\min,\text{total},j}^{-1} = \omega/\pi + \tau_{bf,j}^{-1}. \quad (13)$$

In the derivation of Eq. (12), it is assumed that the oscillations between the layers are affecting the scattering within the layers. Therefore, this approach applies when the layers are sufficiently small and disordered so that minimum scattering occurs within the layers but not so large that the layers and boundaries can be treated independently in a thermal resistor-type model. For example, consider thermally bulk layers so that the atomic bonds at the layer boundaries do not affect phonon mode propagation in the layers. In this case, $\delta v_{g,j}$ in the layers are zero and the total minimum scattering time reduces to π/ω . Note that a boundary scattering rate cannot be constructed in the CP model unless the group velocities in the different layers are independent of each other, which would treat the propagation as thermally bulk.

If interfacial oscillations are not affecting the transport in the layers, then the minimum conductivity models should set a lower limit to thermal conductivity without having to take into account scattering at the layer interfaces. However, the recent measurement on WSe₂ layered structures show a significant reduction beyond the predicted minimum thermal conductivity [10]. Scattering by atoms with a different binding force at the layers' interfaces (Eq. (12)) implies that scattering at interfaces increases the total scattering rate, leading to a decrease in thermal conductivity, which is in line with the recent results on the WSe₂ layered film.

To determine the minimum conductivity, the averaged phase velocity must be determined. In Eq. (12), the velocities were considered independently as per Matthiessen's rule, but the averaged phase velocity of phonon propagation in anisotropic structures is inversely related to the total minimum scattering time given by Eq. (13), so that the minimum thermal conductivity of a layered solid is given by

$$\Lambda_{MOD,layered} = \frac{n^{1/3}}{6\pi^2} \sum_j \int_0^{\omega_{max,j}} \frac{\hbar^2 \omega^4}{k_B T^2} \frac{\exp\left[\frac{\hbar\omega}{k_B T}\right]}{\left(\exp\left[\frac{\hbar\omega}{k_B T}\right] - 1\right)^2} \left(\frac{\omega}{\pi} + \pi^2 \omega \left(1 - \frac{\pi v_{g,j} n^{1/3}}{\omega}\right)\right)^{-2} d\omega \quad (14)$$

Figure 2 shows the minimum cross-plane thermal conductivities of WSe₂ reported in Chiritescu et al. [10]. For comparison, thermal conductivities predicted by the CP model (Eq. (2)) along with the current approach in the limit of a nonlayered (Eq. (4)) as well as a layered solid (Eq. (14)) are given. The predictions are carried out utilizing cross-plane velocities and atomic number density of the WSe₂ films that are given in Chiritescu et al. [10]. The predictions for minimum conductivity in a nonlayered solid agree well with the data on the ion-irradiated film, because the ion bombardment destroyed the layered structure of the WSe₂ sheets. However, the measured conductivities on the films with the layered structure still intact have further reduction beyond the nonlayered limit. Taking into account boundary scattering using the layered model in Eq. (14) causes a reduction in the conductivity minimum to values that agree well with the measured WSe₂ data, indicating that the scattering by atoms with a different binding force is an important parameter in reduction of thermal conductivity.

The binding force scattering is a mechanism that can be used in designing low-thermal-conductivity materials such as material systems used in thermoelectric applications, which require a high electrical conductivity yet a very low thermal conductivity. Superlattice systems exploit this phonon-scattering mechanism by an atom with a different binding force, so that the minimum conductivity of a superlattice can be determined with Eq. (14). Consider, for example, the Si/Ge superlattices measured by Lee et al. [21]. The lowest room temperature thermal conductivity of these superlattices was $\sim 2 \text{ W m}^{-1} \text{ K}^{-1}$. However, the lower limit to thermal conductivity of a Si/Ge superlattice is $0.15 \text{ W m}^{-1} \text{ K}^{-1}$. This lower limit was approximated by averaging the phonon velocities and atomic densities of *a*-Si and *a*-Ge [12] for calculations in Eq. (14). This implies that thermal conductivity of an Si/Ge superlattice can be decreased by an additional order of magnitude through careful fabrication procedures, such as deposition techniques to make the Si and Ge layers amorphous or by inducing disorder to maximize scattering at the interfaces, which has been shown to further reduce thermal conductivity [22, 23].

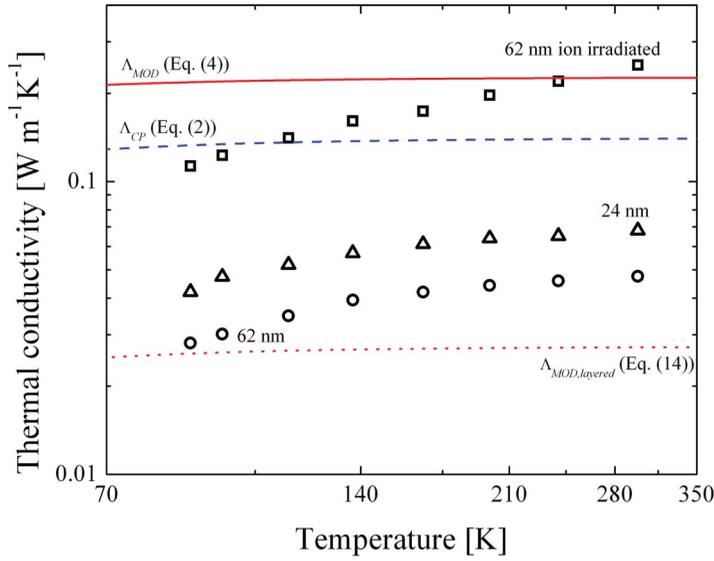


Figure 2. Measured thermal conductivity of various WSe₂ films [10] along with the minimum conductivity predictions of the homogeneous and layered solids. The ion-irradiated film lost its layered structure due to ion bombardment breaking the interlayer bonds, such that these data represent a nonlayered structure and agree well with the minimum conductivity predictions from the CP model (dashed line) and Eq. (4) (solid line). The layered films have interlayer forces that are different than the intralayer forces, which give rise to further scattering and a reduction in the thermal conductivity. The layered model (Eq. (14) shown by the dotted line), which takes into account this scattering mechanism, agrees well with the data and accurately predicts the correct thermal conductivity reduction. For model calculations, the longitudinal and transverse phonon velocities were taken as 1,650 and 1,150 m s⁻¹, respectively, and the atomic density was 1.62×10^{28} m⁻³ [10].

CONCLUSIONS

In summary, the minimum thermal conductivity of amorphous or heavily disordered solids was investigated by considering the phase and group velocities separately as is necessitated by the implementation of Einstein’s original assumptions regarding the minimum thermal conductivity. Assuming that the phase velocity and group velocities are different, a new model for thermal properties of amorphous solids is derived that displays temperature trends similar to those reported for amorphous solids [15]. Through comparisons of the model derived in this work and the traditional CP model with conductivity data of *a*-SiO₂, it is apparent that both the hopping of localized modes as well as the propagation of nonlocalized modes must be considered in the thermal transport of disordered solids. The further reduction of thermal conductivity in layered structures is then accounted for by considering the scattering rate by an atom with a different binding force in the minimum conductivity model. The predictions of the minimum conductivity of a layered structure predict the correct drop in thermal conductivity of WSe₂ layered films and agree well with the reported values. The minimum thermal conductivity layered model provides a lower limit to phonon thermal conductivity of superlattice structures, which has major implications for thermoelectric material design.

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